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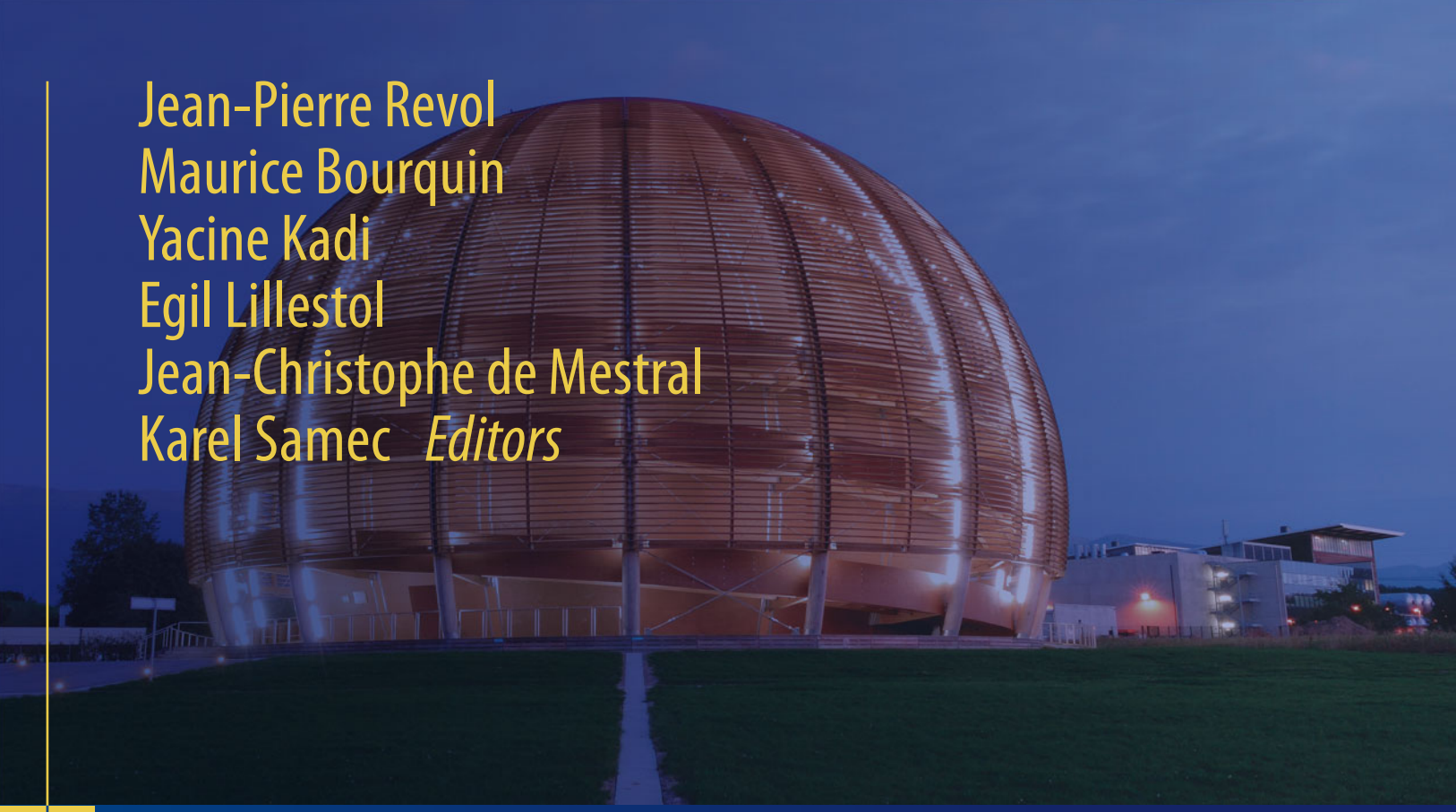
Maurice Bourquin
Yacine Kadi
Egil Lillestol
Jean-Christophe de Mestral
Karel Samec *Editors*



Thorium Energy for the World

Proceedings of the ThEC13 Conference, CERN, Globe of Science and Innovation, Geneva, Switzerland, October 27–31, 2013

iThEC international Thorium Energy Committee www.ithec.org

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Editors

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Conference Poster

Foreword

A large amount of experience has been gained during the last 60 years of construction and operation of nuclear reactors based on uranium. It is now desirable to use this experience to explore and develop alternatives to present-day technologies.

Following the Fukushima accident, fear of radiation has led to snap decisions to phase out nuclear power in Germany. However, there is still room for long-term thinking and financing of innovation in the nuclear sector. The work on thorium-based nuclear reactors is attracting increasing attention around the world, and rightly so: Global thorium resources are assessed to be three to four times greater than those of uranium, and the thorium cycle gives rise to waste that is smaller in volume, less toxic, and less long-lived than waste from uranium fuel.

From a security point of view, two other features are highly appealing: The first is that the use of thorium fuel offers a way to burn up large stocks of plutonium that now sit idle, but require expensive care and protection. The other is that thorium fuel does not give rise to materials that can be used for bombs.

The civilian nuclear community must do what it can to reduce the risk that more nuclear weapons are made from uranium or plutonium. This community can and should use its considerable brainpower to design reactors that can be easily safeguarded along with fuel and fuel supply organizations that do not lend themselves to proliferation. In these regards, the thorium community may have important contributions to make.

Hans Blix

Acknowledgments

The iThEC is grateful to the IThEO for having been granted the organization of the ThEC13, the fourth event in its series of Thorium Energy Conferences.

The ThEC13 organizers would like to express their gratitude to CERN for hosting the conference in the Globe of Science and Innovation, thus adding an inestimable symbolic value to the conference, and for providing additional infrastructure and personnel, which contributed to a stimulating and pleasant working atmosphere.

Our thanks also go to the International Scientific Advisory Committee for helping to shape the program and for proposing world-leading experts as speakers and authors of posters.

We are indebted to our sponsors, who made it possible to select and invite appropriate speakers, some of them coming from far away, thus allowing a global and thorough overview of current thorium nuclear power activities in the world, and of plans for future developments.

To the speakers themselves, we must express our appreciation and gratitude for their excellent presentations and for making the additional effort to provide written contributions for the conference proceedings. Credit also goes to the authors of the posters, who contributed to the many interesting and exciting discussions, in particular during the coffee breaks, and also for their contributions to the proceedings.

Finally we would like to warmly thank all the participants, who made this conference a success by contributing to the varied discussions and by creating the pleasant atmosphere that pervaded the scientific events and the various social activities.

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IBeL, Innovation for a Better Life
iThEC, international Thorium Energy Committee
IThEO, International Thorium Energy Organization
PSI, Paul Scherrer Institut
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Perspective Politique

Le futur énergétique de la planète est un enjeu politique et scientifique. Les deux sont inséparables. Or une partie de l'Europe, pour des raisons politiques, a décidé de renoncer, à terme, à toute forme de production d'énergie à partir du nucléaire. Le résultat immédiat est un envol de la production d'électricité à base de charbon et une dégradation du bilan environnemental consécutif au dégagement de CO_2. Mais surtout cette décision ignore les chances qu'offrent d'autres technologies que celle de la filière nucléaire classique.

Le thorium en particulier est prometteur comme combustible nucléaire. Il permet d'échapper à la plupart des désavantages de la technologie nucléaire fondée sur l'uranium enrichi. Son utilisation dans un réacteur sous-critique rend les risques d'emballement de la réaction nucléaire inexistants. Quant aux déchets, ils sont moins importants et leur dangerosité est très réduite. De plus, les déchets de longue durée provenant des centrales actuelles pourraient être récupérés et intégrés au cycle du thorium pour être transformés en matière stable tout en générant de l'électricité. Il est vrai cependant que le recours au thorium exige encore un effort de recherche mais les chances de succès sont établies.

Dans ces conditions, la frilosité des autorités politiques en charge de l'avenir énergétique est une énigme. Elle est explicable par deux facteurs, l'inquiétude face aux lobbys qui se refusent à distinguer entre les différentes formes d'énergie nucléaire et … l'ignorance des faits scientifiques.

Le symposium ThEC13 tenu au CERN et consacré à l'état d'avancement des travaux sur le thorium répond au besoin d'informations scientifiques destinées à un large public, y compris aux décideurs politiques.

Espérons que la publication des actes de ce symposium contribue à provoquer un débat plus ouvert et rationnel sur l'avenir de la production énergétique et sur les avantages et désavantages des différentes technologies nucléaires.

Pascal Couchepin
Ancien Président de la Confédération suisse

Political Perspective (English Translation)

The energy future of our planet is a political and a scientific issue. The two are inseparable. However, part of Europe, for political reasons, decided to give up for the near future all forms of energy production from nuclear power. The immediate result is a drastic increase of electricity production from coal and a subsequent degradation of the environment due to CO_2 emissions. But mostly, this decision ignores the opportunities offered by technologies other than the conventional nuclear industry.

Thorium, in particular, is promising as a nuclear fuel. It allows most of the disadvantages of conventional nuclear technology based on enriched uranium to be avoided. When used in a subcritical system, the risk of runaway nuclear reactions is nonexistent. As for nuclear waste, it is produced in smaller quantities and its dangerousness is much reduced. Moreover, long-lived waste from present plants could be retrieved and integrated into the thorium fuel cycle to be transformed into a stable material while generating electricity. It is true, however, that the use of thorium still requires much research effort, but the chances of success are established.

Under these conditions, the reluctance of the political authorities in charge of the energy future is a mystery. It can be explained by two factors: concern triggered by the lobbyists who refuse to distinguish between different forms of nuclear energy and … ignorance of scientific facts.

The ThEC13 symposium held at CERN and dedicated to the progress in developments related to the use of thorium addresses the need for scientific information to be provided to a wide audience, including policy makers.

Let us hope that the publication of the proceedings of this symposium will help bring a more open and rational debate on the future of energy production and the advantages and disadvantages of various nuclear technologies.

Pascal Couchepin
Former President of the Swiss Confederation

Conference Photos

Pascal Couchepin, former president of the Swiss Confederation (*left*), in discussion with Carlo Rubbia, Nobel Prize laureate (*right*), during the ThEC13 reception at the Hotel President Wilson, in Geneva; Jean-Pascal Stancu of iThEC (*center*)

Standing in front of the CERN Globe of Science and Innovation, from *left* to *right*, Hongjie Xu, Carlo Rubbia, Claude Haegi, Jean-Christophe de Mestral, Hans Blix, Pascal Couchepin, Jean-Pierre Revol, Egil Lillestol, Anil Kakodkar

From *left* to *right*, Claude Haegi, former mayor of Geneva, Jean-Christophe de Mestral, author of the book «L'Atome Vert», Hans Blix, former director general of IAEA

Rolf Heuer, director general of CERN, and Pascal Couchepin, former president of the Swiss Confederation, sharing a drink at the ThEC13 banquet, at the Hotel President Wilson, in Geneva

On the occasion of the lunch hosted by the president of iThEC at the CERN restaurant: from *left* to *right* Egil Lillestol, Jean-Christophe de Mestral, Jean-Pierre Revol, Hongjie Xu, Pascal Couchepin, Carlo Rubbia, Claude Haegi, Hans Blix, and Anil Kakodkar

Photo taken the first day of the conference, inside the CERN Globe of Science and Innovation. One may recognize among many others, Carlo Rubbia, Anil Kakodkar, Claude Haegi, Rafael Gimalov, Jack Steinberger, Maurice Bourquin, Hamid Ait Abderrahim, Egil Lillestol, Jean-Christophe de Mestral, Claude Gelès, Jean-Pascal Stancu, Claude Joseph, Jean-Pierre Budliger, Jean Borer, Jean-Pierre Revol, etc.

Photo taken the second day of the conference, inside the Globe of Science and Innovation. One may recognize among many others, Andreas Norlin, Hongjie Xu, Claude Joseph, Maurice Bourquin, Oscar Barbalat, Anil Kakodkar, Jean-Pascal Stancu, Hamid Ait Abderrahim, Egil Lillestol, Hans Blix, Pascal Couchepin, Carlo Rubbia, Rolf Heuer, Claude Gelès, etc.

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Contributions: Invited Speakers

Opening Talks

Welcome, Jean-Pierre Revol, president of iThEC
Un Plaidoyer pour l'Innovation, Claude Haegi, Member of the international Thorium Energy Committee (iThEC), President of the Foundation for the Economy and sustainable Development of the Regions of Europe (FEDRE), Former President of the Government of the Canton of Geneva
A Plea for Innovation (English version of Claude Haegi's Presentation)
A Future for Thorium Power? Carlo Rubbia, Nobel Prize Laureate, Senator for Life in the Italian Senate, IASS, Potsdam, Germany, GSSI, L'Aquila, Italy

National and International Thorium Programmes

Towards Sustainable, Secure and Safe Energy Future: Leveraging Opportunities with Thorium, Anil Kakodkar, Former Chairman of the Atomic Energy Commission of India and Secretary to the Government of India
Thorium Energy and Molten Salt Reactor R&D in China, Hongjie Xu, SINAP, China, Head of China MSR Project
The Japanese Thorium Programme, Toshinobu Sasa, JAEA, Japan
Thorium Fuel Cycle Activities in IAEA, Uddharan Basak, IAEA
Overview of European Experience with Thorium Fuels, Didier Haas, Belgium
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Part I
Invited Speakers: Opening Talks

Welcome

Jean-Pierre Revol

Good morning, Ladies and Gentlemen. I would like to welcome you to ThEC13, the international conference dedicated to the use of thorium as a source of energy.

Nuclear energy has the potential to provide humankind with abundant carbon free energy for at least a thousand years. The question we shall be addressing with ThEC13 is: "Can nuclear energy based on thorium fuel fulfill that potential?". The more specific goal of ThEC13 is to contribute to answering that question and show that nuclear energy based on thorium fuel would be acceptable to society, in terms of safety, management of waste and weapon proliferation.

The Organizing Committee is very pleased that, with more than 200 participants from 32 countries, all continents are represented at ThEC13. This is clear testimony to the strong and growing worldwide interest in thorium.

As this conference is entirely dedicated to thorium, I thought that it would be a good idea to have some thorium in the room. There is a sample of 230 g of Monazite (Fig. 1) on display at the entrance of this auditorium. It contains 2–3 % of thorium and was extracted from the Steenkampskraal mine in South Africa. It comes to us courtesy of Trevor Blench, CEO of Steenkampskraal Thorium Limited, whom I would like to specially thank for his gift to ThEC13.

The sample is slightly radioactive. The dose rate at 50 cm is 0.2 μSv/h, to be compared with the natural radioactivity dose rate in Geneva of around 0.1 μSv/h.

Fig. 1 Photo of the Monazite sample that was on display in the CERN Globe of Science and Innovation on the occasion of ThEC13

As we have a rather tight schedule ahead of us for the next three and a half days, let me conclude now, and on behalf of iThEC, the international Thorium Energy Committee, and IThEO, the International Thorium Energy Organisation, wish you a very fruitful conference, here in Geneva, on the use of thorium in the energy domain.

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Un Plaidoyer Pour L'innovation

Claude Haegi

Abstract

Le fait que cette conférence se tienne au CERN symbolise l'importance que le CERN attache à l'innovation.

Mesdames et Messieurs,

Bonjour, et bienvenue à Genève.

Merci Monsieur le Directeur Général du CERN d'accueillir cette importante conférence dans le Globe de la Science et de l'Innovation. Ce globe, offert par la Confédération Suisse, est un signe de reconnaissance à la communauté scientifique internationale, qui est rassemblée ici sur la frontière franco-suisse et dont les résultats des travaux rayonnent à travers le monde. Ce globe est un symbole universel de la recherche scientifique de pointe et du développement durable, par sa structure en bois et son architecture novatrice.

Le CERN a été un projet scientifique prestigieux, rassembleur européen, s'inscrivant comme un acte fort de la réconciliation de l'après-guerre. Ce projet a été porté par le physicien français Louis de Broglie lors de la Conférence Européenne de la Culture, à Lausanne fin 1949, conférence organisée par l'un des pères de la reconstruction européenne, le Suisse Denis de Rougemont. Le Conseiller d'Etat, Ministre genevois, Albert Picot et le physicien suisse Paul Scherrer, totalement acquis à l'idée de créer ce grand laboratoire, obtinrent en 1952 à Amsterdam que le CERN s'installa à Genève. Le CERN fait partie de l'identité genevoise, il en est même l'un des principaux fleurons. Son installation passa sous les fourches caudines de la démocratie directe cantonale helvétique. En 1953 par la voie d'un référendum, 70 % des électeurs du canton ont soutenu l'arrivée du CERN, l'Organisation Européenne pour la Recherche Nucléaire.

Les citoyennes et citoyens ont donc façonné une Genève de la recherche et témoigné de la confiance qu'ils accordaient au génie humain. Dans ce domaine la population genevoise a encore manifesté un énorme intérêt en 1955 et 1958 lors des deux premières Conférences internationales de l'Energie atomique des Nations Unies, «l'Atome pour la Paix». Lors de ces événements, Genève devenait la capitale mondiale de l'énergie atomique pour ses applications pacifiques et le potentiel, notamment médical et industriel qu'elle détenait.

C'est important de se le rappeler face aux velléités de replis et aux freins auxquels la recherche et le développement sont exposés dans ce domaine. Les remises en cause du maintien de certaines centrales nucléaires sont parfaitement légitimes de par leur dangerosité, tout comme de nouvelles exigences de sécurité, mais elles ne sauraient compromettre la recherche pour trouver de nouvelles filières ou maîtriser d'actuels problèmes sécuritaires.

L'industrie nucléaire actuelle contient des risques réels, créés en général par des négligences, de l'inconscience, voire par de coupables recherches d'économies. Ces situations ne justifient pas de jeter le bébé avec l'eau du bain, ce n'est pas la science qui est responsable des divers dramatiques événements nucléaires que nous avons connus et qui ont modifié négativement la perception que l'on a du nucléaire, mais les formes d'irresponsabilité décrites. La science progresse et dans le temps sait répondre à nombre de nos besoins.

Le renommé physicien du CERN, Georges Charpak, Prix Nobel, incarna bien cette confiance en l'innovation conduite avec un discernement humain pour répondre aux besoins énergétiques de tous les habitants de la planète, en préservant la biosphère et l'ensemble de l'environnement naturel. Georges Charpak, homme de justice, donnait une réponse scientifique pour permettre aux habitants des zones en développement d'améliorer leur niveau de vie.

Ancien Président du Gouvernement de La République et Canton de Genève.

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C'est dans cette dynamique que Carlo Rubbia, ancien Directeur Général du CERN et Prix Nobel de Physique, travailla avec son équipe le moyen de disposer d'une énergie sûre et propre en utilisant du thorium plutôt que de l'uranium. Les travaux accomplis par Carlo Rubbia et ses collègues, ont été considérables, mais les relais pour tenter de mener cette recherche à une application pratique, furent régulièrement compromis pour des raisons politiques et économiques. La prise de conscience, de plus en plus large, des futurs besoins énergétiques, devrait faire comprendre que l'on ne fait pas l'économie d'un projet aussi prometteur, qui certes ne s'inscrit pas dans l'immédiateté, mais à plus longue échéance.

Compte tenu de ses avantages: Grandes et accessibles réserves de thorium, sûreté des installations, absence d'usage militaire et, argument majeur, destruction des déchets; il serait irresponsable de ne pas approfondir une telle filière qui représenterait une révolution.

Cette démarche doit s'inscrire dans une politique énergétique globale et visionnaire, partant de la recherche fondamentale pour cheminer vers la recherche appliquée puis si possible l'industrialisation.

Mais une politique énergétique globale et visionnaire ne va pas de soi. Quoi qu'elle prétende, l'Europe y compris la Suisse, n'en n'a pas et les patrons des plus grandes sociétés énergétiques l'on dit récemment à Bruxelles. En ce moment, l'Europe se fragilise entre les USA devenant énergétiquement autonomes et l'Asie innovante et industrielle. L'Europe prend de grands risques de dépendance.

En fait la réponse énergétique doit être internationale et même mondiale. D'un approvisionnement sûr dans des conditions tarifaires stables, dépend l'équilibre des régions développées ou aspirant logiquement à le devenir, grâce à une organisation économique générant notamment des emplois et freinant l'émigration. La qualité des relations Nord Sud en dépend. La recherche fondamentale est certes onéreuse et les retours sur investissement s'inscrivent dans le long terme. D'où la nécessité de disposer de financements partagés.

On dit parfois que la piste du thorium mobiliserait trop de fonds et pénaliserait la progression des énergies renouvelables. Tel n'est pas le but. Il faut trouver des solutions de financement adaptées, et le développement du thorium doit mobiliser des fonds diversifiés venant notamment des plus grands consommateurs mondiaux. La présence à cette conférence de représentants notamment de la Chine, de l'Inde et du Japon, montre l'intérêt que ces grands pays, de la dynamique zone asiatique, portent à la filière du thorium.

Une politique énergétique visionnaire n'écarte pas les actions concrètes immédiates. A court terme on peut obtenir des résultats significatifs en matière d'efficience énergétique en consommant moins, sans renoncer au confort tout en dynamisant les activités économiques.

L'efficience des énergies renouvelables peut aussi être considérablement améliorée par une approche systémique interdépendante, en ne se limitant pas à des applications par filières. C'est la Smart Grid, qui permet de jouer sur la complémentarité des sources renouvelables telles que le vent, le soleil, la géothermie, la biomasse ou l'hydroélectricité afin d'assurer une production continue. Certains pays savent déjà la développer.

On saura aussi, si on le veut, maitriser les pollutions encore générées par des énergies fossiles, notamment le gaz dont on n'a encore bien besoin.

Le maitre mot est d'innover grâce à la recherche.

La coopération et les participations scientifiques devenues mondiales au CERN, confirment sa vocation d'initier de véritables projets de société. Les travaux réalisés au CERN sur le thorium doivent être maintenant repris par des acteurs internationaux, capables de franchir les étapes suivantes. Pour porter ce grand projet des forces scientifiques, industrielles et politiques devront être fédérées dans un nouveau réseau. Souhaitons à cette conférence d'être un moment déterminant, pour faire avancer la voie du thorium permettant la destruction des déchets existants et futurs, et déboucher sur une innovation complétant le bouquet énergétique tout en préservant notre environnement.

A Plea for Innovation (English Version)

Claude Haegi

Abstract

This conference in the Globe of Science and Innovation is a clear sign of the high value that CERN attributes to innovation.

Ladies and Gentlemen,

Good morning and welcome to Geneva.

I would like to thank the Director-General of CERN for allowing us to hold this important meeting in the Globe of Science and Innovation. This Globe, a gift from the Swiss Confederation, is a sign of recognition to the international scientific community assembled here on the Franco-Swiss border, the results of whose work radiate throughout the world. This Globe is a universal symbol of leading-edge scientific research and sustainable development through its wooden structure and novel architecture.

CERN is a prestigious scientific project, a European rallying point, founded as a strong act of reconciliation after the Second World War. This project was supported by the French physicist Louis de Broglie at the European Conference on Culture in Lausanne at the end of 1949, organized by one of the founding fathers of European reconstruction, the Swiss Denis de Rougemont. The State Councillor and Genevan Minister, Albert Picot, and the Swiss physicist Paul Scherrer, totally convinced by the idea of creating this great laboratory, secured the establishment of CERN in Geneva at a meeting in Amsterdam in 1952. CERN is part of the identity of Geneva; it is even one of its main jewels. Its installation passed through the Caudine Forks of direct Swiss cantonal democracy. In 1953, through a referendum, 70 % of the electors of the canton supported the arrival of CERN, the European Organization for Nuclear Research.

Our citizens thus fashioned a Geneva of research and showed confidence in the human spirit. The population of Geneva also showed its great interest in 1955 and 1958 during the first two United Nations International Conferences on Atomic Energy "Atoms for Peace". During these meetings, Geneva was the world capital of atomic energy for peaceful purposes and its potential, most notably, for medical and industrial applications.

We must remember this in the face of attempts to turn back and slow down research and development in this field. Questioning of the maintaining of certain nuclear power stations is perfectly legitimate because of their danger, and of new security requirements, but research to find new outlets or to master current security problems should not be put into question.

The current nuclear industry does contain real risks, generally created by negligence, foolhardiness, or even a dangerous search for savings. This does not justify throwing the baby out with the bath water. It is not the science that is responsible for the different dramatic nuclear events that we have witnessed and that have negatively modified our perception of it, but the irresponsible methods described. Science progresses and with time knows how to respond to our needs.

The renowned CERN physicist Georges Charpak, Nobel Prize laureate, embodied this confidence in innovation carried out with human discernment to fulfil the energy needs of all the inhabitants of the planet, whilst at the same time preserving the biosphere and the whole of the natural environment. Georges Charpak, a man of justice, provided a scientific answer to allow the inhabitants of developing areas to improve their standard of living.

It was through this process that Carlo Rubbia, former Director-General of CERN and Nobel Prize laureate in physics, worked with his team to have a clean and safe energy by using thorium instead of uranium. The work done

Former President of the Government of the Republic and Canton of Geneva.

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by Carlo Rubbia and his colleagues was considerable, but their attempts to get their work to practical applications were regularly compromised for political and economic reasons. The realization of our future energy needs should make one understand that one should not dismiss such a promising project, which cannot be carried out in the short term, but should be considered in the long term.

Looking at the advantages: large and accessible thorium reserves, the safeness of installations, the absence of military usage and, a major argument, the destruction of nuclear waste; it would be irresponsible not to proceed further with such studies that represent a revolution.

This approach must become a global and visionary energy policy, starting with fundamental research, then onwards to applied research, and then, if possible, to becoming an industry.

Having a global and visionary energy policy is not so simple. Whatever they may pretend, the Europeans and the Swiss do not have such a policy, as the bosses of the largest energy companies pointed out recently in Brussels. At the moment, Europe is weakening, with the Americans becoming energy self-sufficient, and the Asians innovative and industrialized. Europe risks becoming dependent.

The answer to the energy question must be international and even global. Safe supplies and stable tariffs will give equilibrium to developed regions and those wishing to be so, thanks to an economic organization generating jobs and slowing down emigration. The quality of North–South relations depends on this. Fundamental research is expensive and the return on investment spans a long time period. For this reason the financing must be shared.

People say to me that the thorium track needs too much money and would penalize the progress of renewable forms of energy. That is not the idea. We must find properly adapted financial solutions, and the development of thorium must use diversified funds coming from the world's biggest consumers. The presence at this conference of representatives, in particular, from China, India, and Japan demonstrates the interest of these great and dynamic countries in the thorium solution.

A visionary energy policy does not remove the need for immediate concrete actions. In the short term, one can obtain significant energy saving results by consuming less, without giving up on comfort and at the same time creating economic activity.

The efficiency of renewable energy sources can also be considerably improved by an interdependent systemic approach, not limited to specific domains. This is the Smart Grid, which allows one to use the complementarity of renewable energy sources such as wind, sun, geothermal energy, biomass, or hydroelectricity in order to have a continuous supply. Certain countries have already developed this.

One can also, if one wishes, master the pollution generated by fossil fuels, especially gas, which one still needs.

The key is innovation thanks to research.

Cooperation and world scientific participation at CERN confirm its vocation to initiate truly societal projects. The work carried out at CERN on thorium must now be taken over by international actors capable of going on to the next steps. For this great project, scientific, industrial and political forces must be brought together in a new network.

Let us hope that this conference will be a decisive step in advancing the thorium channel, allowing for the destruction of existing and future waste and bringing about innovation to complete the range of energy sources, whilst preserving the environment.

A Future for Thorium Power?

Carlo Rubbia

Abstract

The expected duration of today's nuclear reactors, even if persisting at the present relatively low level of 5 % of the primary energy supply, is not appreciably longer than that for energy systems using natural gas and oil and is much less than that for those employing coal. There is no doubt that, for its continued usage, nuclear power must be profoundly modified. For instance, new breeding reactions based on tritium, natural uranium or thorium, which may last for many thousands of years, far beyond fossil fuels, must be pursued, together with much stricter safety levels and a deterministic safety approach. Amongst the breeding alternatives, the use of thorium represents a unique opportunity. The advantages of thorium burning are remarkable; especially if one considers that the same amount of electric energy may be produced from 3 million tons of coal, from U-235 extracted from about 200 tons of natural uranium, or from merely one ton of the vastly abundant natural thorium. Used in an accelerator-driven system, thorium opens options for a safe nuclear power, with a considerably simplified fuel cycle, significantly minimized production of long-lived nuclear waste, as well as the possibility of destroying existing nuclear waste and stockpiles of military plutonium. My own recommendation is to construct a full-scale industrial 600 MW_e subcritical thorium demonstrator, along the lines of the Energy Amplifier engineering design by Aker Solutions ASA, but using salt instead of metal, and at a cost far less than 10 % of what is presently invested in the ITER (International Thermonuclear Experimental Reactor) for fusion. A simplified fuel reprocessing concept may consist of considering the spent thorium, fission fragments, and minor actinides (Pa, Np, Am, and Cm) as "waste" and uranium and plutonium as "seeds". The duration of each fuel cycle would be about ten years and the reactor lifetime could exceed 200 years.

Introduction

The recent Fukushima accident, after the previous warning signs from Three Mile Island, brought to sudden rest the nuclear program of one of the most advanced and heavily nuclear–energy-dependent countries and created a strong movement against the continuation of nuclear power. This accident has proven the inadequacy of the present "probabilistic" safety concept overwhelmingly used by the nuclear community and has shown the necessity for an entirely new, alternative, "deterministic" approach. In order for nuclear power to be actively continued, it must be profoundly modified.

R&D on systems using new fuel breeding reactions based on tritium, natural thorium, or uranium must be pursued, albeit, with much stricter safety levels and a deterministic safety approach, as breeding may provide energy for many thousand years, far beyond the possibilities of oil and gas.

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For such new processes, a distinction between renewable and non-renewable energies is academic. In addition, the long-lived nuclear waste problem has to be solved.

Amongst the various breeding alternatives, the use of thorium represents a unique opportunity. This is a rather old idea, strongly supported by the fathers of nuclear energy, Alvin Weinberg, Eugene Wigner, and Edward Teller, but perhaps ultimately neglected because of its intrinsic absence of military spin-offs. Nowadays, the possibility of using thorium deserves considerable attention.

Today's Nuclear Energy

Commercial nuclear fission is currently based on the highly fissile uranium isotope, U-235, present at a level of 0.71 % in natural uranium. Nuclear reactors may operate either directly using natural uranium (CANada Deuterium Uranium, CANDU), or more often with the help of U-235 enrichment (pressurized water reactor, PWR), leaving behind depleted uranium tailings, still containing typically 0.25 % of U-235. In both alternatives, a substantial fraction of the U-235 remains unused and, in practice, less than one part in a few hundred of the natural uranium is actually burnt.

Burning is somewhat extended with the help of plutonium, more specifically the Pu-239 isotope produced from neutron capture by U-238, through the introduction of the so-called mixed oxide (MOX) fuel cycle, which uses plutonium extracted from spent fuel that is reprocessed and mixed again with (enriched) uranium.

These procedures reduce significantly—typically by 30 %—the consumption of uranium, but increase the amount of minor actinides in the spent fuel by about seven times and consequently the residual long-lived radiotoxicity of the waste.

New, Virtually Unlimited Forms of Nuclear Energy

Although the amount of exploitable ore has not been precisely determined and depends on the lowest concentration level at which uranium can be recovered, in the way it is used today and at the present levels of consumption (5 % of primary energy), uranium reserves are probably not larger than the reserves of oil and gas (Table 1).

Particularly interesting and so far largely unexploited are other fission reactions in which a natural element is first bred into a fissionable one:

Thorium Breeding Cycle

$$^{232}\mathrm{Th} + \mathrm{n} \rightarrow {}^{233}\mathrm{U}; \quad {}^{233}\mathrm{U} + \mathrm{n} \rightarrow \mathrm{fission} + 2.3\mathrm{n}$$

Uranium Breeding Cycle

$$^{238}\mathrm{U} + \mathrm{n} \rightarrow {}^{239}\mathrm{Pu}; \quad {}^{239}\mathrm{Pu} + \mathrm{n} \rightarrow \mathrm{fission} + 2.5\mathrm{n}$$

In each case, more than two neutrons are produced, which are used to initiate another capture on the natural element. Taking exploitable ores into account, these sources of energy represent a potential comparable to that of the deuterium–tritium (D–T) fusion reaction (to be used, for instance, in the International Thermonuclear Experimental Reactor (ITER) [1]).

Tritium Breeding Through Nuclear Fusion

$$\mathrm{Li} + \mathrm{n} \rightarrow \mathrm{T}; \quad \mathrm{T} + \mathrm{D} \rightarrow \mathrm{He} + \mathrm{n}$$

Table 1 Global energy resources at the end of the 20th century

Resource	Type	Yearly consumption in 1999 (ZJ)	Resources (ZJ)	Consumed up to 1999 (ZJ)
Oil	Conventional	0.13	12.08	4.85
	Unconventional	0.01	20.35	0.29
	Total oil	0.14	32.42	5.14
Natural gas	Conventional	0.08	16.56	2.35
	Unconventional	0.00	33.23	0.03
	Total gas	0.08	49.79	2.38
Coal	Total coal	0.09	199.67	5.99
Total fossils		0.31	281.88	13.51
Uranium	Thermal reactors	0.04	5.41 (2000, sw[a])	
	Breeder	0	324 (120,000, sw[a])	
Thorium			1,300,000	

[a]*sw* Resources including those contained in seawater; 1 ZJ (Zeta Joule) = 10^3 EJ (Exa Joule) = 10^{21} J (Joule)

In this case, neutrons produced in the D–T fusion reaction are used to breed new tritium, to replace the tritium consumed by fusion.

How Much Thorium Is Available?

Thorium, which exists naturally as a single isotope, Th-232, is an unexploited energy resource, about four times more abundant than uranium in the Earth's crust. While most of the uranium is dissolved in seawater, thorium is present as mineral deposits. The total abundance in the Earth's crust is estimated to be 1.2×10^{14} tons. Common soil typically contains an average of about six parts per million (ppm) of thorium [2].

Monazite black sand deposits contain 2–22 % thorium. Thorium can be extracted from granite and phosphate rock deposits, rare-earth elements (REE) or tin ores, and coal and uranium mine tailings.

Estimates of available thorium reserves vary widely. See, for instance, Table 1 for an estimate of known energy resources. According to the 2007 International Atomic Energy Agency and Nuclear Energy Agency (IAEA–NEA) report [3], there are 4.4×10^6 tons of known and estimated thorium reserves, but this still excludes data from much of the world.

With well-designed thorium burners, the total electricity production of China in 2007 (3.2 trillion kWh) could be produced with 443 ton/year of thorium by using Th-232/U-233 fuel, which represents only a few percent of the REE domestic production in that country.

Many Different Sources of Thorium

It has even been suggested that thorium could be extracted from the ashes of coal-fired power plants. A 1000 MW_e coal-fired power plant generates in its ashes about 13 tons of thorium per year. One ton of thorium can generate in turn electric power of 1000 MW_e for one year in a well-optimized thorium reactor. Thus, the ashes of one coal-fired power plant can conceptually fuel 13 thorium power plants of the same power.

The six parts per million of thorium contained in common soil have an energetic yield of about 3×10^6 times that of coal. If burnt in a reactor, thorium from the soil would generate $(6 \times 10^{-6}) \times (3 \times 10^6) = 18$ times the energy the soil turned entirely into coal would produce.

Recovering only one part per million of all the thorium in the Earth's crust would be equivalent to producing today's primary world power (15 TW, i.e., 6 kton/y of thorium) for about 20 thousand years.

Conclusion on the Use of Thorium

Thorium reserves represent an attractive potential energy supply for many millennia to come, with little or no CO_2 emissions. For instance, the whole electricity consumption of China could be produced for about 20,000 years by appropriate thorium reactors using 8.9 million tons of thorium: "Thorium is a source of energy essentially sustainable, on the human timescale".

Several conclusions may be drawn from the properties of U-233 (Table 2):

- U-233 is rather insensitive to neutron energy, both for α, the ratio of capture to fission cross sections and for η, the number of neutrons produced per neutron absorbed;
- U-233 is among the best fissile isotopes in the thermal neutron range;
- Th-232/U-233 systems have practical potential for breeding over the whole neutron energy spectrum (Fig. 1), where three separate neutron energy ranges have been considered: (a) thermal or epithermal neutrons (≈0.025–1 eV); (b) the neutron capture "resonance" region (10^3–10^4 eV); (c) fast neutrons (10^5–10^6 eV).

In contrast, U-238/Pu-239 breeding is only favorable with fast neutrons.

Table 2 Properties of the main fissionable nuclei for thermal and fast neutron energy spectra

Isotopes	^{235}U		^{239}Pu		^{233}U	
Spectrum	Thermal	Fast	Thermal	Fast	Thermal	Fast
σ_f (barn)	582	1.81	743	1.76	531	2.79
σ_c (barn)	101	0.52	270	0.46	46	0.33
$\alpha = \sigma_c/\sigma_f$	0.17	0.29	0.36	0.26	0.09	0.12
ν	2.42	2.43	2.87	2.94	2.49	2.53
$\eta = \nu\sigma_f/\sigma_a$	2.07	1.88	2.11	2.33	2.29	2.27
β_{eff} (pcm)	650		210		276	

σ_f, σ_c, and σ_a are the neutron fission, capture and absorption microscopic cross sections, respectively, β_{eff} is the fraction of delayed neutrons, expressed as parts per hundred thousand (pcm)

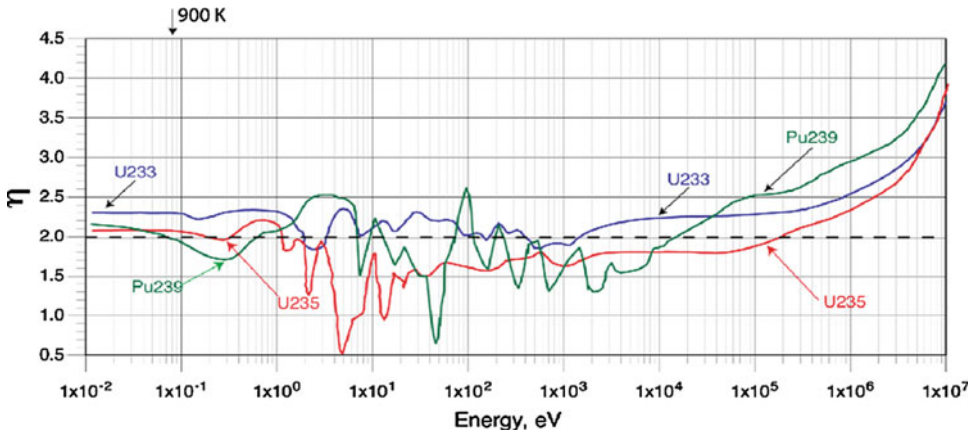

Fig. 1 Number of neutrons produced per neutron absorbed (η) in U-233, U-235, and Pu-239, as a function of neutron kinetic energy. For breeding to be possible, η has to be larger than 2

Thorium Breeders

Comparison of Uranium and Thorium Breeders

Breeding reactions, besides providing an almost unlimited source of energy, require major new developments for several reasons:

- Two neutrons are necessary to close the main breeding cycle, hence η must be larger than 2;
- Enrichment is no longer necessary, as breeding systems consume the entire natural material, either thorium or uranium;
- Breeding systems generate 200 times more energy than currently achieved in PWRs, in which only the U-235 isotope is used.

Early Attempts: Blending a U-235 Reactor with Thorium

In the early attempts, thorium was mostly considered as a secondary source of U-233. Fissile material containing U-235 was added repeatedly to the thorium fuel. One practical way of doing this consisted of adding thorium to a U-235 reactor in the form of a "blanket" surrounding the core in order to breed U-233.

Several newer alternatives to such a scenario have been considered:

- The Radkowsky reactor [4], in which uranium "seeds" enriched to about 20 % in U-235 are kept separated from a surrounding thorium–uranium "blanket". U-235 produces the neutrons that sustain the chain reaction while slowly creating U-233 in the blanket;
- The CANDU heavy water reactor with a thorium blanket associated with a core enriched in U-235 or Pu-239. The technology is similar to the Radkowsky reactor.

As burnup progresses, the newly created U-233 picks up an increasingly greater share of the fission load. In practice, somewhere between 30 and 50 % of the energy comes from thorium through the production of U-233.

A Purely Thermal Thorium Breeder?

The pioneers of the idea of a thorium–uranium breeder were Alvin Weinberg, Ralph Moir, and Edward Teller. The first step, the Molten Salt Reactor Experiment (MSRE) [5], not yet a breeder, ran successfully until 1969 at the Oak Ridge National Laboratory (ORNL) in the USA. It was followed by the Molten Salt Breeder Reactor (MSBR) project [6] (Fig. 2, left), which was terminated in 1976, despite the success of the MSRE. The project was followed up, in particular, by the FUJI project in Japan [7] (Fig. 2, right)

The liquid fuel was a molten mixture of uranium and thorium salts—$LiF–BeF_2–ThF_4–UF_4$—which had to be continuously reprocessed by an adjacent on-line chemical plant. All the salt had to be reprocessed within a period of about ten days in order to ensure criticality. Reprocessing consisted of:

- Fluorine removing U-233 from the salt;
- A molten bismuth column separating Pa-233 from the salt before Pa-233 decays;
- A fluoride salt system that distils the salts;
- Thorium salts that must be separated from the fission fragments, which constitute the "waste".

The MSBR fuel processing flow sheet is illustrated in Fig. 3.

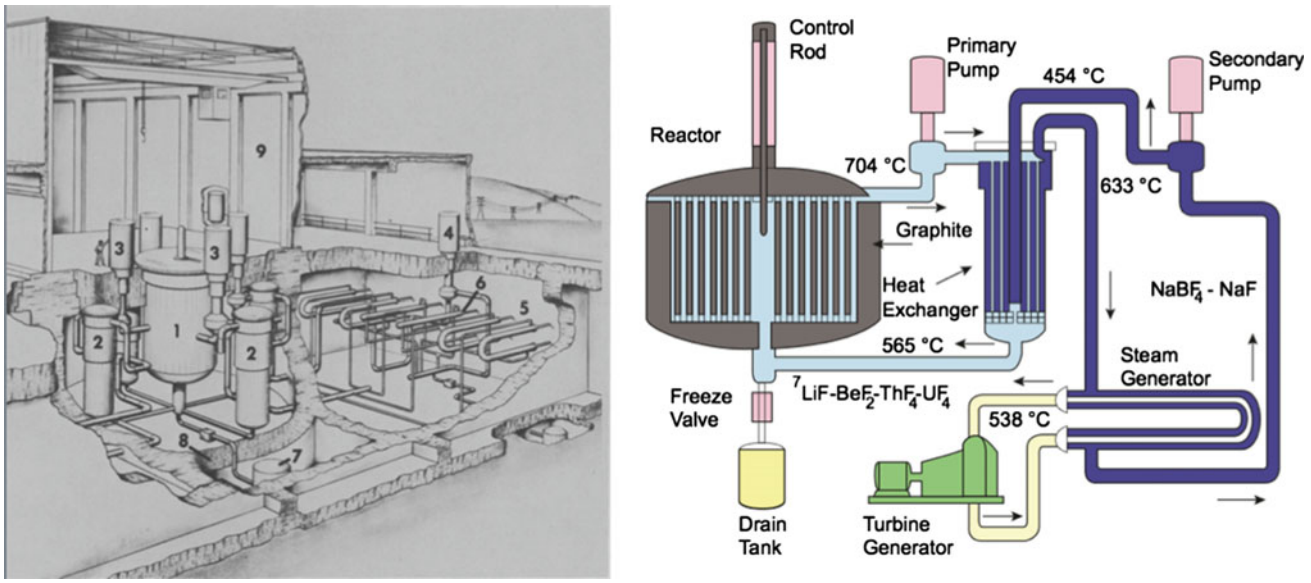

Fig. 2 *Left* Cut-away perspective of the MSBR reactor and steam cell, taken from [6]. *Right* Simplified schematic diagram of the FUJI reactor, taken from [7]

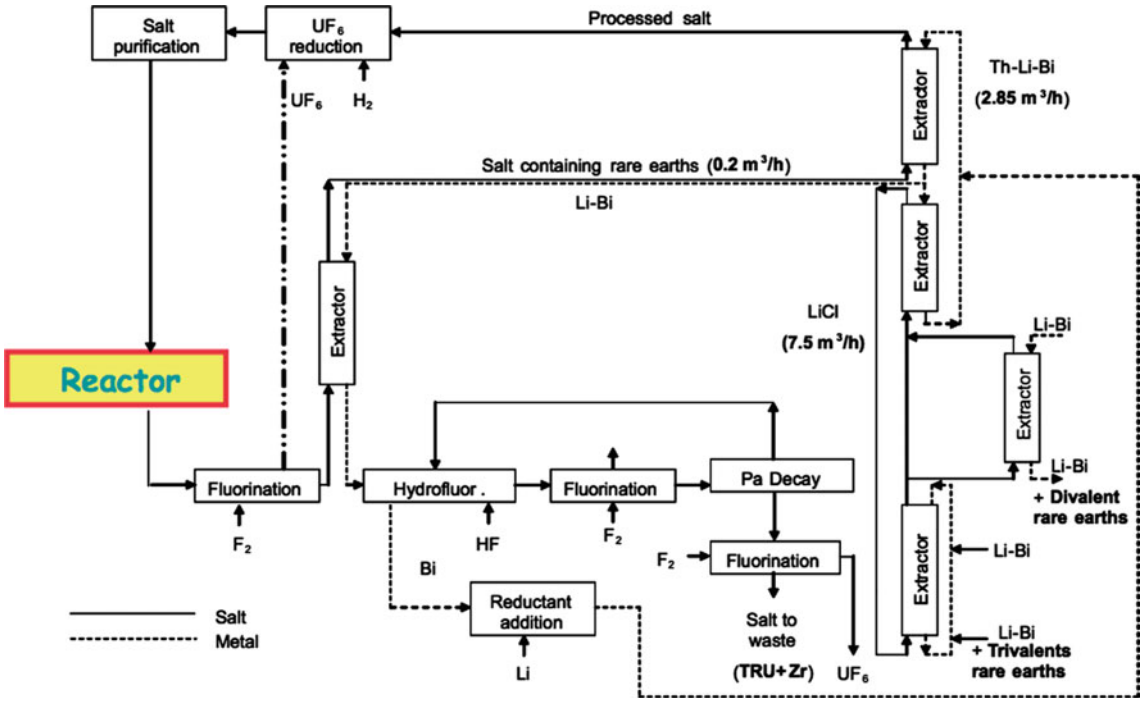

Fig. 3 MSBR fuel processing flow chart showing the extraction of fission fragments, protactinium, residual uranium, and Transuranic (TRU). Taken from [8]

Only one month before he passed away, Edward Teller wrote:

> The power plant could operate for up to 200 years with no transport of fissile material to the reactor or of wastes from the reactor during this period. Advantages that include utilization of an abundant fuel, inaccessibility of that fuel to terrorists or for diversion to weapons use, together with good economics and safety features such as an underground location will diminish public concerns. We call for the construction of a small prototype thorium-burning reactor.

Not all of his assumptions are actually correct, but the idea is tantalizing! The main advantage of thermal breeders is that the technology is well-known. In a thermal neutron flux, the

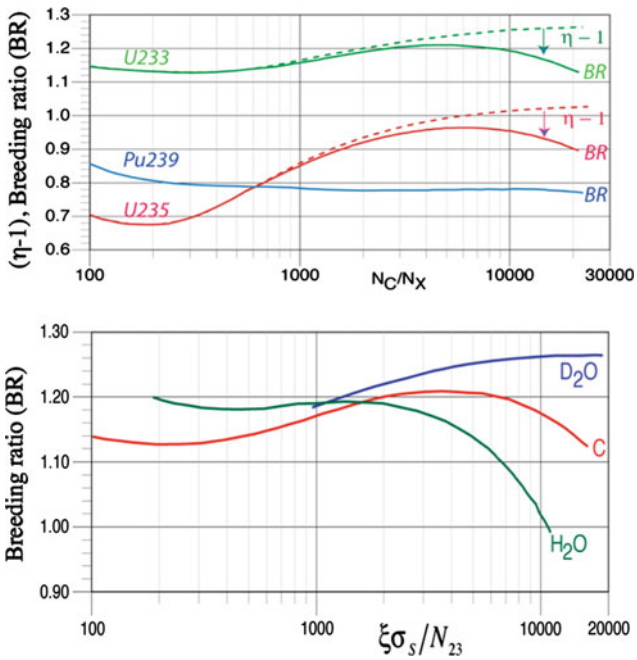

Fig. 4 *Top* Breeding ratios (*full lines*, labelled BR) and breeding potential ($\eta - 1$) (*dashed lines*) as a function of carbon concentration, evaluated at a temperature of 900 K for U-233, Pu-239, and U-235. *Bottom* Breeding ratios of U-233 for moderation with heavy water (D_2O), carbon, and light water (H_2O) as a function of the slowing down power per atom of fuel, where ξ is the average neutron lethargy, σ_s is the elastic cross section, and N_{23} is the number of U-233 atoms. Adapted from [9]

breeding potential, $(\eta - 1)$, is favorable for U-233 with respect to Pu-239 and U-235. This is illustrated in Fig. 4 for an infinite size system, neglecting the presence of fission fragments.

However, effects other than moderation reduce the breeding ratio, by amounts usually expressed as fractional neutron losses (ΔL). These effects include:

- Xenon-poisoning fraction, $\Delta L = 0.047$ at a power density $S = 3$ MW_{th}/kg;
- Losses due to captures on intermediate Pa-233, reducing the U-233 breeding process, $\Delta L = 0.1$ at a power density $S = 3$ MW_{th}/kg;
- A sizable ΔL due to fission product build-up;
- The build-up of the less readily fissionable uranium isotope, U-234.

Conclusion for Purely Thermal Thorium Operation

A thermal neutron reactor with H_2O, D_2O, or carbon coolant inevitably requires a delicate on-line fuel reprocessing system to operate satisfactorily. For example:

- In a critical reactor, without such chemistry, the small initial excess of k above criticality will be quickly wiped out by the accumulation of fission fragments, the presence of fuel cladding, and so on. In comparison, an ordinary CANDU reactor with natural uranium has $k_\infty(t = 0) = 1.47$, far larger than the k_∞ value for thorium breeding.
- If operated as an accelerator-driven subcritical system (ADS) with $k_0 < 1$, the active size of the fuel around the beam impact on the spallation target is proportional to the inverse of the neutron cross section times the factor $1/(1 - k)$. For thermal neutrons, the cross section is large; hence, the beam fission activity is narrowly concentrated (exponentially) around the location of the target. Therefore, in a thermal ADS, the beam-activated volume for the fission energy production for any reasonable value of k is strongly localized and not very uniform.

The thermal thorium configuration demands a complex, flawless, and risky on-line reprocessing of fuel in order to remove fission products and protactinium (Pa-233, prior to its decay to U-233), which both capture neutrons. This local reprocessing produces an additional strong radioactive background due to delayed neutron and gamma emissions generated outside the reactor core. It is unlikely that such a technology becomes widely accepted by society in view of the presently very highly critical public concerns.

The Thorium-Driven Fast-Neutron Energy Amplifier

The thorium-based fast-neutron energy amplifier (EA) is a subcritical system [9] (Fig. 5, left) driven by a proton accelerator. It uses fast neutrons and a closed fuel cycle based on natural thorium. In the closed fuel cycle, all actinides are recycled indefinitely. The "waste" consists of fission fragments and structural materials, the radiotoxicity of which is relatively short-lived.

Lead is used as a target for protons, as a neutron moderator, and as a heat carrier. The safety is based on a deterministic approach, relying on passive elements to eliminate criticality, meltdown, and decay heat. The system is equipped with seismic protection. In the conceptual design [9], the electrical power considered is 600 MW.

The fuel cycle is considerably simplified compared with molten salt critical reactors, or with the MOX cycle for uranium fuel in PWRs. The EA fuel cycle, as envisaged by Aker Solutions in the UK in 2010, who performed a detailed engineering study [11] for a system with 1500 MW of thermal power, 600 MW of electrical power, is shown in

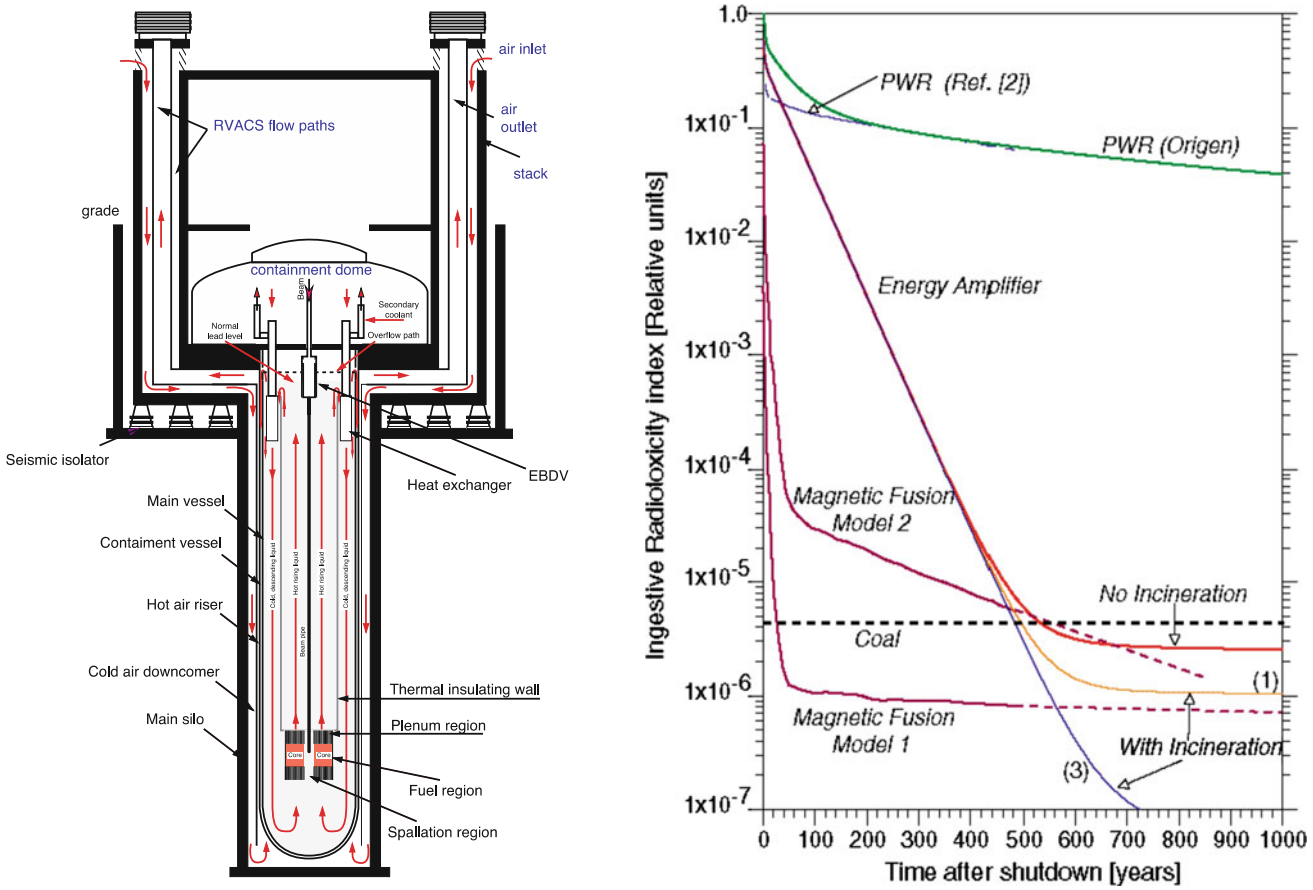

Fig. 5 *Left* Schematic diagram of the energy amplifier, taken from [9]. The proton beam is inserted vertically from the top in order to produce neutrons by spallation on the lead target at the center of the core. *Right* Relative time evolution of the ingestive radiotoxicity of nuclear waste produced by a PWR with an open fuel cycle, an energy amplifier under the three scenarios described in [9], by magnetic fusion under the two scenarios considered in [10], and coal ashes, corresponding to the same energy production

Fig. 6. It is a system with a subcritical core using thorium oxide fuel, driven by a proton beam introduced through a central beam tube, with molten lead as coolant at a temperature of 400 to 540 °C. It is equipped with two axial flow pumps and four annular heat exchangers for direct lead–water heat exchange. The design can be modified into a minor actinide ADS burner.

Comparing Alternatives

To continuously generate an electric power output of 1 GW for one year requires 3.5 million tons of coal, 200 tons of uranium in a PWR, or 1 ton of thorium in an EA. Coal burning has a significant impact on the environment through CO_2 emission and air pollution. Burning uranium in PWRs has a low CO_2 impact, but presents challenges with fuel reprocessing and requires very long-term storage of hazardous wastes. Enrichment is needed and this is prone to nuclear weapons proliferation. On the other hand, burning thorium in ADS systems has a low CO_2 impact, it can be used to eliminate plutonium and long-term radioactive waste, it reduces both the quantity and duration of storage needed for hazardous waste, it does not require enrichment, and is proliferation resistant.

Typical Operation of the Energy Amplifier

A particle accelerator supplies the missing neutron fraction and controls the energy produced by the fission reactions. For a proton beam energy of 1 GeV and the production of 1.5 GW of thermal power, the general characteristics of the system are:

- 2.7×10^{20} fission/s ($k_{\text{eff}} = 0.997$ and an energy gain, $G = 700$);
- 3.8×10^{17} spallation neutrons/s (30 neutrons per incident proton);
- A beam intensity of 1.3×10^{16} protons/s, or a current of 2.12 mA, corresponding to a beam power of 2.12 MW, feasible today with the state-of-the-art accelerator and spallation target technologies.

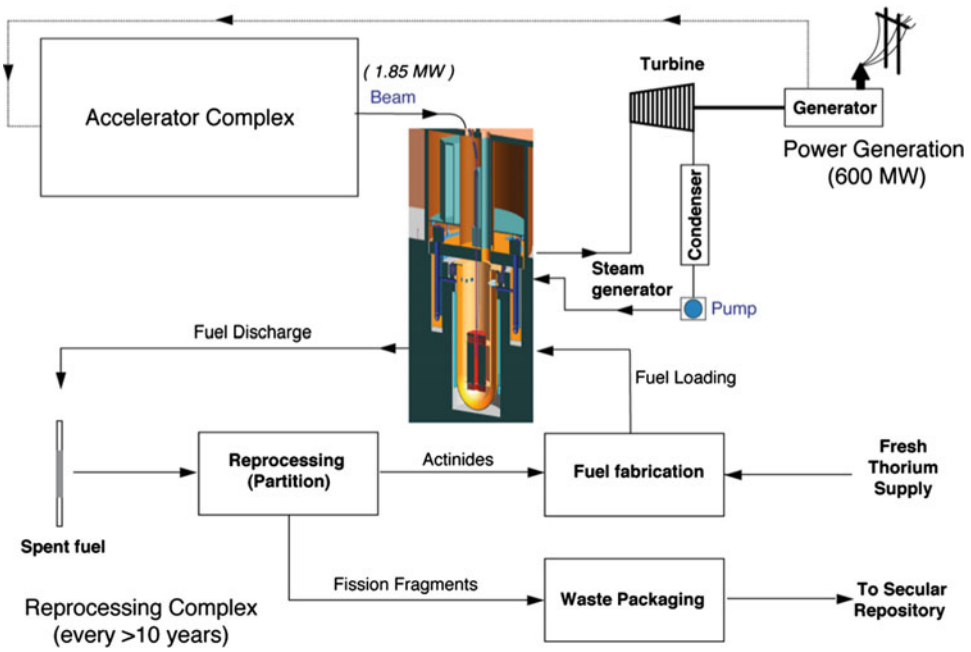

Fig. 6 The EA fuel cycle studied by Aker Solution in 2010. Taken from [11]

For a fast energy amplifier, the total neutron losses, ΔL, is 0.11, which is distributed as follows:

- Lead coolant: $\Delta L = 0.0626$
- Cladding: $\Delta L = 0.0378$
- Beam window: $\Delta L = 0.0004$
- Main vessel: $\Delta L = 0.0071$
- Leakage: $\Delta L = 0.0012$

Due to the accumulation of fission products, neutron losses grow linearly with increased burnup (Fig. 7) up to $\Delta L \approx 0.06$, including buckling changes due to the reduction of fuel mass.

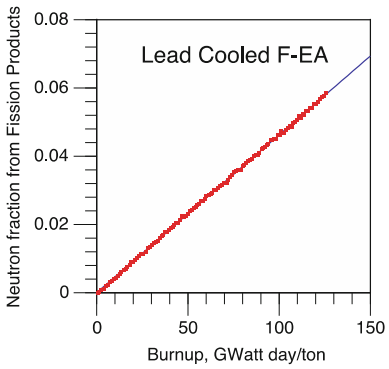

Fig. 7 Fraction of neutrons captured by fission fragments as a function of burnup in a fast energy amplifier

General Considerations on Thorium-Driven Fission Systems

A thorium-driven fission system has requirements very different from those envisaged for uranium-driven systems, both for thermal and fast-neutron energy spectra.

A subcritical or a critical thorium-driven breeding nuclear reactor can be considered as the indefinite repetition of very long-lasting and nearly identical breeding nuclear fuel cycles, progressively approaching an equilibrium concentration configuration of the fission-generating thorium–uranium mixture. This specific configuration, called "secular configuration", is characterized by an extreme fuel stability during the very long duration of the cycle and, in particular, an extremely constant spontaneous neutron multiplication factor, k.

Each cycle lasts between 10 and 15 years, a duration determined by the integrity of the fuel structural materials due to radiation damage (usually expressed in displacements per atom, dpa).

Basic Features of the Breeding Process

Let's assume for simplicity that the main iterative breeding process of a simple binary mixture, Th-232/U-233, is the one in which neutrons from U-233 fissions are transforming Th-232 into fresh U-233 in an indefinitely continuing process. Under steady neutron-flux conditions, the chain will

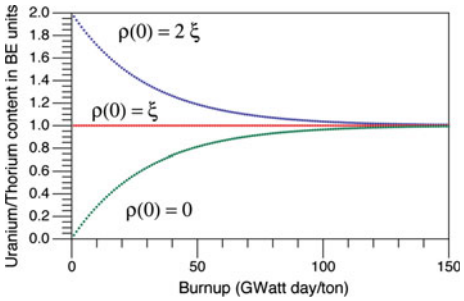

Fig. 8 Evolution of the relative U-233 to Th-232 concentration ratio, ρ, as a function of burnup, for three initial conditions: (a) starting at equilibrium, $\rho(0) = \xi$; (b) with an initial excess of U-233, $\rho(0) = 2\,\xi$; and (c) with an initially pure Th-232 fuel, $\rho(0) = 0$

tend toward equilibrium, namely a condition in which each fissioned nucleus is replaced by a newly bred fuel nucleus.

To a first approximation, the spectrum-averaged equilibrium concentration is given by:

$$\phi\langle\sigma_c^{232\mathrm{Th}}\rangle N(^{232}\mathrm{Th}) = \frac{N(^{233}\mathrm{Pa})}{\tau(^{233}\mathrm{Pa} \rightarrow {}^{233}\mathrm{U})} = \phi\langle\sigma_c^{233\mathrm{U}} + \sigma_\mathrm{f}^{233\mathrm{U}}\rangle N(^{233}\mathrm{U})$$

where ϕ is the neutron flux, σ_c is the neutron capture cross section, σ_f is the neutron fission cross section, N is the number of atoms of the element considered, and τ is the half-life of the element. $\langle\sigma\rangle$ indicates that the cross section was averaged over the neutron energy spectrum.

The breeding equilibrium turns out to be a simple function of averaged cross sections:

$$\xi = \frac{N(^{233}\mathrm{U})}{N(^{232}\mathrm{Th})} = \frac{\langle\sigma_c^{232\mathrm{Th}}\rangle}{\langle\sigma_c^{233\mathrm{U}} + \sigma_\mathrm{f}^{233\mathrm{U}}\rangle}$$

For thermal neutrons ξ = 0.0135 and for fast neutrons ξ = 0.125.

The Emergence of a Breeding Equilibrium

It is informative to study the ratio $\rho = N(^{233}\mathrm{U})/N(^{232}\mathrm{Th})$ as a function of burnup for three typical initial ^{233}U concentrations (Fig. 8). All configurations tend to progressively approach the breeding equilibrium, ξ.

Magic Equilibrium for Breeding

Assuming that the breeding equilibrium has been nearly reached at the end of the previous cycle, the addition of fresh thorium to the recovered uranium shifts the effective neutron multiplication factor, k, below the breeding equilibrium. Hence, k will increase. However, the build-up of neutron captures on the fission fragments will reduce the value of k. This "magic cancellation" (Fig. 9, left) [9] may be optimized, leading to a nearly constant value of k (Fig. 9, right) [9].

Plutonium Elimination and Fissile Uranium Build-Up

Over many fuel cycles, the simulation shows clearly the progressive disappearance of plutonium, and the build-up of U-233, until an equilibrium concentration is reached (Figs. 10 and 11). It is assumed that at the end of each cycle, all actinides are reprocessed and burnt Th-232 is replaced with fresh Th-232.

The fine adjustment of the appropriate value of k ($k < 1$) is obtained with the help of control bars.

Short Duration of Nuclear Waste and Fuel Reprocessing

The scenario envisaged is that of uninterrupted operation for about ten years, after which the only waste consists of fission fragments. Their radioactivity is intense, but limited to a few

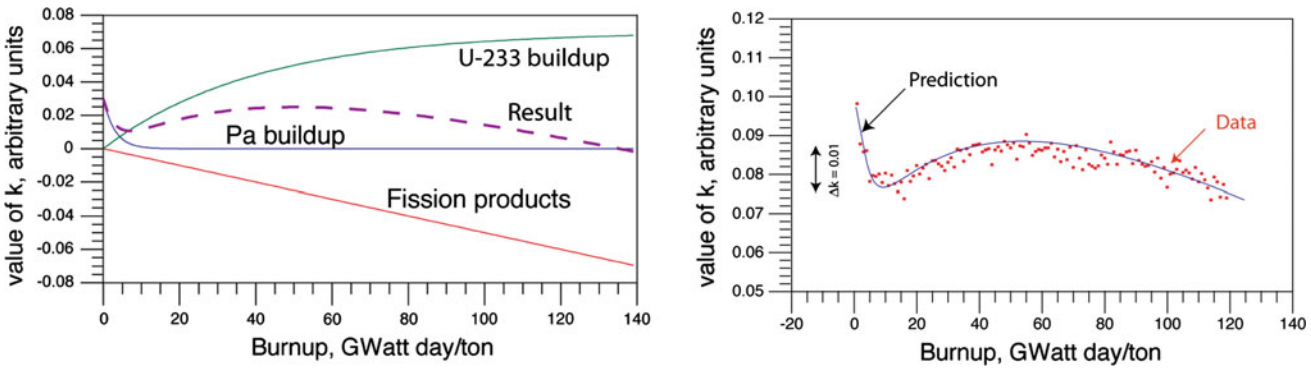

Fig. 9 Variation of the neutron multiplication factor, k, as a function of burnup. *Left* Contributions to k from fissions of U-233 and captures on fission fragments [9]. *Right* Monte Carlo simulation, compared with an analytic prediction [9]

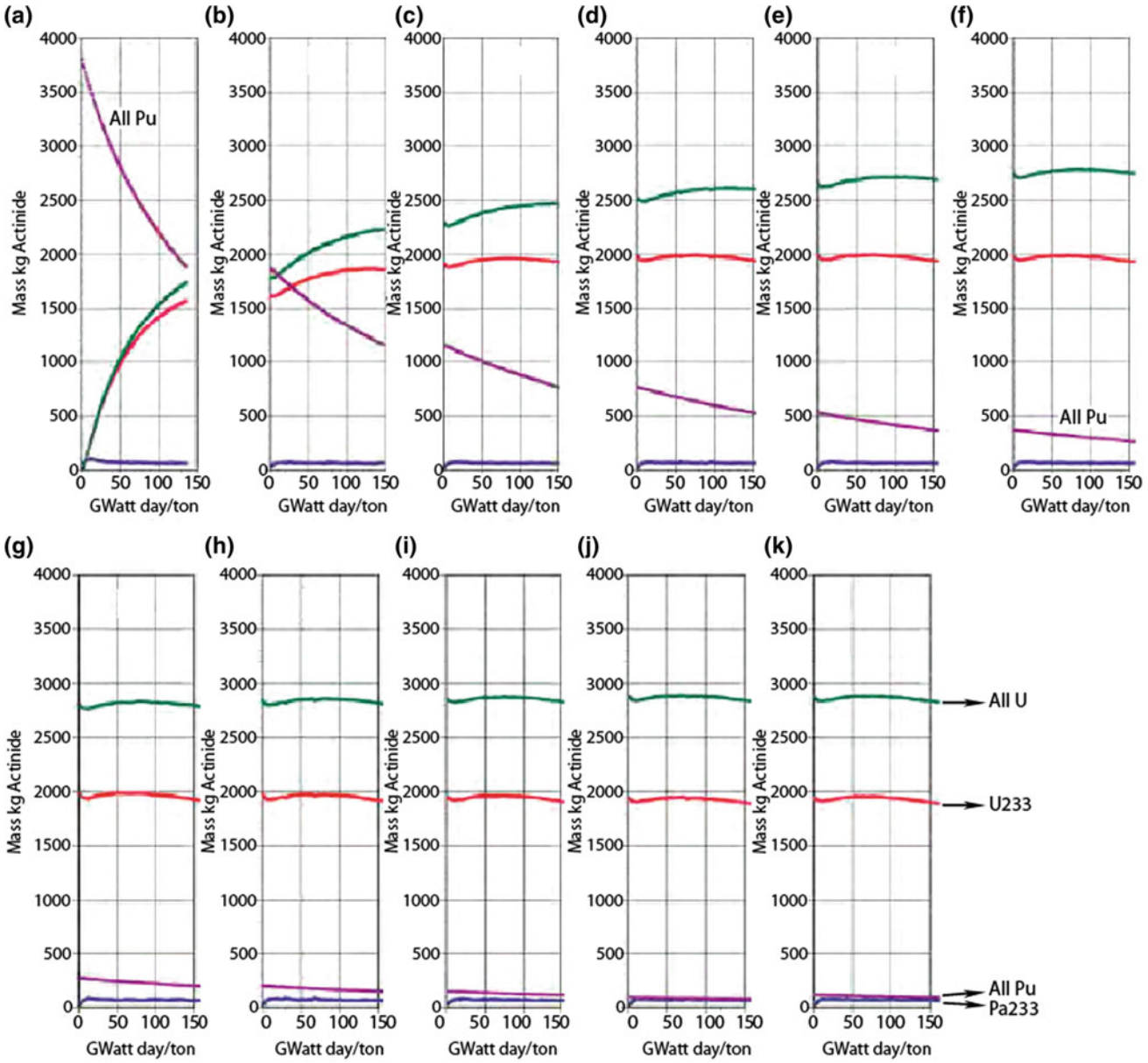

Fig. 10 Evolution of the metal content of the fuel (plutonium, U-233, all uranium isotopes, and Pa-233) as a function of the burnup in an energy amplifier over 11 cycles (labelled **a**–**k**), each extending to a burnup of 150 GW × day/t. The initial fuel is a MOX mixture with 84.5 % of ThO_2 and 15.5 % of PuO_2. Taken from [11]

hundred years (Fig. 5, right). This is to be compared to the one-million-year lifetime of the long-term waste in the present uranium fuel cycle. Actinides are recovered without separation and are the "seeds" of the next fuel load, after being topped with about 10–15 % of fresh breeding elements (Th-232 or U-238), in order to compensate for the consumption of these elements. A small fraction of actinides is not recovered and ends up with the "waste".

The fuel cycle is "closed" in the sense that the only material inflow is the natural elements and the only "outflow" corresponds to fission fragments.

Conclusion on Thorium Breeders

A pure thorium breeder may operate in a variety of configurations, namely epithermal, resonance, or fast neutrons, all with rather similar nuclear performances (i.e., similar values of η).

The basic reaction chain from Th-232 to Pa-233 to U-233 will converge to a "breeding equilibrium", namely to a steady-state condition in which each fissioned nucleus is replaced by a newly bred U-233 fuel nucleus.

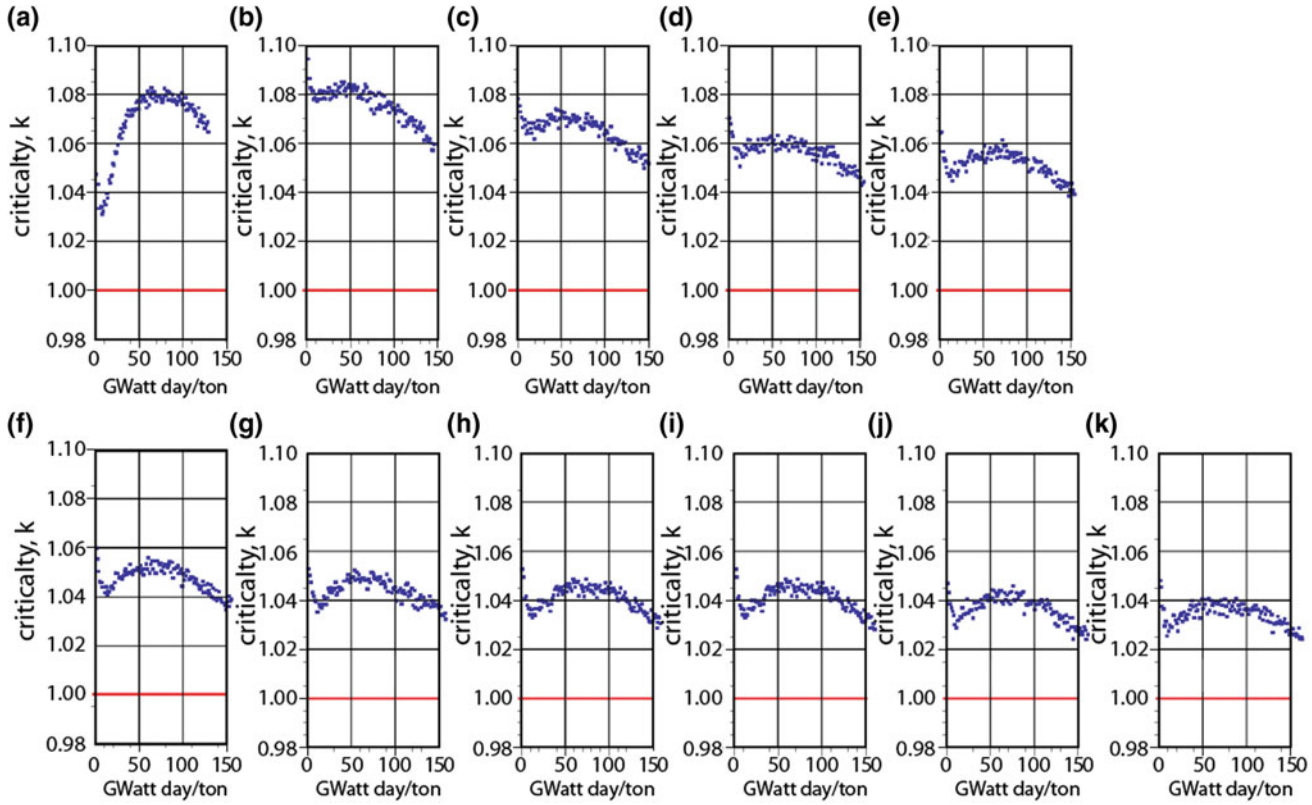

Fig. 11 For the same fuel cycles as illustrated in Fig. 10, the corresponding evolution as a function of burnup of the neutron multiplication factor, *k*. Taken from [11]

Fresh natural Th-232 supply is added periodically. Under ideal conditions, the cycle is closed, that is, Th-232 is converted into fission fragments.

Proliferation Issues

The breeding reaction on natural uranium is a problem for nuclear proliferation, as it involves the production of vast amounts of plutonium. Conversely, the breeding reaction on thorium is robustly immune from proliferation risks. The three main elements of the discharge, if chemically extracted —namely uranium, neptunium, and plutonium (Pu-238)— exclude the feasibility of an explosive device. See Table 3 for a comparison of the properties of the critical masses of each of the relevant elements with U-233. With one critical mass (28 kg) of the uranium mixture extracted at the end of a thorium fuel cycle, it would take only 10 min to get a lethal exposure at a distance of one metre, mainly because of the strong 2.6 MeV gamma radiation from Tl-208.

The extended presence of the fuel inside the energy amplifier (ten years) allows the system to be kept sealed under international control, avoiding illegal insertion of any other possible bomb-like materials.

Table 3 Properties of critical masses of bomb-grade plutonium, U-233, Np-237, and Pu-239

Element	Bomb-grade Pu-239	Uranium U-233	Neptunium[c] Np-237	Plutonium[c] Pu-238
Critical mass (CM) (kg)	3	28.0	56.5	10.4
Decay heat[a] for CM (W)	8	380	1.13	4400
Gamma activity (Ci/CM)	Negligible	1300	Small	Small
Neutron yield[b] (n/g/s)	66	3000	2.1×10^5	2600

Adapted from [9]
[a]Equilibrium temperature ~190 °C for 100 W, a concern due to the presence of high-power explosive shield
[b]Neutron yield must be ≤1000 n/g/s
[c]Very small amount produced at discharge

General Considerations About the Choice of Coolant

Comparing Pb, Pb/Bi Eutectic and Na Coolants

The radiotoxicity of the lead–bismuth eutectic (LBE) mixture is dominated by the Po-210 activity, resultant mainly from a direct (n, γ) capture on bismuth. The equilibrium amount for the EA1600 is 1.89 kg and 260 kW of decay power.

Pure lead still has a dominant activity due to Po-210, but with only 0.534 g, at equilibrium in the EA. Here, the early radiotoxicity of the Pb–Bi eutectic mixture is orders of magnitude larger than that of pure lead (Fig. 12).

Sodium has a simpler radiation spectrum that is characterized by Na-22 ((β^+, γ), with a γ-line at 1.27 MeV, half-life 2.6 years) and Na-24 (β^-, γ, with two γ-lines, one at 1.38 MeV and one at 2.75 MeV, half-life 14.6 h). The early radiotoxicity of sodium is substantially higher than that of pure lead. They become equal after about 2.5 days (Table 4).

Corrosion Studies

To optimize the efficiency of the Carnot cycle in the transformation of heat into electricity, it is desirable to operate energy systems at the highest possible temperature. One limitation is the corrosion of structural material by high

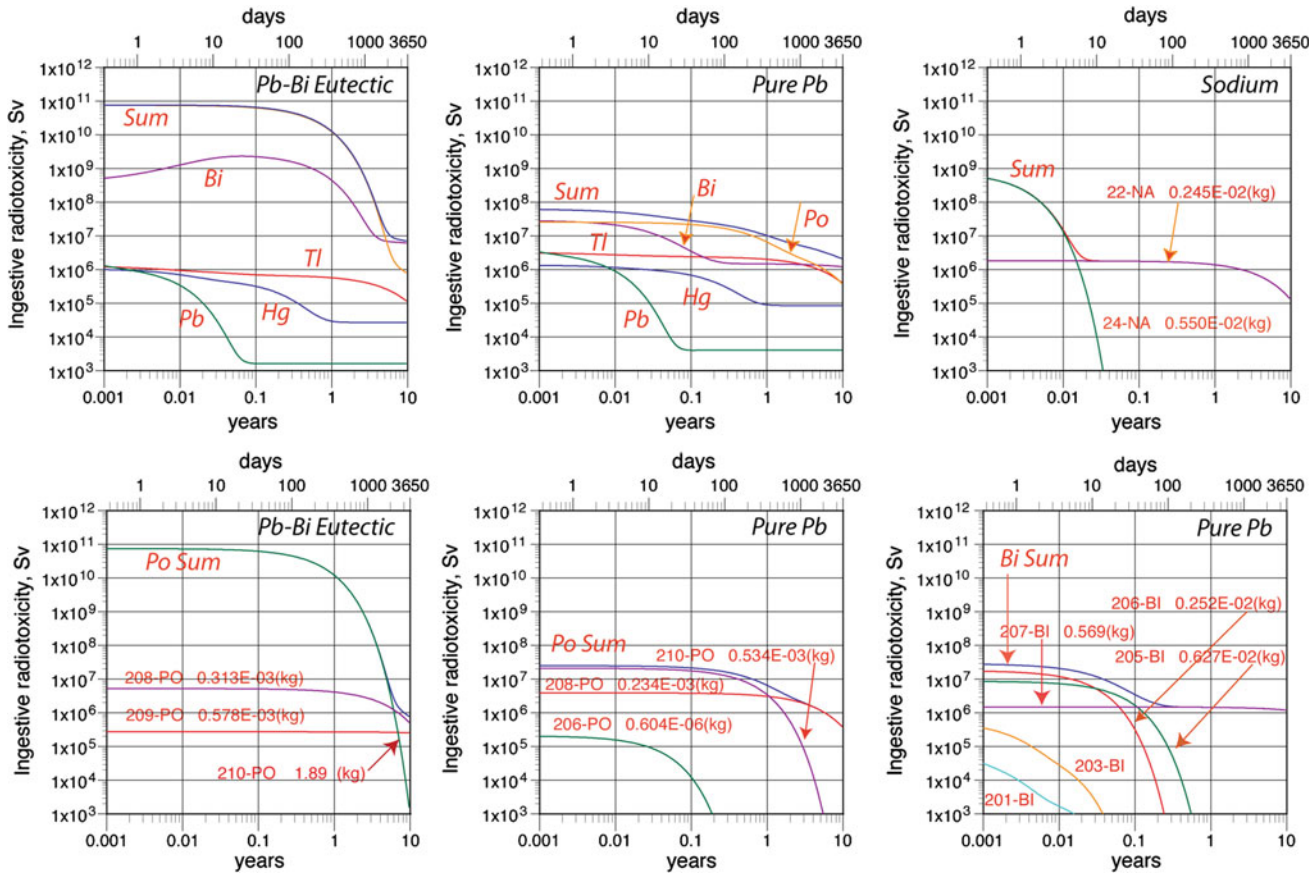

Fig. 12 Contributions to the ingestive radiotoxicity of Pb–Bi eutectic mixture, pure lead, and sodium. Taken from [11]

Table 4 Comparison of the ingestive radiotoxicities expressed in Sievert (Sv) of eutectic Pb–Bi mixture, lead, and sodium as a function of time after exposure to neutron irradiation, over a period of 0.365–100 days

After	0.365	1	3	10	30	100	Days
Pb–Bi	7.56×10^{10}	7.55×10^{10}	7.52×10^{10}	7.35×10^{10}	6.69×10^{10}	4.71×10^{10}	Sv
Pb	6.07×10^{7}	5.78×10^{7}	5.20×10^{7}	4.08×10^{7}	2.96×10^{7}	2.08×10^{7}	Sv
Na	5.11×10^{8}	2.52×10^{8}	2.81×10^{7}	1.81×10^{6}	1.77×10^{6}	1.68×10^{6}	Sv

Taken from [11]

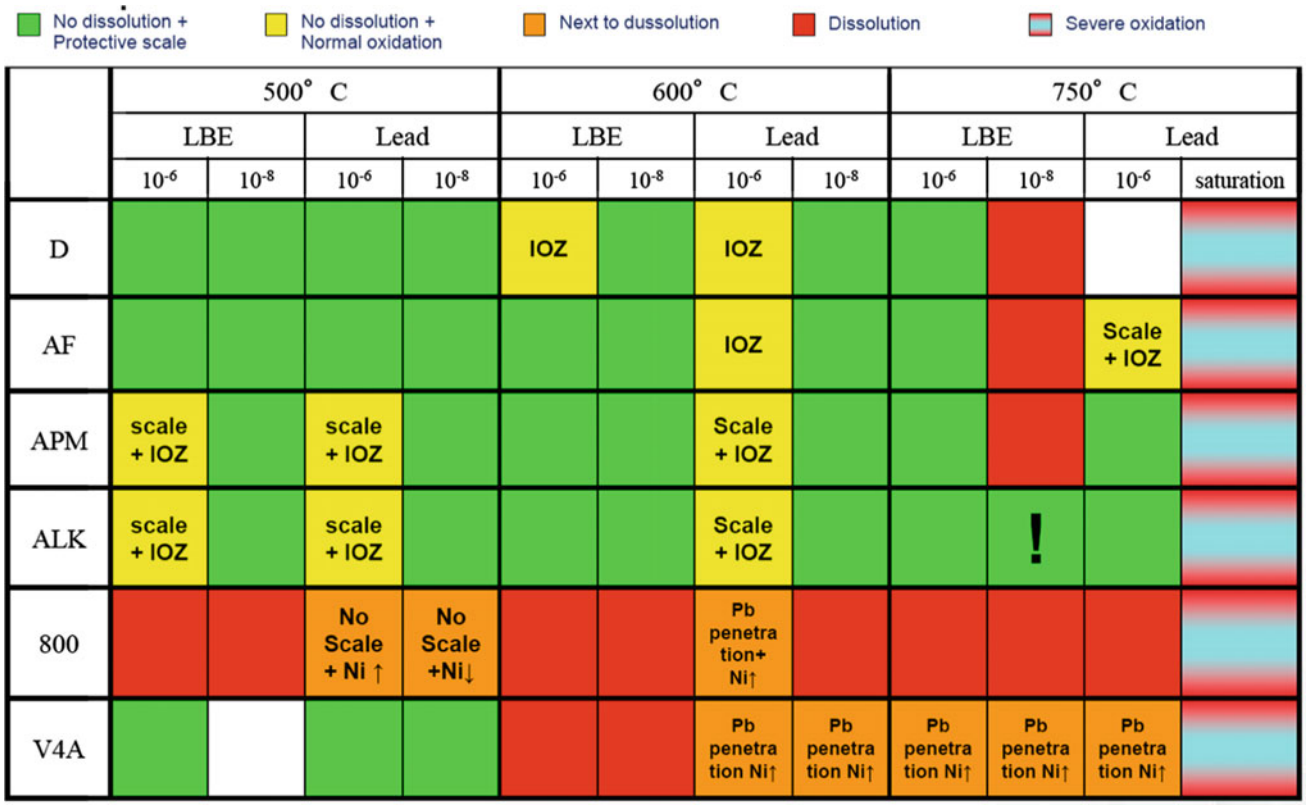

Fig. 13 Compatibility of various steels and alloys with lead and lead–bismuth eutectic mixture (LBE) up to 750 °C. From the Karlsruhe Liquid Metal Laboratory (KALLA) at KIT

temperature lead or lead–bismuth coolant. A lot of progress was made in the past 15 years in the development of corrosion resistant materials by acting on the composition, surface condition, and secondary treatment of the materials. Today, it is possible to control corrosion up to temperatures of about 600 °C for pure lead and LBE mixture (Fig. 13).

Conclusion on the Choice of Coolant

The ex-USSR military program has produced extensive experience on the use of the molten LBE mixture as a reactor coolant. With the help of controlled oxygen, the corrosion of stainless steel cladding is manageable. The residual problem is associated with the large amount of polonium produced by the neutron capture on bismuth. Our Russian colleagues claim that the majority of the polonium remains in the eutectic mixture and does not volatize within the molten metal, but this has to be better verified. In spite of this, for a civil international program, the presence of industrial quantities of polonium is to be excluded. Note that in presence of air at 55 °C, about 50 % of Po-210 is volatized in 24 h!

It is concluded that pure lead is highly preferable. The Pb–Bi eutectic mixture should be excluded for any wider commercial deployment.

A fast metal-cooled ADS may represent a practical solution to the exploitation of a pure thorium breeder. A detailed engineering design based on reference [11] has been produced by Acker Solutions. A priori, the metal coolant may be either sodium, LBE, or lead. However, some problems persist as (a) sodium is highly flammable; (b) LBE is plagued by the very large Po-210 radioactivity; and (c) lead is far less radioactive, but needs higher temperatures, which may be limited by corrosion.

Working in the Neutron Capture Resonance Region

Neutrons from a fluoride salt with graphite coolant are thermal and, as already pointed out, require on-line reprocessing. However, it is possible to considerably harden the spectrum with an appropriate choice of actinide-loaded fluoride and high density through a high content of thorium and uranium of the order of 20 % of (Th + U)F_4.

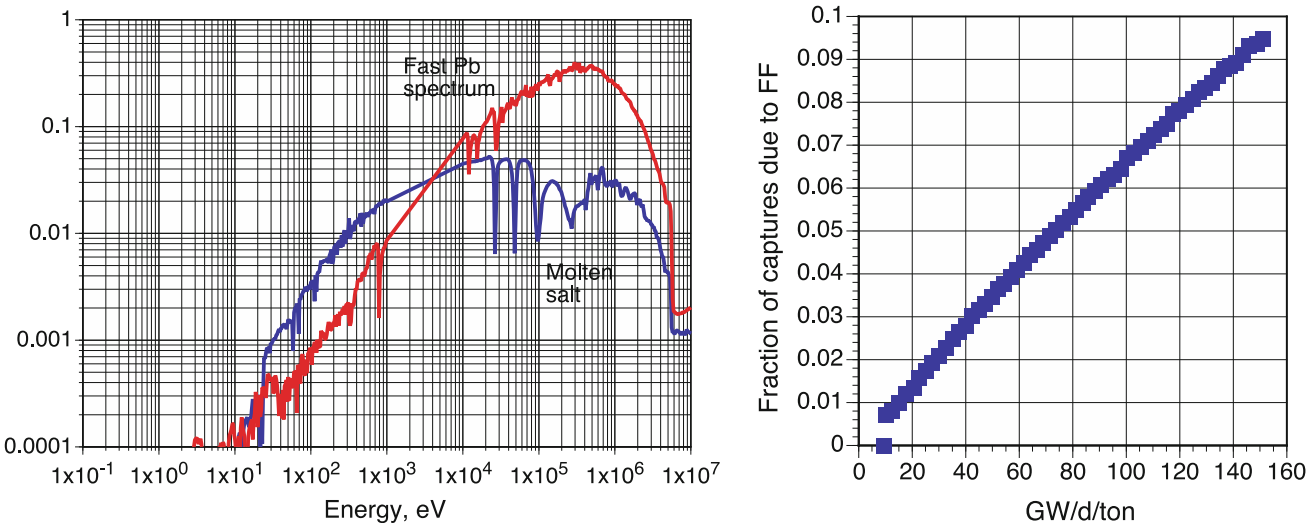

Fig. 14 *Left* Comparison of the neutron energy spectrum in a lead-cooled fast energy amplifier and in a high-density molten salt (20 % (Th + U)F_4) system [11]. *Right* Corresponding fraction of neutrons captured by fission fragments as a function of burnup in the same high-density molten salt system [11]

The resulting energy spectrum is characterized by several resonances above 3×10^4 eV and a negligible thermal contribution (Fig. 14, left).

Captures on fission fragments are limited to 10 % after a burnup of 150 GW/d/t, which corresponds to 8–10 years of continuous operation (Fig. 14, right).

Accelerator-Driven Pure Thorium System Without On-Line Reprocessing

The subcritical operation of a molten salt ADS driven by a high-energy proton accelerator (Fig. 15) can be a far simpler alternative to a thermal critical reactor with quick, integral on-line reprocessing. A spread out beam directly hits the molten salt (with 1 GeV protons, the number of neutrons per incident proton is about 15). The operating temperature is about 640 °C. All metal parts in contact with the salt are made of an alloy developed at ORNL, Hastelloy-N, which has proved to be nicely compatible with fluoride salts. The heat exchanger may use LiF–BeF_2 (66:34 mol%) as a secondary salt. Such systems should be simple and relatively cheap to build. It provides multiple containments to protect against molten salt leaks.

Some Considerations for the Optimal Arrangement

Although on the face of it, a critical molten salt system is attractive for its simplicity and cost, these systems have some severe drawbacks, including:

- The value of the criticality coefficient, k, depends on the actual location of the liquid salt inside the reactor. Can the absolute stability of k to the required level ($<10^{-4}$) be ensured?
- The radiotoxicity of the molten fuel in its liquid form is huge. The classic alternative with many separate solid fuel rods may represent a better protection against accidents.

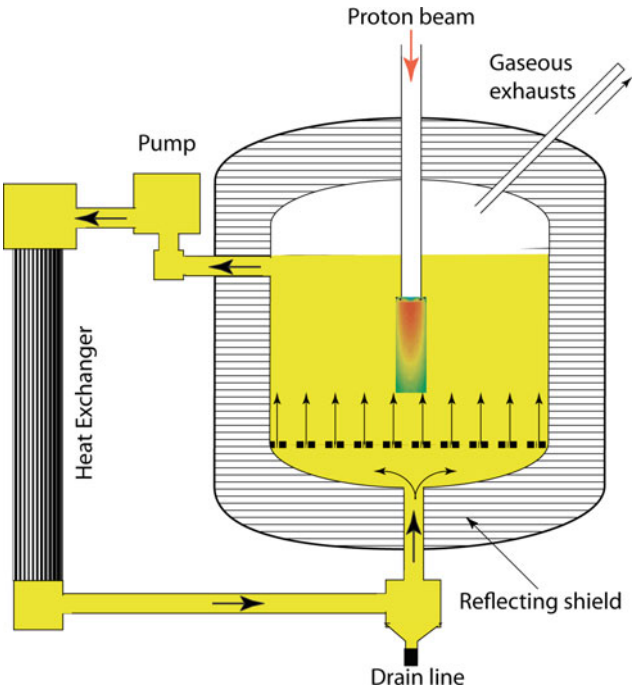

Fig. 15 Molten salt reactor driven by a high-energy proton accelerator inserted from the top into a vacuum beam tube. The molten salt is drained and reprocessed possibly only after a burnup of 150 GW/d/t

However, I believe that the molten salt, per se, has many interesting properties, such as the higher operating temperature (640 °C) and the widely proven compatibility with appropriate steel cladding and other materials.

An arrangement, which may be worth considering, is an Acker Solutions-like configuration [11], but with molten salt coolant replacing both the lead in a fast neutron configuration and the conventional fuel pins.

New Reprocessing Methods

The end of the fuel cycle in the case of thorium needs major new developments and it deserves new methods. It would not be appropriate to throw away the seeds at the end of each cycle as this would prevent the indefinite repetition of several very long-lasting breeding thorium-fed nuclear fuel cycles. It is presumably possible to apply standard aqueous processes, such as THOREX, PUREX, etc., to the thorium fuel cycle, to allow actinides to be separated and later recovered by a centralized reprocessing facility, but it will certainly be both complicated and costly. Aqueous solution systems use an organic solvent, which is easily decomposed and deteriorated in the case of the closed thorium–uranium cycle by the substantial effects of radiation damage. The creation of a new, appropriate end-of-cycle fuel reprocessing has been, so far, an undisclosed necessity for the practical realization of any closed thorium cycle.

A New, Simple Dry Reprocessing Method for Uranium and Plutonium Seeds

The volatility of fluorides has been widely utilized to extract actinides with a valence of five or greater (uranium, neptunium, and plutonium). Fluoridation is one of the universally used processes. It is a fortunate circumstance that uranium and plutonium are the main "seeds" in the case of fast energy amplifier thorium fuel. The initial UO_2 is converted with hydrofluoric acid (HF) to uranium tetrafluoride, UF_4. Oxidation with fluorine yields the end product UF_6 through the following process:

$$UO_2 + 4HF + [500\ ^\circ C] \rightarrow UF_4 + 2H_2O;$$
$$UF_4 + F_2 + [350\ ^\circ C] \rightarrow UF_6$$

Spent thorium, fission fragments, and the minor actinides, protactinium, americium, and curium, constitute the "waste". Uranium and plutonium are the "seeds". New thorium may be injected into each cycle as thorium is cheap and abundant. The final transformation from fluoride to oxide is currently performed with the help of hydrogen and steam/hydrogen flowing at appropriate temperatures (Fig. 16).

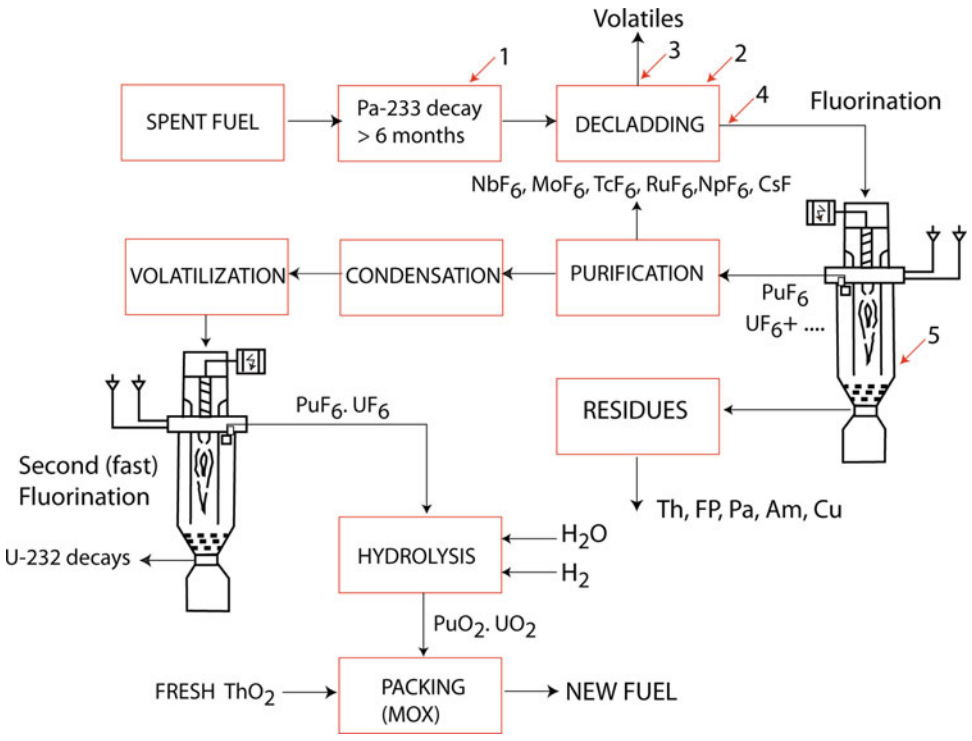

Fig. 16 Schematics of the reprocessing of spent thorium fuel to separate out fission fragments and minor actinides, and to regenerate thorium and uranium oxides, which are reintroduced as seeds in the fuel, in addition to fresh thorium oxide

Today, about 95 % of the depleted uranium produced is stored as uranium hexafluoride, UF_6, in steel cylinders in open-air yards, close to enrichment plants.

Long Lasting Toxicities for the Simple Fluorination Method

The long-term radiotoxicity of the resulting waste (Fig. 17) is due to two main contaminants:

- The long-lived Pa-231 isotope, which may be recovered with the help of bismuth;
- The Th-230 isotope, which is present in the discharged thorium and is radioactive.

Thorium may be further recycled as "seed" with an appropriate chemical extraction. The remaining "waste", namely the uranium and plutonium leaks, curium and americium, have a radiotoxicity that is negligible compared to that of the fission fragments.

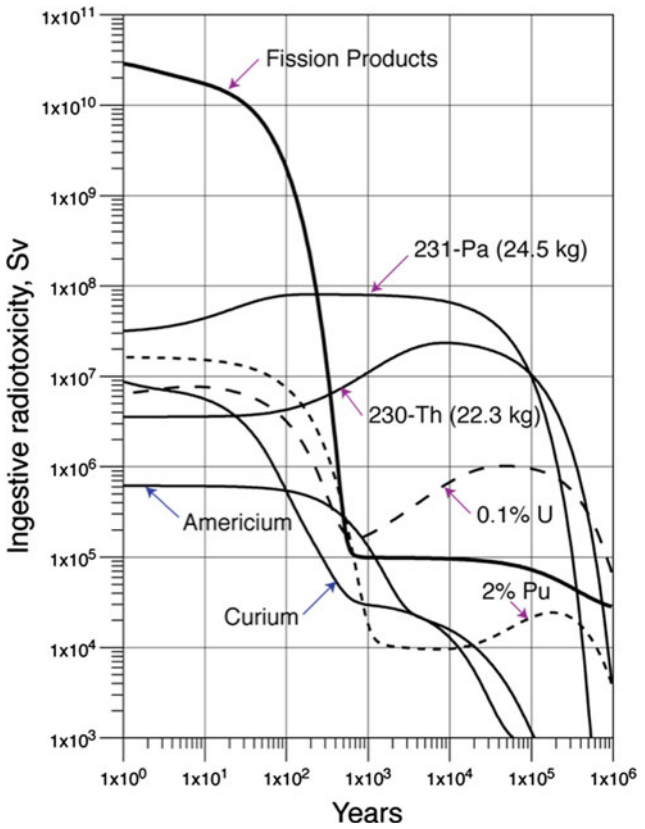

Fig. 17 Ingestive radiotoxicity of the EA1600 asymptotic load after 120 GW/d/t. Taken from [11]

Removing the Tl-208 Radioactivity to Make New MOX Fuel

Fluorination offers an attractive and simple method for separating the uranium seeds from the gamma-active equilibrium contaminants. Assuming a second (quick) batch fluorination, uranium and plutonium hexafluorides are separated out from the other locally generated decay products, which include those from the whole decay chains of U-232 and Pu-236, progressively building up the strong gamma-lines from Tl-208. In the manufacturing phase that follows, MOX pins are quickly assembled, while the newly built-up gamma-ray background is still small. The final conversion into oxides is performed with the help of hydrogen and hot steam. Oxides are reduced to a MOX-type powder and mechanically mixed with fresh ThO_2 oxide (Fig. 18).

New Fuel Batches: A Tentative Example

The new fuel typically consists of 19 tons of new metallic thorium, 3.4 tons of metallic uranium, and a residue of 130 kg of metallic plutonium, from the seeds of the previous batch. Individual fuel pins contain about 6 kg of MOX fuel each.

The total Tl-208 equilibrium contamination in the spent fuel is huge; it corresponds to a gamma activity of 8.1×10^5 Ci. Each cycle is expected to contribute to the manufacturing of about 15 new pins, that is, ≈90 kg of new MOX fuel. The regenerated gamma-contamination grows progressively with time (Fig. 19). For 90 kg of new MOX fuel, it is about 1 mCi

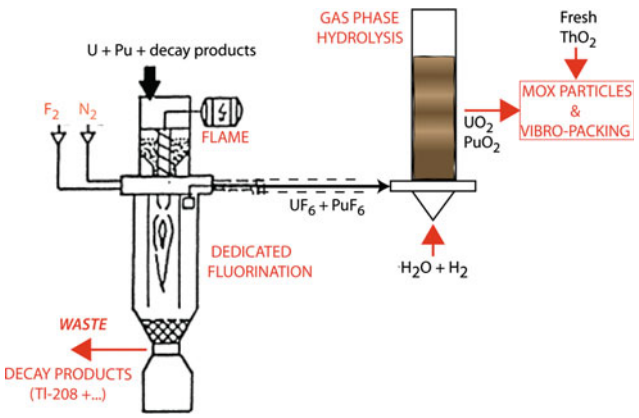

Fig. 18 Schematic of the process generating MOX thorium fuel shortly after the extraction of the decay products coming from uranium and its decay chain, but before significant build-up of the strong gamma-radiation

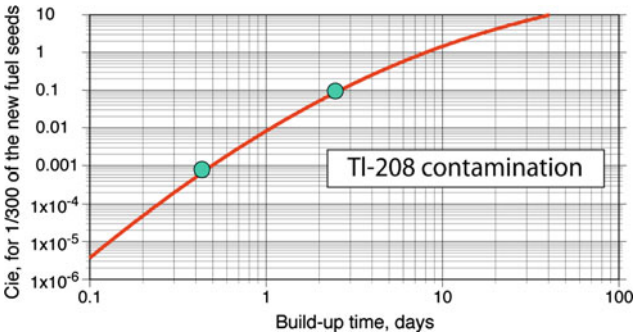

Fig. 19 Build-up of the gamma activity due to Tl-208 as a function of time after the extraction of the uranium and plutonium decay-chain products. Taken from [11]

after 5 h, but 0.1 Ci after 2.5 days. The manufacturing process must be performed as quickly as possible in order to minimize the emitted radioactivity.

Conclusion on Thorium Fuel Reprocessing

Fuel "reprocessing" or, more precisely, fuel "regeneration" is an essential part of the thorium process. It is a fortunate circumstance that most of the seeds are uranium. UF_6 is the most volatile uranium compound known to exist (PuF_6 is also volatile). Uranium and plutonium are therefore the ideal "seeds" for a breeding process. A simplified reprocessing may then consist of keeping the spent thorium, fission fragments, and minor actinides (Pa, Np, Am, and Cm) as "waste" and uranium and plutonium as "seeds".

My Own Recommendations

My own recommendation is to consider a 600 MW_e sub-critical, proton beam-driven reactor prototype, along the lines of that in reference [11] and the energy amplifier feasibility study by Aker Solutions ASA, but using salt instead of metal.

The fuel is contained in a conventional structure made of stainless steel rods and metal oxides operating at about 640 °C. All parts in contact with the LiF–BeF_2 cooling fluoride coolant are made of Hastelloy-N alloy—which has been proven to be compatible with LiF–BeF_2. The start-up fuel is conventional thorium–MOX, progressively bred into fissile U-233 and other uranium isotopes. The duration of each cycle is about ten years and the reactor lifetime exceeds 200 years. A simple in situ fuel reprocessing/regeneration is performed, based on the transformation of the uranium and plutonium seeds into hexafluorides.

Spent thorium, fission fragments, and the minor actinides americium and curium are "waste". Uranium and plutonium are "seeds". Long-lived Pa-231 from protactinium may be recovered as "seed" with the help of bismuth.

The discharged thorium is radioactive as it contains some Th-230. New thorium may be injected at each cycle or recovered as "seed".

Given the potential of the energy amplifier (Table 5) in terms of abundant, safe and clean energy production it would be wise to build an industrial demonstrator as soon as possible.

Table 5 Summary of main properties of the proposed energy amplifier

Item	Energy amplifier
Fuel	Natural thorium
Fuel availability	Practically unlimited
Chemistry of fuel	Regenerated every 10 years
Waste disposal	At level of coal ashes after 600 years
Operation	Extrapolated from present reactors
Technology	No major barrier
Safety	System not critical; no meltdown
Proliferation resistance	Excellent; sealed fuel tanks
Cost of energy	Competitive with fossil fuels
Credibility	Proven at zero power

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Part II

Invited Speakers: National and International Thorium Programmes

Towards Sustainable, Secure, and Safe Energy Future: Leveraging Opportunities with Thorium

Anil Kakodkar and S.B. Degweker

Abstract

The growing economic empowerment of an increasing part of the world's population and small amount of carbon space available, necessitates a quick shift to non-fossil energy sources that are large enough to meet future energy requirements. Apart from electricity, energy in fluid form, derived through non-fossil means is also needed. It is, thus, important to progressively replace fossil fuels and increase the share of nuclear and solar power in the overall energy mix. For expansion of nuclear capacity, it is important to squarely address various fears associated with nuclear energy by eliminating the possibility of any adverse impact in the public domain. Since time is running out, we need to explore how to achieve this objective through reconfiguration of available technologies even as we develop new technologies specifically for the purpose. This paper argues that the use of thorium, together with innovative reactor designs such as that of the advanced heavy water reactor (AHWR), under development in India can, by and large, eliminate many of the perceived risks associated with nuclear energy. In the long run, a mix of advanced technologies such as molten salt reactors, accelerator-driven systems, and fast reactors will be necessary to sustain nuclear energy.

Keywords

Sustainable energy • Thorium fuel • Advanced heavy water reactor • Fast reactors • ADS and molten salt reactors

Introduction

Carbon emissions from fossil fuels used for powering motor vehicles and for generating electricity are predicted to cause large-scale climatic changes [1] with catastrophic effects on the global environment, economics, and politics. Public concern on this account has led to international negotiations under the UN Framework on Climate Change, such as the Kyoto Protocol, aiming to limit carbon dioxide and other greenhouse gas emissions and setting targets for their phased reduction. At the same time, economic growth in an increasing part of the world continues to drive the demand for energy upwards. Much of this demand is expected to be met with fossil fuels. This clearly is not sustainable both in terms of energy resources as well as stability of the climate. It is, thus, imperative to look at solutions beyond the "business as usual" approach.

Among the various alternatives to fossil fuels are those based on renewable sources, such as wind, hydro, solar, biomass, etc. Although these are important in themselves, they have limitations and large-scale growth in much of the developing world will have to be powered by nuclear energy. Presently, nuclear energy has hit several barriers and is perceived to be too risky to be acceptable in some countries. In this paper, we examine these risks and argue that the use of thorium fuel together with new innovative reactor

Anil Kakodkar presented this paper at the Thorium Energy Conference 2013 (ThEC13)

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systems can, by and large, eliminate these risks. In the long run, a mix of advanced technologies such as molten salt reactors, accelerator-driven systems, and fast reactors will be necessary to sustain nuclear energy.

We also examine the problems associated with nuclear energy, and we discuss how the properties of thorium fuel help in mitigating these risks. We also look at the design of the advanced heavy water reactor (AHWR) in this connection. Finally, we touch upon other advanced nuclear technologies and concepts that might play an important role in making large-scale deployment of nuclear energy around the world more acceptable.

Sustainable Development of the Energy Sector

According to estimates by the International Energy Outlook 2013, the primary energy consumption is expected to increase by 56 % by 2040 and if things continue as they are, most of this will be derived from fossil fuels. Table 1 depicts the projected [1] environmental consequences, including average temperature change and the consequent rise in sea level, etc., under different scenarios. The need to limit the temperature rise to 2 °C seems to be generally recognized [2]. This would need a 70 % cut in greenhouse gas (GHG) emission, requiring 85 % of energy to come from clean sources. This would mean that we urgently replace fossil hydrocarbon and derive most of the primary energy from nuclear and solar. Also, we should explore the possibility of recycling carbon dioxide in the atmosphere by sequestering [3] or by producing non-fossil hydrocarbons by using atmospheric CO_2 and hydrogen derived from nuclear or solar as the primary energy source and the use of biomass. This requires high temperature capability to trigger the necessary reactions. A possible path for a transition to a fossil-carbon-free energy cycle is shown in Fig. 1.

Nuclear energy showed great promise and rapid growth in the early years. However, the growth has slowed down since the mid-eighties and has not picked up since then. In spite of the strong motivation for a switch over to nuclear energy, there continues to be resistance to its large-scale deployment. It is possible to identify the following main reasons for this resistance:

1. Irrational fear of radiation caused by linear no threshold (LNT) logic;
2. Potential for large-scale displacement of people following a severe accident;
3. Panic potential following a terrorist action;
4. Unresolved issues related to spent fuel disposal and constraints on recycling;
5. Regulatory delays.

With regard to the first two points, we merely note the following evidence to the contrary. There are several areas around the world that have radiation levels much above the average, but the residents of these areas show no adverse health effects [4–6]. Colorado, USA, with a population over five million, is one such place [4]. Colorado should have an excess of 200 cancer deaths per year as per the LNT logic. In fact, it has a rate lower than the US national average. Residents of Ramasar, Iran, receive an annual dose many times higher than the radiation level in the Chernobyl and Fukushima exclusion zones. Nevertheless, people living in Ramsar live longer and healthier lives [4–6]. We also know that other countries, such as India, China, and Norway, have areas where radiation level is many times higher than 2.4 mSv/y world average.

In spite of the lack of evidence on the health consequences due to radiation exposure below a threshold, the mindset driven by LNT logic leads to disproportionately large-scale displacement of the population following a severe accident. This leads to fear with regard to the impact of a potential accident in the public domain. Without getting into the merits or demerits of the LNT logic, we need to recognize that it is here to stay and it implies significant off-site impact in a severe accident, which is no longer acceptable.

Table 1 Projected environmental consequences for various assumed periods when carbon dioxide emission into the atmosphere reaches a peak

Category	CO_2 equivalent conc. at stabilisation (ppm)	Peaking year for CO_2 emissions	% Change (over year 2000) in global CO_2 emissions in 2050	Global ave. temp. increase above pre-industrial (°C)	Global ave. sea level rise above pre-industrial (m)	No. of accessed scenarios
1.	445–490	2000–2015	−85 to −50	2.0–2.4	0.4–1.4	6
2.	490–535	2000–2020	−60 to −30	2.4–2.8	0.5–1.7	18
3.	535–590	2010–2030	−30 to +5	2.8–3.2	0.6–1.9	21
4.	590–710	2020–2060	+10 to +60	3.2–4.0	0.6–2.4	118
5.	710–855	2050–2080	+25 to +85	4.0–4.9	0.8–2.9	9
6.	855–1130	2060–2090	−90 to +140	4.9–6.1	1.0–3.7	5

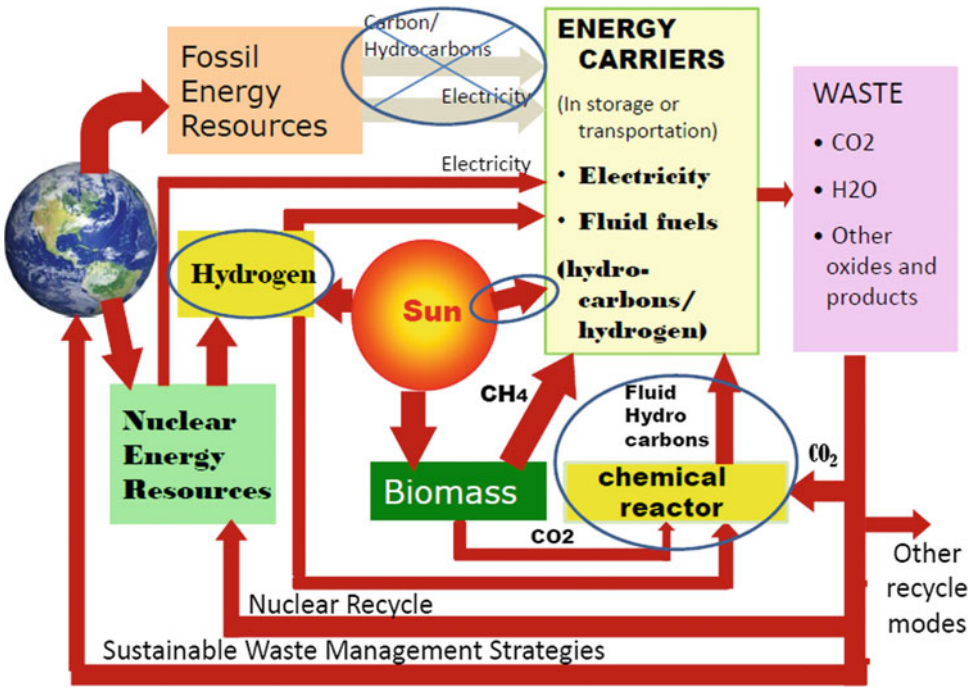


Fig. 1 Possible pathways for transition to a fossil-carbon-free energy cycle

This problem has been further accentuated due to the rise of terrorism. Accidents such as the ones at Chernobyl or Fukushima also give a jolt to the regulatory mechanism, leading to regulatory ratcheting and consequent delays as well as financial uncertainties.

With regard to the problem of waste disposal, a catch-22 situation arises as a result of the policy of not reprocessing and recycling spent fuel and treating it as waste following one-time use in the nuclear reactor. Spent fuel contains legally defined nuclear material, which must be secured and safeguarded at all times and, therefore, cannot be permanently disposed. Disposal of spent fuel in once-through mode is, thus, an issue that is not likely to be resolved. Hence, it is necessary that the once-through fuel cycle must be replaced by a closed fuel cycle, which some countries are already adopting. This is necessary not only for better resource utilization, but also for reducing waste volumes and facilitating waste disposal without the fear of loss of control over fissile material such as plutonium. As we will see later, use of thorium in the fuel cycle makes it much more proliferation resistant while enhancing its energy resource potential.

By adopting innovative system configurations, it is possible to design reactors that would have no impact in the public domain and would also address the other issues, even with existing technologies. In India, we are making a beginning in this direction through the development of an advanced heavy water reactor (AHWR) [7] using thorium. The design also addresses the concerns relating to insider and outsider security threats. Thorium plays a major role in burning plutonium and making the fuel cycle inherently more proliferation resistant, as we show in the next section. In the long run, these efforts will, of course, need to be supplemented with advanced technologies involving fast reactors, molten salt reactors, and accelerator-driven systems.

Thorium as a Solution: The Advanced Heavy Water Reactor

Thermo-Mechanical and Neutronic Properties of Thorium

The favorable physical and chemical properties of thorium fuel over uranium are well known [8]. Thorium oxide has a higher melting point and thermal conductivity (Fig. 2) and a lower thermal expansion coefficient. Fission gas release is one order of magnitude lower. Thorium oxide is chemically stable and, unlike UO_2, it does not get oxidized or react with water. This means that for a given power level, thorium fuel would operate at lower fuel temperatures, with less fission gas release, and we can expect better dimensional stability of the fuel and chemical stability upon long-term storage.

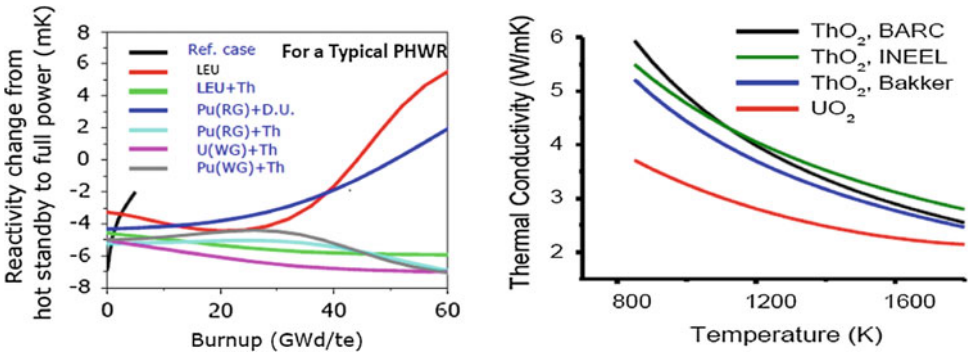


Fig. 2 Properties of thorium fuel: (*left*) reactivity change from hot standby to full power in a thorium-fueled reactor; (*right*) thermal conductivity of the oxide

Thorium and its derived fuel, ^{233}U, also have better neutronic properties such as the highest value of η at thermal energies, which cause thorium-based reactors to sustain long burnups, a point discussed in greater detail later. In Fig. 2, we also show the change in reactivity from hot standby condition to full power for a variety of fuels. As a result of their nuclear and thermo-physical properties, Th-based fuels tend to have favorable reactivity coefficients leading to more stable reactor performance.

The Advanced Heavy Water Reactor (AHWR)

The advanced heavy water reactor combines the above-mentioned thermo-physical properties of Th fuel and some innovative design features to achieve the objectives stated. It is a heavy water moderated, boiling light water cooled vertical pressure tube type reactor under development at the Bhabha Atomic Research Centre (BARC). It is an innovative configuration, using available technologies and it

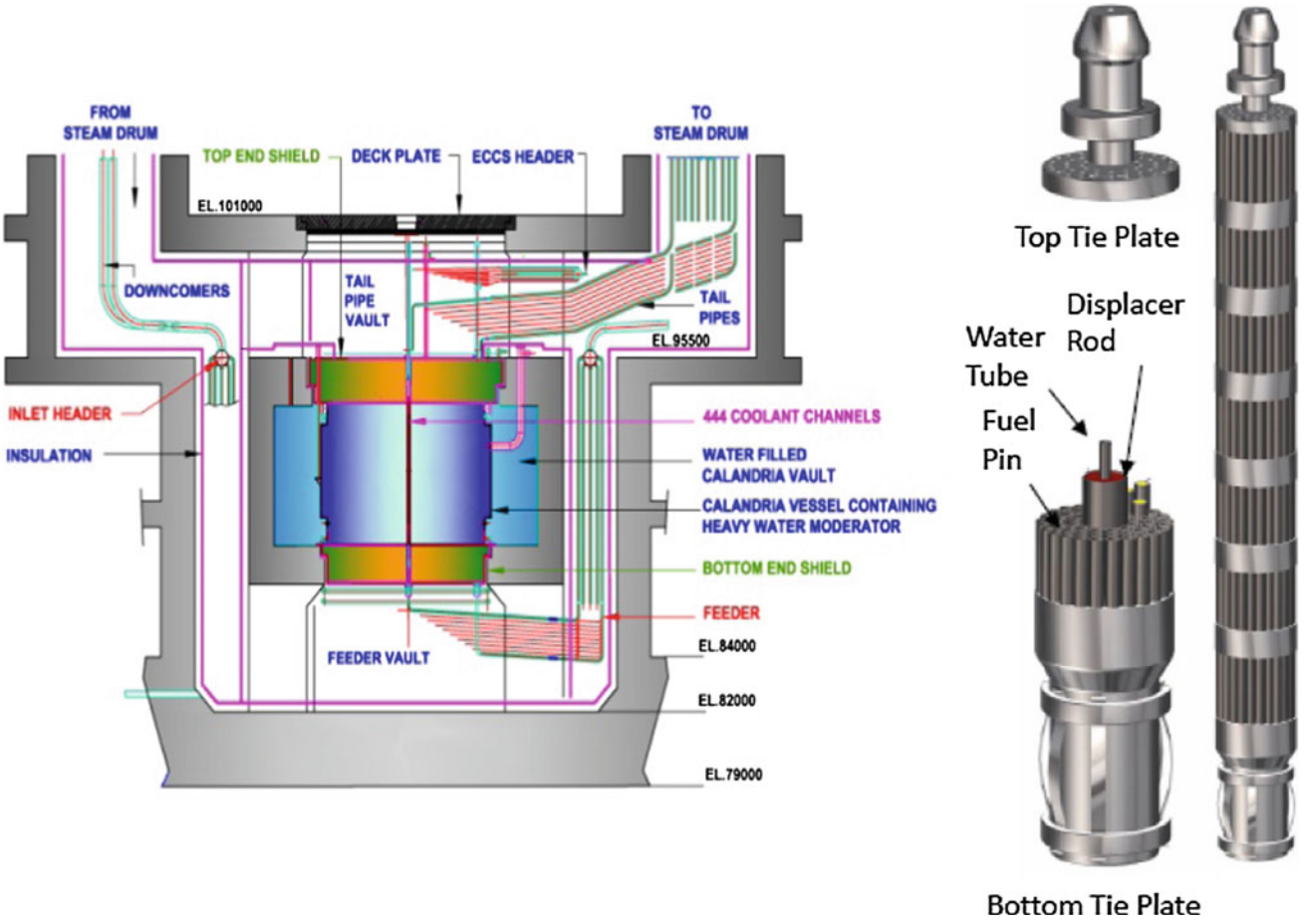


Fig. 3 *Left* Schematic of the AHWR; *Right* Fuel design of the AHWR

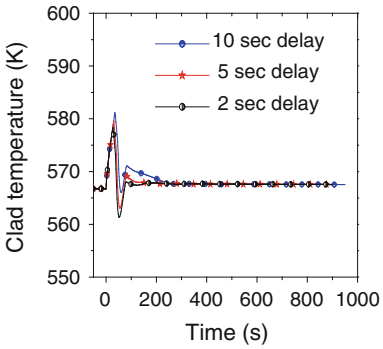


Fig. 4 Expected peak clad temperature in the event of station blackout and failure of the shutdown systems in the AHWR

should nearly eliminate any impact on the public domain. The design can use a range of fuels viz., low-enriched uranium (LEU), U–Pu, Th–Pu, LEU–Th, and ^{233}U–Th, in the entire core. The design incorporates several passive features such as passive on-power and decay heat removal, a passive shut-down system to address insider threat scenarios, and it provides a grace period of more than three days in the event of an accident with no radiological impact on the public domain. The system is designed for a life of one hundred years, with easily replaceable coolant channels. The design power is 300 MW_e with a significant fraction of its energy derived from Th. A schematic of AHWR and its fuel is shown in Fig. 3.

The AHWR 300-LEU [9], a recent version of the AHWR, is a simple 300 MW_e system fueled with LEU–Th; it has advanced passive features, a high degree of operator forgiving characteristics, no adverse impact on the public domain, high proliferation resistance, and inherent security strength. It provides a robust design against external and internal security threats, including insider malevolent acts. The system is designed such that even in the extreme event of station blackout and simultaneous failure of the two shut-down systems, the peak clad temperature rises only marginally, as shown in Fig. 4, thereby completely precluding any impact on the public domain, owing to the passive elements incorporated into the design.

Probabilistic safety analysis (PSA) studies for the AHWR have been carried out [10]. The study includes plant familiarization and identification of design aspects important to severe accidents; PSA level-1, viz., identification of significant events with large contribution to the core damage frequency (CDF); PSA level-2, viz., source term evaluation within the containment; and PSA level-3, viz., release from the containment and atmospheric dispersion with analysis of consequences. The results of the study are illustrated in Fig. 5. The results show that the probability of exceeding permissible thyroid dose is lower by six to seven orders of

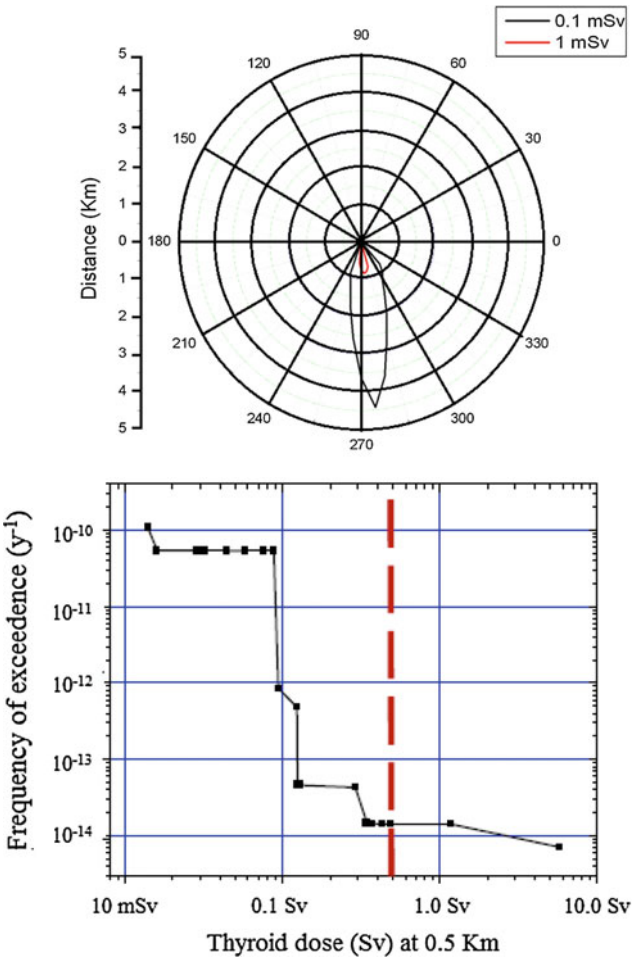


Fig. 5 Results of probabilistic safety analysis of the AHWR. *Top* Iso-dose contours for thyroid—200 % break of reactor inlet header (RIH) and wired shutdown system unavailable (using wind conditions in January on western Indian side). *Bottom* Variation of dose with frequency of exceedence

magnitude. In the event of a double-ended header break together with the failure of the wired shut-down systems, any dose in excess of 1 mSv remains well within the plant area, and outside the plant limits the dose is well below the 1 mSv limit. Thus, the system has practically no impact on the public domain.

Thorium as a Solution to Other Issues

Utilization of Uranium Resources

^{233}U has the smallest value of α (the capture-to-fission ratio) among the fissile nuclides, which is responsible for the highest thermal value of η. On the other hand, thorium has a higher thermal capture cross section compared to ^{238}U. For these reasons, if we consider an initial mixture of ^{235}U and

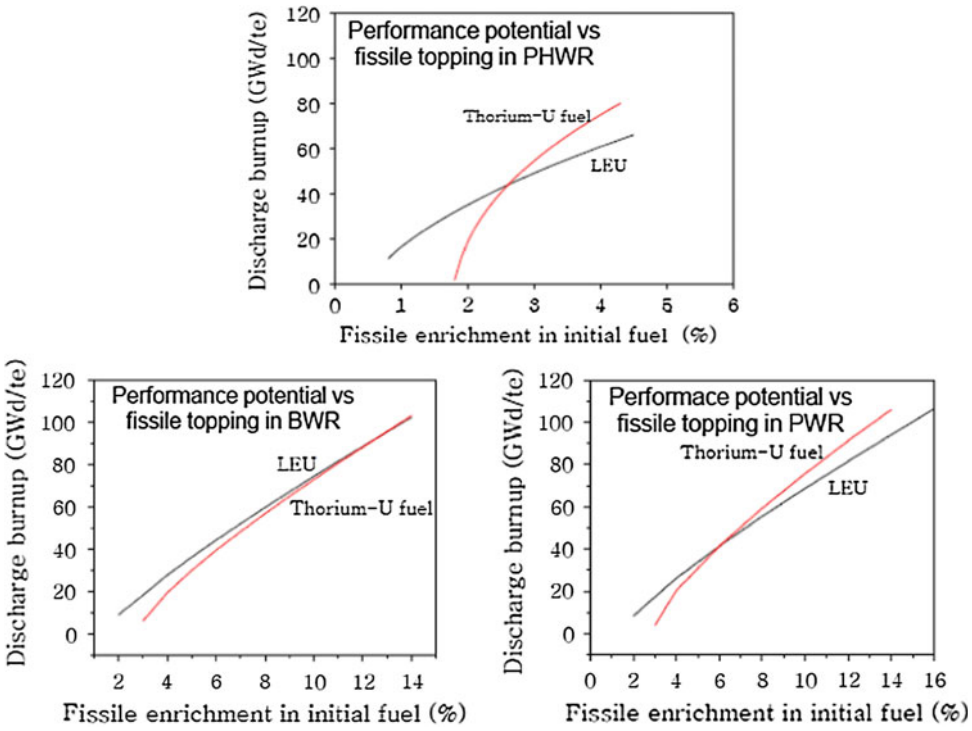


Fig. 6 Comparison of thorium and ^{238}U as fertile hosts with regard to discharge burnup achievable for a given initial fissile enrichment, for PHWR, BWR and PWR

Th and compare its performance with that of ^{235}U and ^{238}U (LEU), it is found that the amount of initial fissile material required to yield a given value of the discharge burnup is lower for LEU at low burnups, but as we increase the burnup, typically beyond 40 GWd/t, the ^{235}U–Th mixture requires lower amounts of initial fissile material and, thus, shows an advantage. As high burnup fuels are becoming more common nowadays in thermal reactors, this is an advantage that needs to be exploited.

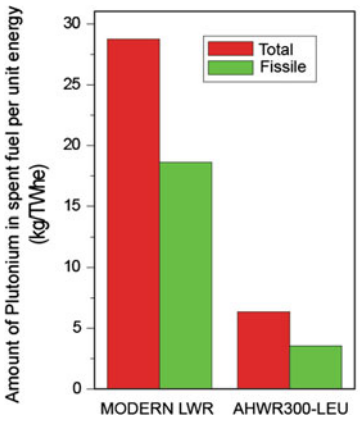
Figure 6 illustrates this effect. By using ^{235}U as the initial fissile material and thorium as the fertile host, in pressurized heavy water reactors (PHWRS), pressurized water reactors (PWRs), and boiling water reactors (BWRs), we see that at higher burnups there is better fertile to fissile conversion, a smaller reactivity swing, greater energy generation from the in situ generated fissile material, and consequently better uranium utilization in terms of GWd per ton of mined uranium. In particular, as a consequence, the AHWR300-LEU provides better utilization of natural uranium owing to its much higher burnup.

Long-Lived Waste

Disposal of spent fuel remains an unresolved issue. There is already a large (>200,000 tons) spent fuel inventory. Another 400,000 tons are likely to be generated by the year 2030 (World Nuclear Association estimate). Permanent disposal of the fuel remains an unresolved issue fraught with unacceptable safety and security risks. We need to adopt ways to destroy the spent fuel through recycling.

Thorium provides an effective answer for the safe recycling of spent nuclear fuel as may be seen from Fig. 7. The production of Pu and minor actinides is much lower if Th is used as the fertile host instead of ^{238}U. Moreover, the isotopic vector of both uranium and Pu in the spent fuel makes the fuel cycle proliferation resistant. The Pu in the spent fuel contains a much lower fraction of the fissile isotopes ^{239}Pu and ^{241}Pu. Owing to the much larger fraction of ^{238}Pu, which produces heat and the hard gamma emissions of ^{232}U, the fuel is rendered completely unsuitable for weapons. Nevertheless, the uranium in the spent fuel is suitable for recycle and reuse in other reactors owing to the significant fissile content. Moreover, the Pu can be recycled in fast spectrum reactors.

Thorium is an excellent host for disposal of excess plutonium. Recycling Pu mixed oxides (MOX), as is practiced in current PWRs and BWRs, does not lead to significant Pu burning due to its continued formation from ^{238}U. Another option is the use of an inert matrix fuel for burning Pu. Although Pu burning is more in this scenario, due to degraded reactor kinetics parameters, only a fraction of the core can be loaded with this fuel. Thorium enables more effective burning of Pu while maintaining acceptable performance characteristics. This is illustrated in Fig. 8, which shows the discharge burnup dependence on the initial fissile Pu content in the fuel. Also shown is the fissile content in the discharged fuel. As the initial fuel content increases, we get higher burnup. Beyond a certain burnup, most of the initial Pu is consumed and any additional increase in burnup is due to the ^{233}U formed.



Isotope	Modern LWRs (%)	AHWR300 LEU (%)
^{238}Pu	3.50	9.54
^{239}Pu	51.87	41.65
^{240}Pu	23.81	21.14
^{241}Pu	12.91	13.96
^{242}Pu	7.91	13.70
^{232}U	0.00	0.02
^{233}U	0.00	6.51
^{234}U	0.00	1.24
^{235}U	0.82	1.62
^{236}U	0.59	3.27
^{238}U	98.59	87.35

Fig. 7 Comparison of spent fuel composition in LWRs and AHWR300-LEU. *Left* Total and fissile Pu content; *Right* Pu and U vectors

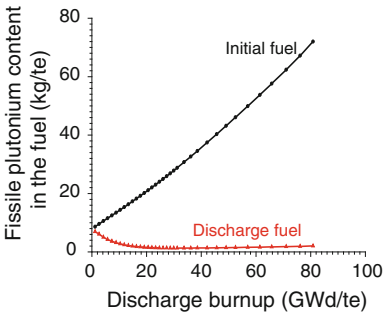


Fig. 8 Variation of initial and final fissile Pu content with discharge burnup in a Th host

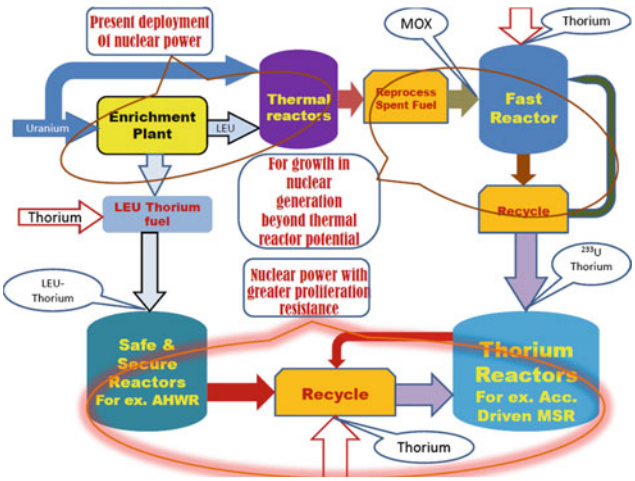


Fig. 9 The role of thorium in securing a sustainable future for nuclear energy

Proliferation Resistance of Th Fuels

The uranium produced as a result of thorium irradiation invariably contains the isotope ^{232}U, the daughter products of which produce a hard gamma dose. Although this presents a major technological challenge for the recycling of thorium fuels, it has potential benefit from a non-proliferation point of view. Firstly, it makes the uranium easy to detect in case of theft. Secondly, fabrication of any device based on such uranium requires a degree of sophistication not readily available. This renders the making of a device based on uranium derived from thorium extremely difficult.

Importance of Recycling and the Role of Fast Breeder Reactors, ADS, MSR

Although the AHWR300-LEU enables comparable utilization of uranium in a safe manner, and also reduces the production of minor actinides (MA), issues related to long-term sustainability of nuclear fuel resources and waste management would still need to be addressed through the recycling of fissile and fertile materials in fast reactors and accelerator-driven systems (ADS). Minor actinides fission better in the fast spectrum. The rate of incineration would be highest if the entire core is made up of MA or a mix of Pu and MA fuel. However, due to the much lower value of the delayed neutron fraction of MA fuels, a critical core with such fuel would be unacceptable. Moreover, such a core has a practically zero Doppler coefficient of reactivity and a large positive void coefficient. Accelerator-driven reactors are insensitive to these parameters and can be operated safely. Hence, the fast ADS would play an important role in waste transmutation.

Presently operated once-through cycle reactors utilize less than 1 % of the mined uranium. To improve resource utilization, it is necessary to have recycling with breeding. This will mean deployment of fast breeder reactors in the long run. Eventually, we would have to change to the Th cycle. To ensure the sustainability of thorium resources through breeding, accelerator-driven molten salt reactor (MSR) systems seem to have high potential. Figure 9 depicts a possible approach to making the transition to large-scale thorium utilization that would be sustainable in terms of energy resources for the future, would address issues of long-term waste management, safety, security, and, most importantly, transition to non-fossil energy supply.

Conclusion

Thorium is a good host for efficient and safe utilization of fissile materials. It can support a greater geographical spread of nuclear energy with lower risk. Thorium can facilitate the resolution of waste management issues and enable realization of the full potential of the available uranium. Fast breeder reactors would, however, be necessary for growth in

nuclear power capacity well beyond thermal reactor potential. Fast reactors as well as uranium fuel enrichment and recycling would, however, need to be kept within a more responsible domain.

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Thorium Energy and Molten Salt Reactor R&D in China

Hongjie Xu

Abstract

Thorium-based molten salt reactors (TMSRs) have interesting characteristics and applications, including thorium energy utilization, hydrogen production at high temperature, water-free cooling, and small modular design. These properties make TMSRs one of the best approaches to solve the energy and environment issues of China. In January 2011, the Chinese Academy of Sciences (CAS) launched the TMSR project, which strives to realize effective thorium energy utilization and composite utilization of nuclear energy in 20–30 years.

Energy Demand and Environmental Challenges

China is the world's most populous country with a fast-growing economy that has led it to be the largest energy consumer in the world. However, fossil fuels, particularly coal, continue to be the leading source of the electricity generation and installed capacity [1] (Fig. 1). Subsequently, China is also the world's leading CO_2 emitter, releasing 8715 million metric tons of CO_2 in 2011. Its government plans to reduce both carbon intensity (carbon emissions per unit of GDP) by 17 % and energy intensity (energy use per unit of GDP) by 16 % between 2010 and 2015. It intends to reduce its overall CO_2 emissions by at least 40 % between 2005 and 2020.

Air pollution is another issue induced by the huge coal consumption. It is indicated that about 75 % of China's total air pollution comes from combustion of fossil fuels. Options for coal substitution by carbon-free or low-carbon energy sources and improving energy efficiency are not only the requirements for CO_2 emission mitigation, but also the requirement for local environmental protection and air pollution reduction. Benefiting from high energy density, low-carbon emissions, and the potential for sustainable development, development of nuclear power has become one of the strategic focuses of China's medium and long-term energy development plan.

At present, China has 21 nuclear power units in service with an installed capacity of 18 GW_e, which accounts for ~1 % of the total power generation. Another 28 reactors, with a total capacity of 29 GW_e, are under construction [2]. It was estimated by the World Nuclear Association (WNA) that, in 2060, China's nuclear power capacity will be about 150–700 GW_e [3] and the corresponding natural uranium consumption would be approximately 40,000–170,000 ton/year. Clearly, the estimated deposits of natural uranium in China would not support this in the long term.

Th–U Fuel Cycle and Molten Salt Reactors

Nuclear fission energy can be divided into two categories, uranium-based and thorium-based, where the entire nuclear power industry is all uranium-based currently. In both the U and Th fuel cases, three types of fuel cycles are proposed; a once-though fuel cycle, a modified open fuel cycle, and a fully closed fuel cycle.

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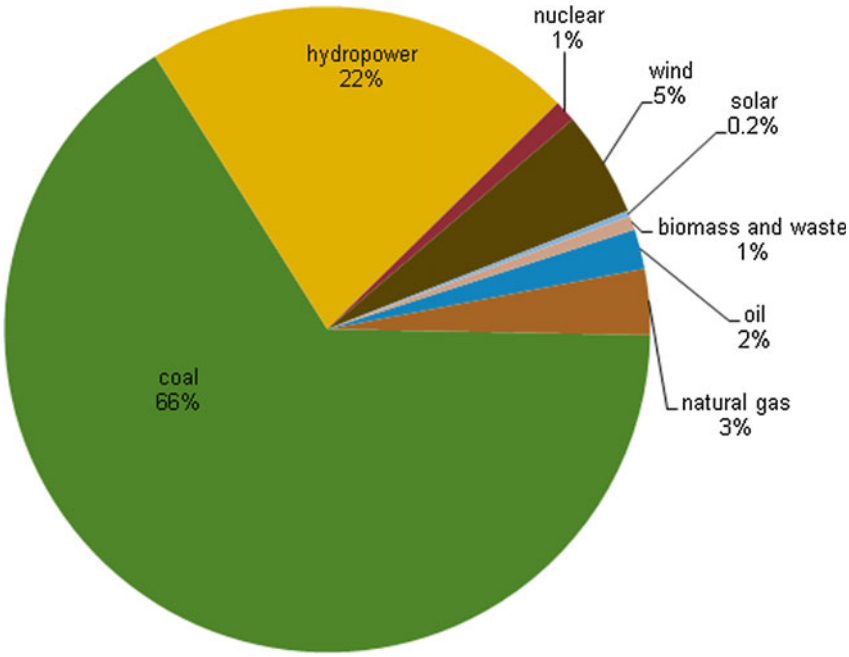

Fig. 1 China's installed electricity capacity by fuel in 2012 (installed capacity: 1,145 GW)

The Th–U fuel cycle has several advantages compared with the U–Pu fuel cycle. The thermal capture cross section of ^{232}Th (7.4 barns) is about three times higher than that of ^{238}U (2.7 barns) and that of ^{233}U (45.76 barns) is much smaller than ^{239}Pu (268 barns). This means that ^{233}U can also be bred in thermal reactors. In addition, the long-lived minor actinides (MA) resulting from fission are much lower in the Th–U fuel cycle compared with the U–Pu fuel cycle [4]. The fission gas release rate of Th-based fuel is smaller than that of U-based fuel, which allows Th-based reactors to be operated in deeper burnup [5–7]. There are also several challenges associated with the Th–U fuel cycle. The strong gamma irradiation of the daughter products of ^{232}U will cause difficulties in storage and transport. The intermediate nucleus ^{233}Pa in the Th–U conversion chain has a 27-day half-life [8], which will have a significant impact on reactor operation and fuel conversion.

Being one of the six candidates of Generation IV reactors, molten salt reactors (MSRs) have two main subclasses. The first subclass is known as a liquid-fueled MSR (MSR-LF) [9, 10], in which nuclear fuel is dissolved in the molten fluoride salt and the salt serves both as the fuel and the coolant in the primary loop. Dry reprocessing processes can be applied to an MSR-LF system so that fuel breeding can be achieved. Such a scenario is particularly suitable for the use of thorium fuel. The second subclass is the fluoride-salt-cooled high-temperature reactor (FHR) [11], also known as a solid-fueled MSR (MSR-SF). It uses solid fuel elements composed of TRISO particles inside a graphite substrate and molten fluoride salt as the coolant. This type of reactor can achieve excellent performance on safety and economy with a high temperature output. Recently, investigations on MSRs have drawn fresh attention around the globe.

Thorium-Based Molten Salt Reactor Nuclear Energy Systems

The MSR-LF and MSR-SF have interesting characteristics and applications, including thorium energy utilization, hydrogen production at high temperature, water-free cooling, and a small modular design. These properties make MSR one of the best approaches to solve the energy and environmental issues of China. In January 2011, the Chinese Academy of Sciences (CAS) launched the strategic pioneer science and technology project: thorium molten salt reactor nuclear energy systems (TMSR). The TMSR project will strive to realize effective thorium energy utilization and composite utilization of nuclear energy in 20–30 years (Fig. 2) [12].

Th utilization in MSRs can be realized step by step, depending on the fuel cycle modes and the related technology development. TMSR-SF can be operated in a once-through fuel cycle mode for simplicity, which means that the nuclear fuel is used only once. In principle, Th

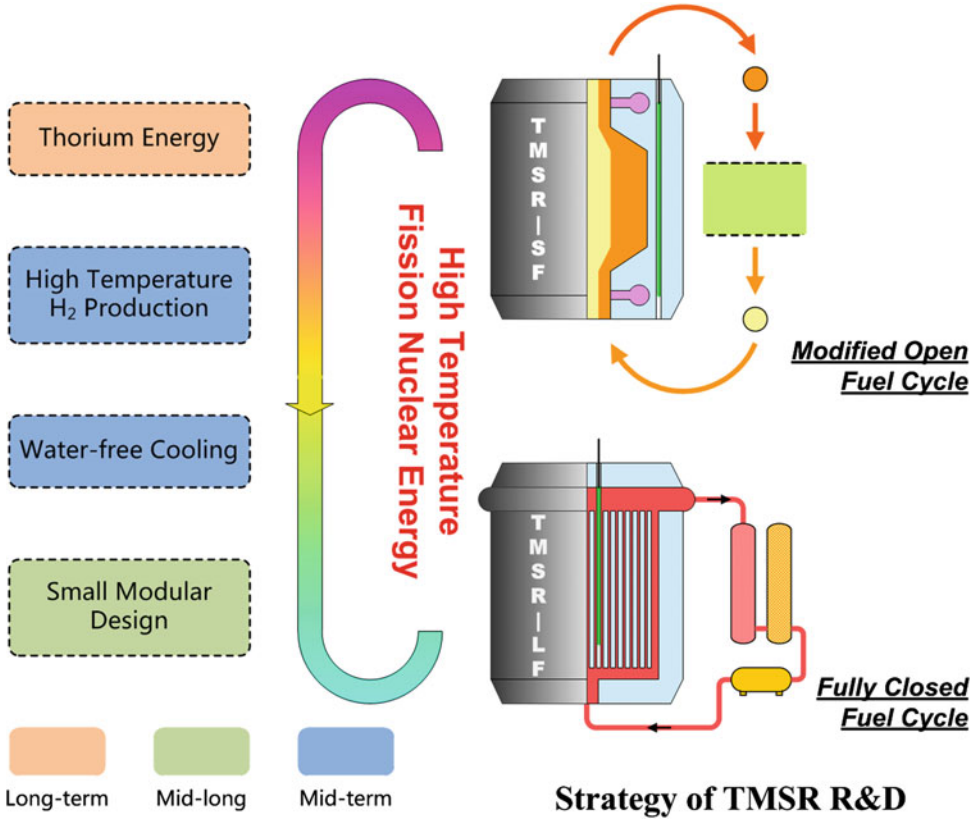

Fig. 2 Strategy of the TMSR R&D approach

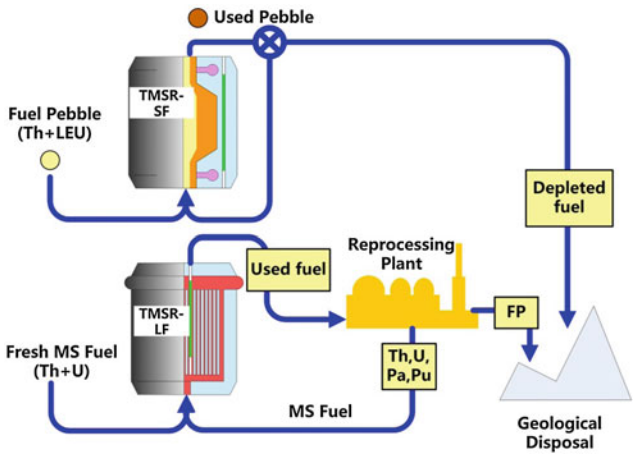

Fig. 3 Material flow of TMSRs with various fuel cycles

utilization can be realized in TMSR-LF with the modified open or even fully closed fuel cycle owing to its unique on-line chemical reprocessing technology (Fig. 3). After separation, fission products are geologically disposed directly and, simultaneously, ^{233}U, ^{232}Th, and minor actinides (MA) are recycled for further burning.

It should be noted that fuel utilization, waste generation, together with economical facts, non-proliferation, safety, and technology availability all play significant roles and should be taken into consideration in any comprehensive analysis of the various nuclear fuel cycles.

Research Activities

The TMSR project has already carried out research work and achieved some results in key technologies, including conceptual designs of experimental TMSRs, development of fuel reprocessing technologies, establishment of experimental platforms, and theoretical research, etc. Foreign cooperation has progressed steadily. The TMSR center has productively cooperated with the American Nuclear Society (ANS) in setting the safety standards for the TMSR-SF and with American Society of Mechanical Engineers (ASME) in setting the material processing standards for high-temperature reactors. Furthermore, the current TMSR plans have been strongly supported and approved by the National Nuclear Safety Administration (NNSA) and the Shanghai Committee municipal government.

Conceptual Design of TMSR Experimental Reactors

A set of methods and tools for the design and safety analysis of the experimental TMSRs have been constructed, initially based on mature commercial software. The accuracy of these computational methods has been preliminarily tested using both cross checks from different software and a few experimental data. In addition, some key experimental verification is also planned.

Based on the TMSR design platform built so far, the world's first 10 MW solid-fuel molten salt experimental reactor (TMSR-SF1) and the 2 MW liquid-fuel molten salt experimental reactor (TMSR-LF1) are beginning to be designed (Fig. 4). The goal of the TMSR-SF1 and TMSR-LF1 is to realize the integration, construction, operation, and maintenance of the TMSR system, to verify the physical behaviors, thermal-hydraulic and intrinsic safety characteristics, and to provide a comprehensive experimental platform for the design of future commercial reactors. Furthermore, the construction of TMSR-LF1 will also provide an experimental platform for pyro-process technologies and complete the preliminary validation of Th utilization.

The TMSRs are different from other kinds of reactors in their lack of ready-made products. The TMSR research team has successfully developed a high-temperature molten salt pump, molten salt air exchanger, molten salt frozen valve, and other key pieces of equipment. The constructed FLiNaK experimental loop was operated for more than 1000 h at 500–650 °C. The humidity and O_2 concentration in the loop were controlled to protect the system from corrosion and solve high-temperature seal problems. A series of tests have been finished, including an equipment performance test, loop technical identification, a heat transfer experiment, and a test of the compatibility of the salt with nickel alloy. In particular, the system design, construction, and operation of a high-temperature fluoride molten salt loop have been achieved.

Fuel Reprocessing Technologies

We have designed a process flowsheet for the TMSR fuel cycle based on pyro-processing techniques. The targets of this flowsheet are separating and recycling the most valuable UF_4 and carrier salts on-line by using pyro-processing techniques and separating ^{233}U (decay from ^{233}Pa) and Th from the residue after cooling for several months. The fluoride volatility method and low-pressure distillation are the crucial techniques in the above-mentioned flowsheet. Such a process not only reduces intensity and difficulty of on-site fuel processes, but also recycles precious ^{7}LiF in time to reduce the inventory on-site.

For the fluoride volatility technique, we have determined a pathway that includes IR spectroscopy to monitor the process, an adsorption method to purify the products, and gradient condensation to collect the volatilized UF_6. The equipment has been built and an on-line monitoring technique has also been developed. A series experiments have proven that UF_6 can be recovered using the fluoride volatility process from UF_4 powder or eutectic UF_4–FKZr, and the recovery ratio of U is over 95 %. To prevent the corrosion between molten fluoride salt and construction materials during the fluoride volatility process, we are planning to develop the "frozen wall" technique, which is extensively applied in the metallurgical industry (Fig. 5). This technique will effectively prolong the lifetimes of the construction materials. The preliminary results with nitride salt indicate that the frozen wall can be formed and maintained by controlling the heat exchange rate.

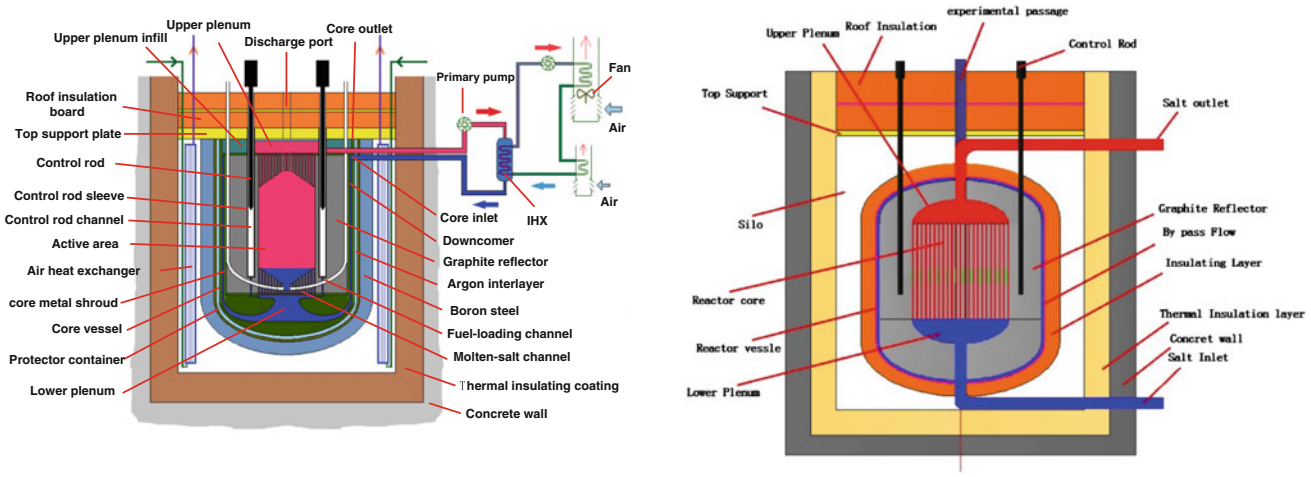

Fig. 4 Schematic structure of the TMSR-SF1 (*left*) and the TMSR-LF1 (*right*)

Fig. 5 Picture of frozen-wall facility and image of the formed frozen wall (*Inset*)

Fig. 6 Picture of the horizontal distillation facility

A gram-scale vacuum thermogravimetric system has been developed, and the distillation rate of FLiNaK and the relative volatility of a series of rare-earth fluorides have been determined experimentally using this system (Fig. 6). We have developed a 100 g-scale fully sealed distillation device in which the distillation, condensation, and collection of the molten salt are driven by the temperature gradient. The dependence of the collection efficiency on the temperature field was investigated in detail, and a recovery ratio of more than 98 % was obtained. Kilogram-scale distillation of FLiNaK was performed at a horizontal distillation facility with a large evaporation surface. The evaporation rate of FLiNaK reached 1 kg/h, and the collection efficiency was more than 94 %. The results suggest that the application of low-pressure distillation for purification and recovery of the carrier fluoride salts is feasible. Further work will focus on eutectic FLiBe, which is the real coolant or carrier salt for the TMSRs.

Structural Materials

The structural materials of TMSRs will be subject to the extreme environments, that is, high temperature, high neutron doses, and corrosive coolant. Particularly in the case of TMSR-LF, the fuel-dissolving fluoride salt in the core will produce a few radioactive or corrosive products (such as Xe, F, I, Cs) under neutron irradiation. Hence, the development

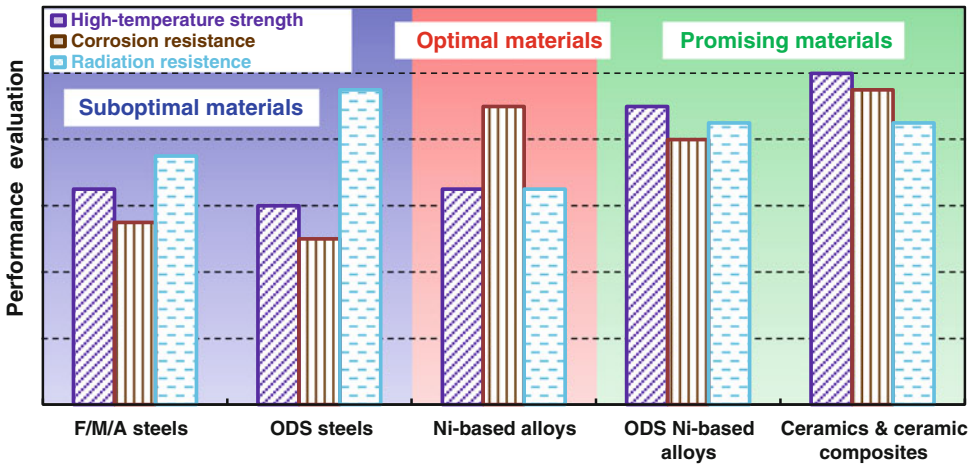

Fig. 7 Comprehensive performance evaluation of candidate structural materials

of the TMSRs much depends on the high-temperature structural materials. Several candidates have been widely discussed, such as ferritic/martensitic/austenitic (F/M/A) steels, oxide dispersion strengthened (ODS) steels, Ni-based alloys, including ODS Ni-based alloys, ceramics and ceramic composites [13]. Figure 7 presents their comprehensive performance evaluation for high-temperature strength, corrosion and radiation resistance. The Ni-based alloys are considered to be the primary option for metallic structural materials in TMSRs. The Hastelloy-N, a Ni–Mo–Cr based alloy developed at ORNL shows good strength at 923 K and good chemical compatibility with FLiBe salt [14].

A high-temperature Ni-based alloy (GH3535) has been developed in China, and its conventional performance parameters have reached those of Hastelloy-N alloy. Small-scale and pilot-scale production of GH3535 alloy has been completed. The technology for mass production has also been established and it ensures a sufficient supply of high-temperature molten salt corrosion-resistant alloys in China.

We have developed the processing technologies of Hastelloy-N alloy, for example, hot extrusion and rolling process technologies for large-caliber pipes (ϕ = 141.3 mm) of the reactor primary loop (Fig. 8). Several technological challenges in dealing with alloys with high molybdenum content have been solved. We have optimized the welding procedures where the manual tungsten inert gas (TIG) welding procedure was tentatively selected and validated.

Fig. 8 Hot extrusion of alloy pipes: **a** hot extrusion process, **b** ϕ = 141.3 mm pipe, **c** ϕ = 88.93 mm pipe

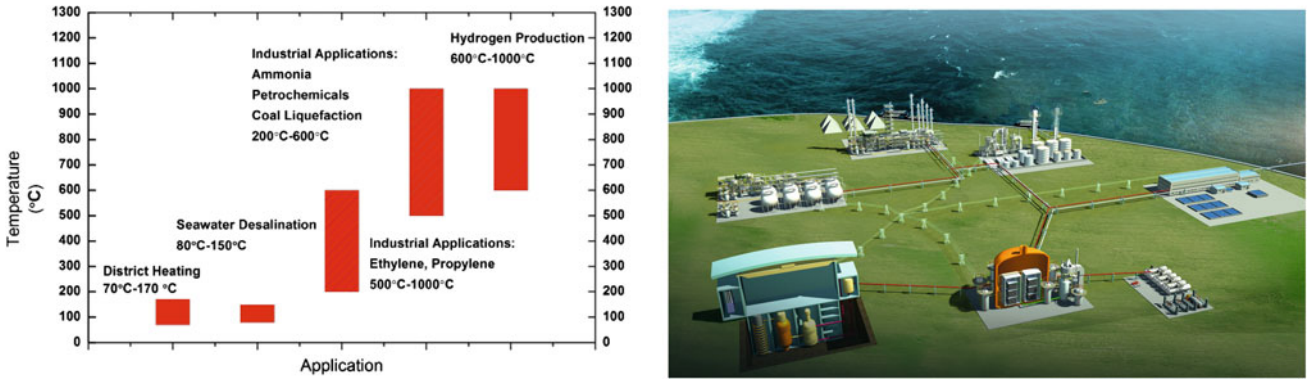

Fig. 9 *Left* Applications of nuclear heat in different temperature ranges. *Right* A sketch for a nuclear hybrid energy system based on TMSR

TMSR Hybrid Energy System

TMSRs are designed to provide very high temperatures (600–1000 °C). This heat can be used not only for high efficient electricity generation, but also for hydrogen production, industrial processes, and seawater desalination [15]. A hybrid energy system (HES) based on TMSR will improve the energy utilization efficiency, meet the clean energy demand, diminish the use of coal and oil, and diminish the emission of pollutants (Fig. 9) [16]. It is predicted that TMSRs can produce hydrogen much more efficiency (up to 50 %) by high-temperature stream electrolysis (HTSE) or thermochemical cycles. Moreover, molten salt coolants can provide a better solution for heat transfer from nuclear reactor to hydrogen production plant.

Compared with conventional low-temperature (<100 °C) electrolysis, HTSE increases the performance and the electricity-to-hydrogen efficiency by minimizing the Gibbs free energy of the reaction. We have integrated the first 1 kW solid oxide electrolyser cell (SOEC) hydrogen production system and reveal the influence of different parameters, such as current density, temperature, and steam concentration on the stack performance. The facility shown in Fig. 10 has run stably for more than 1000 h and the hydrogen production rate has reached more than 100 L/min.

Fig. 10 kW integrated SOEC evaluation platform

Summary and Outlook

The purpose of China's TMSR project is to achieve a sustained thorium-based nuclear system with high-temperature output, maximized thorium utilization, and minimized radiotoxicity of the spent nuclear fuel. Although there are still several challenges facing Th–U fuel cycle and MSR development, it is reasonable to expect that this TMSR project will shed light on the energy problem in China and help realize sustainable development in the long term.

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The Japanese Thorium Program

Toshinobu Sasa

Abstract

The Japanese nuclear energy policy is based on the uranium–plutonium cycle including application of a fast breeder reactor. In 2010, the Japan Atomic Energy Commission launched a council to construct a new framework of nuclear energy policy. Although progress has been halted after the Fukushima accident caused by the Great East Japan Earthquake, the application of thorium fuel is discussed as one of the alternative options for future nuclear fuel cycles and it is recommended to precede in basic research and developments. Two research working groups related to thorium-fueled reactors and the thorium fuel cycle were set up in the Atomic Energy Society of Japan in 2013. One is the "Research Committee on Nuclear Applications of Molten Salt". The committee was established to survey molten salt technology including molten-salt-cooled reactors, molten-salt-fueled reactors, and dry reprocessing processes. The committee planned to summarize the current state of the art and issues of molten-salt-fueled systems. The other is the "Working Group for Thorium Fuel Applications in Light Water Reactors and Fast Reactors". The objective of the working group is to summarize the current status and issues for application of solid-form thorium fuel in a U–Pu fuel cycle. Through these two different approaches, it is expected that the opinion on thorium fuel applications in Japanese nuclear fuel cycle policy will be summarized from an academic point of view. This paper summarizes the outline of activities for the two groups and introduces the application of accelerator-driven systems for thorium breeding.

Introduction

The Japanese nuclear energy policy is based on the uranium–plutonium fuel cycle for application in both light water reactors (LWR) and fast breeder reactors (FBR). This was decided at an early phase of nuclear power installation in Japan and research and development (R&D) activities for nuclear power generation was based on the national policy. In 2010, the Japan Atomic Energy Commission launched a council to construct a new framework of nuclear energy policy. Although this council has been halted after the accident at Fukushima-Daiichi Nuclear Power Plant caused by the Great East Japan Earthquake, the application of the thorium fuel cycle is discussed as one of the alternative options for future nuclear fuel cycles and it is recommended to enhance basic R&D activities in Japan.

Two research working groups related to thorium-fueled reactors and the thorium fuel cycle were set up in the Atomic Energy Society of Japan in the summer, 2013. One is the "Research Committee on Nuclear Applications of Molten Salt". The committee was established to survey molten salt technology, including molten-salt-cooled reactors, molten-salt-fueled reactors, and dry reprocessing processes. The committee planned to summarize the current state of the art and issues of molten-salt-fueled systems. The other is the "Working group for Thorium Fuel Application in Light Water Reactors and Fast Reactors". The objective of the working group is to summarize the current status and issues

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for application of solid-form thorium fuel in LWR/FBR fuel cycles.

Through these two different approaches, it is expected that the opinion on thorium fuel usage in Japanese nuclear fuel cycle policy will be summarized from an academic point of view. This paper summarizes the activities of these two research working groups and gives examples for thorium utilization research using accelerator-driven systems (ADS) for nuclear transmutation as studied at Japan Atomic Energy Agency (JAEA).

Research on Thorium Utilization in Japan

Activities for Molten Salt Applications

The committee was established in 2013 to discuss current molten salt technology, including molten-salt-cooled reactors, molten-salt-fueled reactors, accelerator-driven systems, fusion reactor blankets, and dry reprocessing processes. The committee consists of about thirty members from universities, research institutes, and industrial companies. The committee planned to summarize the current state of the art and issues of molten-salt-fueled systems. The committee also discussed the handling technologies for molten salt reactors especially those in China and the United Kingdom; issues of molten salt application to fusion reactors, dry reprocessing of spent nuclear fuel, and non-nuclear applications of molten salts.

Activities for Solid Thorium Fuel Applications

The "Working Group for Thorium Fuel Application in Light Water Reactors and Fast Reactors" was started before the Fukushima accident to enhance the basic R&D for thorium-loaded fuel applications. The objective of the working group is to summarize the status and issues for application of solid-form thorium fuel in LWR/FBR fuel cycles. The working group will summarize the current status of thorium-loaded fuel research. Physical and chemical properties of thorium oxide fuel, the effect of inert materials and/or fission products, and the application of thorium–plutonium mixed oxide (MOX) fuels were discussed. The status of thorium-loaded nuclear reactors from a neutronic point of view was also discussed. Nuclear data preparation for the thorium cycle, including cross sections, burnup chain, fission product yields, and delayed neutron emission fractions, were reviewed. As a survey of international activities, the working group reviewed the reports summarized by International Atomic Energy Agency (IAEA), OECD/Nuclear Energy Agency (NEA), and the US Nuclear Regulatory Committee.

Application of ADS to Thorium Breeding

JAEA performs R&D on ADS for transmutation of long-lived radioactive nuclides to reduce the environmental impact caused by radioactive waste disposal. Although the primary candidate of the ADS is a lead–bismuth eutectic alloy (LBE) target/cooled system using minor actinide (MA) nitride solid fuel [1], JAEA has also performed studies on liquid-fueled ADS as an alternative option for a primary candidate.

Figure 1 shows the conceptual view of a chloride-based molten salt ADS [2]. In this system, the molten salt acts as a spallation target, the fuel of the subcritical core, and primary coolant. For the requirement of the higher transuranic species (TRU) solubility and a harder neutron spectrum in the subcritical core, a chloride molten salt was selected as a fluid material. The fuel composition of the molten salt is 64NaCl–5$PuCl_3$–31$MACl_3$ or 60$PbCl_2$–60(Pu + MA)Cl_3. There are no specific differences for neutronic and burnup performances caused by the difference in the fuel composition. The operation temperature of the salt is set at 650–750 °C. Molten salt circulation pumps and heat exchangers are placed in a tank-type reactor vessel to reduce the in-core fuel inventory. Thermal power of 800 MW is generated by the injection of 1.5 GeV, 25 mA protons and 250 kg of actinides can be transmuted annually. Fuel composition is continuously controlled to extract reaction products and stabilize the core subcriticality through the on-line fuel processing circuits.

To evaluate the feasibility of the JAEA-proposed molten salt ADS for thorium cycle use, neutronics studies were performed [3]. In this evaluation, the dimensions of the system were not changed and only the material composition of the chloride fuel was changed to 60NaCl–34$ThCl_4$–6$^{233}UCl_4$. When the concentration of ^{233}U was set to a higher value, the melting point changed to a lower value than that of FLiBe fuel. The trends of the subcritical core

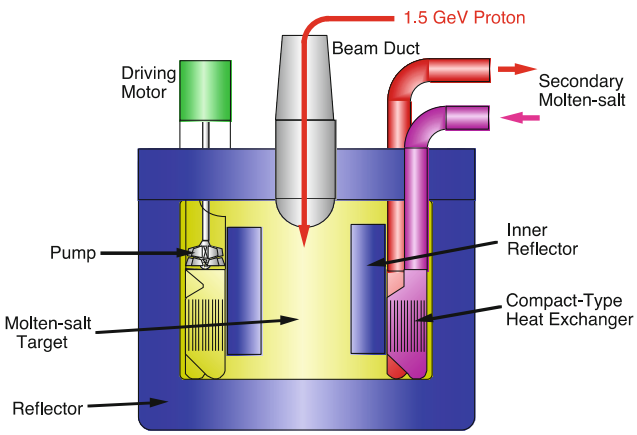

Fig. 1 Molten salt target/cooled ADS for MA transmutation

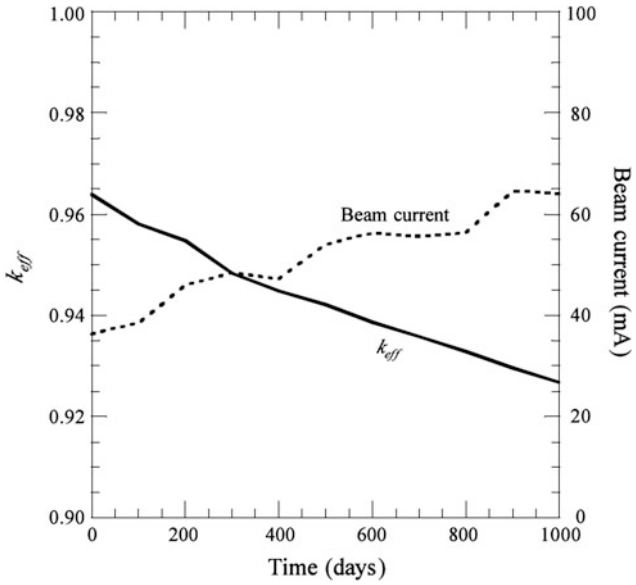

Fig. 2 Time evolution of k_{eff} and the operation beam current

multiplication factor, k_{eff}, and the required beam current for rated operation according to burnup without on-line fuel treatment are shown in Fig. 2.

Because the core of the ADS is kept in subcritical condition, this system is not sensitive to the accumulation of ^{233}Pa. The k_{eff} slightly decreases because of accumulation of fission products. The required beam current to generate 800 MW of fission power increases from 30–70 mA over 1000 days of operation. In this case, spallation neutron yields from the injection of 1.5 GeV protons are calculated to be 27 neutrons per incident proton. If a more efficient spallation target, such as liquid LBE, is used, the required beam current for rated operation can be decreased.

Design studies for a revised liquid fuel ADS with separated spallation target as shown in Fig. 3 is underway [4].

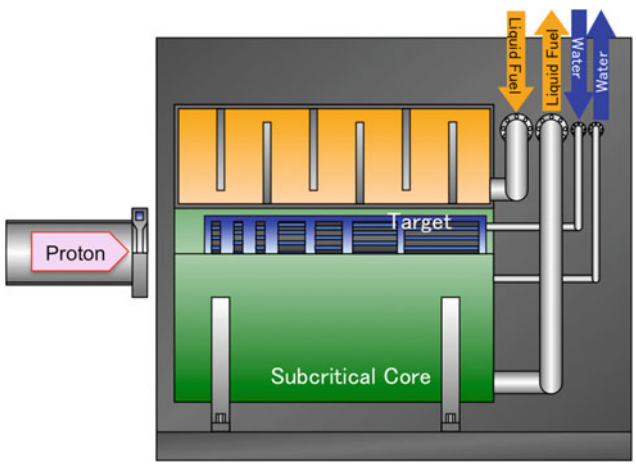

Fig. 3 Revised liquid-fueled ADS concept for MA transmutation

This system aims at extracting americium and curium from the current fuel cycle and transmute them into plutonium, which has better chemical stability and safety in MOX fuels. By adding these concepts to the current power generation cycle, fuels for power reactors can maintain a conventional fuel composition. It is useful to improve safety and cost efficiency of electric power generation. From the preliminary analysis results, there is a possibility to make the system a self-standing one to reduce the environmental impact of minor actinides. It is also found that this system, reflecting liquid-fueled ADS concepts, also gives fairly good transmutation performances. Adoption of on-line reprocessing by batch processing gives very small burnup reactivity change compared with the solid-fueled systems. The major parameters for the above-mentioned JAEA liquid-fueled ADS are summarized in Table 1.

National Review of Partitioning and Transmutation Technologies

To realize full ADS usage, there are many issues to be solved. JAEA proposes to establish a transmutation experimental facility (TEF), which is illustrated in Fig. 4, within the framework of the Japan Proton Accelerator Research Complex (J-PARC) project [5]. TEF consists of two buildings; the transmutation physics experimental facility (TEF-P) [6] and the ADS target test facility (TEF-T) [7]. The two facilities are connected by a proton beam line with a low-power beam extraction mechanism using a laser beam [8]. TEF-P is a facility with a zero-power critical assembly where a low-power proton beam is delivered to study the reactor physics and the controllability of ADS. TEF-T is planned as an irradiation test facility, which can accept a maximum 400 MeV, 250 kW proton beam into the LBE spallation target. Using these two facilities, the basic physical properties of subcritical systems and engineering tests of spallation targets will be studied.

In 2013, a national review working party for partitioning and transmutation (P-T) technologies using ADS was launched by the Ministry of Education, Culture, Sports, Science and Technology in Japan. The working party aims at reviewing the state of the art of P-T, the feasibility for construction of TEF, and a cost and benefit analysis of cooperation with European MYRRHA project. Five meetings were held during August to October 2013 and an interim report was issued in November 2013. In the interim report, the working party specified a roadmap to realize an ADS-based P-T fuel cycle. The working party also recommended to promote P-T technology as an important alternative option for future nuclear waste management and agrees that establishment of facilities to handle certain amount of MAs are required and the R&D to establish these

Table 1 Major parameters for JAEA liquid-fueled ADS

Molten salt ADS for MA transmutation	
Proton beam	1.5 GeV, 25 mA
k_{eff}	0.92
Thermal power	800 MW
Molten salt fuel	$64NaCl–5PuCl_3–31MACl_3$ or $60PbCl_2–60(Pu + MA)Cl_3$
MA loading	5000 kg
Fuel operation temperature	650 °C/750 °C
Fuel flow speed	3.6 m/s
Secondary coolant	$92NaBF_4–8NaF$
Liquid-fueled ADS for the U–Th cycle	
Proton beam	1.5 GeV, 36 mA
k_{eff}	0.964
Thermal power	1500 MW
Fuel	$60NaCl–34ThCl_4–6^{233}UCl_4$
MA loading	5000 kg
Initial ^{233}U loading	~12 t
Fuel melting temperature	370 °C
Fuel average temperature	640 °C
Revised liquid-fueled ADS for MA transmutation	
Proton beam	600 MeV, 5 MW
Initial k_{eff}	~0.98
Thermal power	320 MW
Fuel zone radius/height (cm)	18–38/~120
Fuel density (g/cm^3)	3.80
MA fraction (Am:Cm)	88:12
Fuel fraction $MACl_3$:NaCl	30:70
Target radius/height (cm)	12.0/30.0
Target material/coolant	W/water

Fig. 4 Transmutation experimental facility

facilities should be continued. As for the construction of J-PARC TEF, a step up from the current basic study stage to the next stage is noted as "suitable". It is also noted that a detailed technical review should be held before initiating construction. As for the participation with the MYRRHA project, it is proper to begin negotiations with Belgium with comprehensive cooperation not only by JAEA but also by universities and the commercial sector. The working party continues a review works of P-T technology, including the status of the TEF project and negotiations with the MYRRHA project.

In 2014, the working party held two meetings to review the progress of R&D, particularly for the activities related to TEF construction. Various activities to realize the LBE spallation target for TEF-T, the MA-loaded core for TEF-P, and other efforts to enhance R&D activities such as international collaboration and human resource development were discussed. After the meeting, the working party summarized that, the working party approved the R&D for TEF construction as "appropriate" and that it had progressed steadily. The working party also approved enhancing activities related to the TEF construction, such as a ground survey for candidate areas for the TEF building.

Summary

There were no specifications for thorium utilization in Japanese energy policy; thus, two research working groups, the "Research Committee on Nuclear Applications of Molten Salt" and the "Working Group for Thorium Fuel Application in Light Water Reactors and Fast Reactors" were set up in the Atomic Energy Society in Japan. Through these two different approaches, it is expected that the opinions for thorium fuel usage in Japanese nuclear fuel cycle policy will be summarized from an academic point of view.

Meanwhile, JAEA performs R&D for P-T technologies using ADS. Several kinds of liquid-fueled ADS are being studied as an alternative option for ADS. The transmutation performance of these liquid-fueled ADS is comparable with primary candidate LBE target/cooled ADS.

JAEA also proposes a transmutation experimental facility within the framework of the J-PARC project. A 250 kW LBE spallation target and MA-fueled critical assembly is planned to be installed. The construction of TEF is being discussed under the national review working party launched in 2013.

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Thorium Fuel Cycle Activities in IAEA

Uddharan Basak

The Nuclear Fuel Cycle and Materials Section of the International Atomic Energy Agency (IAEA) organizes technical meetings and symposia, facilitates technical cooperation, coordinates research projects, prepares state of the art technical documents and maintains and updates databases on nuclear fuels and fuel cycles. The IAEA conducts a programme on "Advanced Nuclear Fuels and Fuel Cycle Options", in particular, on the thorium fuel cycle, for which the main incentives and challenges are well known.

Thorium-based fuels have been studied for their potential applications in almost all types of reactors including water reactors (WRs), boiling water reactors (BWRs), pressurized heavy water reactors (PHWRs), high-temperature reactors (HTRs), fast breeder reactors (FBRs) and molten salt reactors (MSR), though on a small scale compared with U/U–Pu fuels. The motivation for developing thorium technologies are: utilization of the large energy potential of thorium resources, transuranic actinide (Pu and/or minor actinides (MAs)) consumption/management and a proliferation resistant fuel cycle. Sustainable development of nuclear energy is possible through the deployment of the thorium fuel cycle.

During the period 2011–2013, IAEA technical meetings were held on world thorium resources, on fuel integrity during normal operating and accidental conditions in PHWR and on advanced fuel cycles in PHWRs.

During the period 2012–2015, Coordinated Research Activities (CRP) have progressed on "near-term and promising long-term options for deployment of thorium-based nuclear energy". Participating Member States are Canada, China, Czech Republic, Germany, Italy, Israel, India, Russian Federation, UK and USA. Preliminary neutronic analysis shows that thorium enables sustained transuranics burning in all reactor systems analyzed: reduced-moderation pressurized water reactors (RMPWRs), reduced-moderation boiling water reactors (RMBWRs) and fast reactors (FRs). Furthermore, an investigation indicates that using the Th/U-233 fuel cycle in HTR-PM (pebble module) is feasible and realistic. A new CRP was launched on the "reliability of high-power, extended burnup and advanced PHWR fuels (2013–2017)". IAEA publications on the thorium fuel cycle and documents on world thorium resources can be found at http://infcis.iaea.org.

In summary, the use of thorium-based fuels was demonstrated in almost all types of reactors during the period between the 1960s and the mid-1980s. Large resources of thorium, along with favorable waste management and intrinsic proliferation characteristics of the thorium–uranium fuel cycle, instill the potential for the sustainable development of nuclear energy. Technological challenges remain for remote and automated processes even for the front end of the thorium–uranium fuel cycle in well-shielded facilities. Global attitudes towards deployment of the thorium fuel cycle are fast changing.

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Overview of European Experience with Thorium Fuels

Didier Haas, M. Hugon, and M. Verwerft

Abstract

Since the early 1970s, studies and experimental projects have been undertaken in Europe to examine the potential of thorium-based fuels in a variety of reactor types. The first trials were mainly devoted to the use of thorium in high temperature reactors (HTR). These projects can be seen as scientific successes, but were not pursued on a commercial basis, owing to the priority given in Europe to the development of light water reactors (LWR). Since then, thorium oxide has been considered as a potential matrix for burning plutonium (possibly also minor actinides), and several core design studies, as well as experiments, were undertaken. The most recent ones concern the BR2 and HFR material test reactor (MTR) irradiations in Belgium and the Netherlands, respectively, as well as the KWO pressurized water reactor (PWR) in Obrigheim in Germany, in which thorium–plutonium oxide fuel (Th–MOX) was successfully irradiated up to 38 GWd/tHM. The results of these experiments have shown that Th–MOX behaves in a comparable way to conventional uranium–plutonium oxide fuel (U–MOX). More work is still needed before Th–MOX will reach sufficient maturity to implement it on a large scale in power reactors, but all currently available results indicate that licensing Th–MOX for LWRs should be feasible. Finally, European research projects are still devoted to the study of thorium salts in molten salt reactors (MSRs), a design that incorporates on-line reprocessing and needs no specific thorium fabrication, adding, therefore, the benefits of thorium without its main challenges.

Keywords

Thorium • Fuel • Light water reactor • High temperature reactors • Molten salt reactor • Plutonium

Introduction

Natural thorium (Th) has only one isotope, ^{232}Th, which is fertile. In a thermal reactor, Th can absorb neutrons and the subsequent nuclear reactions produce ^{233}U, which is fissile. Under optimized breeding conditions, a sustainable Th–^{233}U cycle can be reached, but the thorium cycle needs a seed or driver fuel, which can be based on ^{235}U or on Pu.

^{233}U as a fissile nuclide features high neutron production in thermal and epithermal neutron spectra. This offers improved neutron economy for reactors fueled with ^{233}U rather than ^{235}U or ^{239}Pu, particularly at thermal energies in light water reactors (LWRs). In theory, breeding (formation of fissile nuclides) is achievable at thermal energies with a Th–^{233}U fuel, which is not the case with a U–MOX fuel. However, even though breeding can be demonstrated at the experimental level, optimal breeding has not been achieved in the current fleet of LWRs. In today's context, U–MOX fuels are not reprocessed and here, Th–MOX offers perhaps

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its best advantage over U–MOX. The excellent chemical stability of the thorium oxide matrix makes it an excellent candidate for direct disposal, and thus also for once-through fuels that allow the burning of excess Pu without production of higher actinides. Another alternative would be to use Th–MOX fuels in LWRs as a means to initiate the breeding of ^{233}U for future use in other reactor types; this presents an option to save natural U and/or to further improve the U–Pu fuel cycle.

Besides the LWR/fast reactor scenario, two reactor types have been considered for a breeding Th fuel cycle in the future: high temperature reactors (HTRs) and molten salt reactors (MSRs). HTRs represent the fastest route to implement a closed breeding Th fuel cycle. The technology exists conceptually, but needs to be developed before commercialization (which is pending). Also, supporting technologies associated with fuel manufacturing, reprocessing, transport, waste management, and final disposal need to be developed. MSRs represent a longer-term development option for Th fuel cycles. In MSRs loaded with Th-based fuels, breeding may be achieved over a wide range of neutron energies. Online reprocessing is an important feature of MSRs, which enables continuous re-use of the nuclear fuel by extracting the fission products.

The potential development of a closed Th fuel cycle faces some obstacles. Reprocessing is one of these as Th oxide is more stable that U oxide. Contrary to the Purex process, which has been industrially operational in the U–Pu fuel cycle for more than 30 years, the Thorex process, which has been investigated for many years in laboratories, faces some difficulties: it requires stronger acids (and therefore more advanced corrosion-free materials for process vessels) and longer dissolution times. Remote-controlled fuel manufacturing represents another challenge, as Th-based fuels have high-energy gamma radiation owing to the presence of ^{232}U after irradiation. This will require remote fabrication and handling in heavily shielded facilities. This makes fuel fabrication, transport, and reprocessing more complex than the present practice for U oxide fuel, for instance.

This presentation summarizes the history and status of the main European research programs (www.cordis.europa.eu) with Th use. These programs concern HTRs, LWRs, and MSRs. Emphasis is given here on the last two developments.

European Thorium Research Program History

During the early years of nuclear energy R&D in Europe, between 1960 and 1980, the main experimental projects involving Th fuels were related to HTRs (DRAGON, OECD international project in the UK, and the ATR and THTR reactors in Germany) and also to an irradiation of Th–MOX fuel in the Lingen BWR in Germany. These projects can be seen as scientific successes, but were not pursued on a commercial basis, owing to the priority given in Europe to the development of LWRs (except in the UK where low temperature gas-cooled reactors were developed) with UO_2 as a reference fuel, and, for countries having selected the reprocessing cycle strategy, the recycling of recovered Pu as a mixed oxide (MOX) fuel.

Since then, several studies have been undertaken to examine the worldwide interest in Th. In 1997, M. Lung wrote a report entitled “A present review of the thorium fuel cycle” [1] at the request of the European Commission. Then in the 4th EURATOM Framework Program, a review of the benefits of the Th cycle as a waste management option was carried out [2].

As a result of these studies, it was recognized that this option presented major advantages in terms of actinides management through the “burning” of excess Pu in a non-U matrix (Th oxide), at least for those countries in Europe that considered Pu as a waste and not as a source of energy for future utilization in fast reactors (FRs). These assessments opened the door to several European irradiation experiments during the 5th EURATOM Framework Program using Th–MOX, namely in the KWO pressurized water reactor (PWR) in Obrigheim (Germany), in the HFR material test reactor (MTR) in the Netherlands (operated by NRG), and in the BR2 MTR in Mol (SCK•CEN) (“THORIUM CYCLE [3]” and “OMICO [4]” projects). These efforts were pursued and completed within the 6th EURATOM Framework Program, with the demonstration at the laboratory scale that this fuel would behave in a comparable way to current MOX fuels (see below). In the 6th EURATOM Framework Program, the fuels irradiated in the programs THORIUM CYCLE and OMICO were further investigated (post-irradiation examination, radiochemical analysis, and leaching tests) in the “LWR-DEPUTY” project [5] and a strategy study on the “Impact of Partitioning, Transmutation and Waste Reduction Technologies on the Final Nuclear Waste Disposal” (“RED-IMPACT”) was performed [6].

In parallel, efforts at European level started in the early 2000 and are still under way, concerning the development of the MSR, using a Th–^{233}U cycle in liquid Th fluoride fuel. Between the 5th and the 7th EURATOM Framework Programs, several projects (MOST, ALISIA, EVOL) were funded (see below).

Within the European nuclear research community, a Technology Platform named SNETP (Sustainable Nuclear Energy Technology Platform: www.snetp.eu) gathers most of the stakeholders involved in reactor research. SNETP issued a “Strategic Research Agenda” in May 2009 (revised in 2013, following the Fukushima accident) with an Annex (in January 2011) devoted to Th. In the annex, Th systems are noted as having significant long-term potentialities, but

also significant challenges before reaching industrial implementation. The two aspects (Pu management, molten salts) mentioned in this paper were specifically recognized in the Th Annex to the Strategic Research Agenda.

Th–MOX Fuels Irradiated Under LWR Conditions

Within the European Framework Programs, the study of Th fuels behavior in LWRs was primarily aimed at comparing the behavior and the applicability of various matrices to be used for the transmutation of Pu and minor actinides (projects THORIUM CYCLE, LWR-DEPUTY, OMICO). Comparisons were made with standard fuels (UO_2, MOX), and also with so-called "inert matrices" fuels (using, for example, Mo or MgO as a matrix in, respectively, CERMET and CERCER fuel types). As explained earlier, irradiation experiments were performed in three facilities, namely the KWO PWR, HFR, and BR2 material test reactors.

The THORIUM CYCLE project was a four-year project with the following participants: the coordinator NRG (NL), BNFL (UK), CEA (F), FZK and KWO (D), JRC-IE and JRC-ITU (EU). It started on October 1st, 2000. The goals of this project were to supply key data for application of the Th–cycle in LWRs. In particular, it included the study of:

- the behavior of Th-based fuels at extended burnup through an irradiation experiment offour4 short fuel pins (UO_2, (U, Pu)O_2, ThO_2, and (Th, Pu)O_2) up to 55 GWd/tHM in HFR, and an irradiation experiment of one short fuel pin ((Th, Pu)O_2) to 38 GWd/tHM in a PWR (KWO). It should be noted that a previous irradiation of (Th, Pu)O_2 in Germany (Lingen) achieved a burnup of 20 GWd/tHM [7];
- the core calculations for Th-based fuels, including code-to-code validation, sensitivity checks for the significant isotopes ^{232}Th and ^{233}U, and the calculation up to 80–100 GWd/tHM for Th–MOX fuel.

The irradiation test in KWO enabled the investigation of the operational safety of Th–MOX rod behavior under realistic PWR conditions. The short test rod was inserted into a MOX assembly to provide the most realistic boundary conditions possible. The MOX carrier assembly had been irradiated for one cycle already. The cladding appeared in good condition after irradiation and its creep down, measured at the reactor site during the shut-down periods, as well as its general behavior were well within the bounds of experience for UO_2 fuels. The fission gas (Xe and Kr) release was about 0.5 % [8], which is about half that for equivalent MOX fuels at the same burnup, but the linear power was lower than in equivalent U–MOX studies. Taking into account experimental uncertainties, the fuel behavior seems to be at least as good as U–MOX.

The THORIUM CYCLE project was completed in 2006, but the post-irradiation experiments were performed under a subsequent experiment called LWR-DEPUTY (coordinator SCK•CEN). In this program, the main tests on Th–MOX consisted of additional fuels studies (microscopy, radial distributions of elements and isotopes), and radiochemical analyses. The objective of these analyses was to obtain a reliable experimental database for burnup analysis and to evaluate changes in the heavy nuclide content in order to:

- optimize the dissolution and analysis strategies;
- establish the first dataset on heavy nuclide and fission product content in irradiated Th–MOX to assess the overall uncertainties;
- use this dataset in a benchmark analysis program.

The OMICO Project [4] was conducted from 2001–2007. Its scope included the study and modelling of the influence of microstructure and matrix composition on Th–MOX fuel in-pile behavior under normal PWR conditions. The following tasks were undertaken:

- fabrication of the Th–MOX fuels at the JRC-ITU;
- irradiation in the "CALLISTO" PWR loop in BR2, representing real PWR conditions; the burnup achieved at the end of this project was about 13 GWd/tHM.
- non-destructive examinations (gamma-spectrometry, visual examinations) and microstructure studies.

It should be noted that the pins were instrumented for pressure and fuel temperature determination. The test matrix was such that the Th–MOX could be compared with U–MOX and UO_2 fuels. Another test parameter consisted of the fabrication process (homogeneous versus heterogeneous powder mixtures). The results of the temperature/pressure readings were primarily used to benchmark computer code models for behavior of Th–MOX fuels in the first stage of their lives.

In addition to the irradiation, fuel characterization was performed, including thermal diffusivity measurements, the results of which were published in references [4, 9]. The results demonstrate a higher thermal conductivity for (non-irradiated) Th–MOX compared with U–MOX.

In the LWR-DEPUTY [5] project, selected samples of the OMICO and THORIUM CYCLE programs were extensively studied to provide experimental datasets suitable for evaluating their in-pile performance. The experimental data were the basis of a benchmark exercise on the Th–MOX fuel pin irradiated at the NPP KWO to investigate the

qualification of the numerical tools and software packages. A study to determine the scope of the leaching behavior was also conducted. In addition to the experimental work, steady-state and transient analyses were performed for different PWR designs fueled completely or partially with Th–MOX fuel. An assessment of steady-state parameters (reactivity, shutdown margin, and reactivity feedback coefficients) has been performed in comparison with UO_2. All feedback coefficients are favorable for safe operation under steady-state conditions. A comparative analysis of control rod ejection scenarios has also been performed, and it was found that the maximum values obtained for fuel and clad temperature and maximum fuel enthalpy are in line with the acceptance criteria for the current generation PWRs.

After 10 years of research sponsored through the EURATOM programs, the following conclusions can be drawn regarding the behavior of Th–MOX fuel under LWR conditions:

- Th–MOX has great potential and its fabrication as an oxide fuel is feasible;
- even with a laboratory-scale production route, Th–MOX shows good in-pile performance;
- know-how on Th–MOX has increased, but:
- fuel performance clearly needs to be further improved before code calculations can predict the specific Th–MOX behavior.

As a general conclusion, the results of these experiments have shown that Th–MOX behaves in a comparable way (even better on some aspects) than MOX, and that licensing Th–MOX in a LWR should not be problematic, although more experimental data on fuels representative of the future commercial ones would be needed. Experimental data also demonstrate that Th fuels will be more resistant to corrosion than U fuels in the case of spent fuel geological disposal.

The Molten Salt Reactor

The MSR, which incorporates the reprocessing on-line and needs no specific Th fabrication, adds the benefits of Th without its main challenges. In particular, breeding may be achieved over a wide range of neutron energies, which is not the case for the U–Pu cycle.

Under the European Framework Programs, conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts opens promising possibilities to exploit the ^{232}Th–^{233}U cycle. In addition, they can also contribute to significantly diminish the radiotoxic inventory from present reactors' spent fuels, in particular, by lowering the masses of transuranic elements. Finally, if required as a result of expansion of nuclear electricity generation, breeding beyond iso-generation could be achieved. With the Th–U cycle, doubling times values are only slightly higher than those predicted for solid-fuel fast reactors working in the U/Pu cycle (in the range 40–60 years). The characteristics of different launching modes of the MSFR with a thorium fuel cycle have been studied, in terms of safety, proliferation, breeding, and deployment capacities of these reactor configurations [10].

Between Framework Programs 5 and 7, several projects (MOST, ALISIA, EVOL) were conducted, and promising developments and results were obtained in particular in following areas:

- conceptual design studies;
- safety developments, in particular, the study of residual heat extraction; tests with liquid salts have been undertaken to prove the ability of the cold plug system to play the role of a security valve on the loop circuit;
- fabrication of the salt mixture (LiF–NaF–KF) to be used in the French molten salt loop (FFFER project) has been achieved;
- experimental investigation of physicochemical properties of fluoride salts;
- experimental tests of the metallic-phase extraction process;
- corrosion studies and experiments (this remains one of the main challenges for the development of the reactor system).

Finally, it should be noted that the MSR with its Th cycle is one of the six reference systems selected for R&D collaboration in the framework of the Generation IV International forum. The main contributors are the European partners, supported by Russia as an observer.

Conclusion

Since the early 1970s, studies and experimental projects have been undertaken in Europe to examine the potential of Th-based fuels in a variety of reactor types. These projects have all been successful from a scientific point of view, but not all were followed-up in relation to the overall development of the nuclear industry in Europe. HTRs, although very well suited for Th use, have not been deployed to the benefit of LWRs. Results on the use of Th matrices in Th–MOX fuels in LWRs are encouraging, but still need demonstration on a larger scale in commercial conditions. Finally, the probably most efficient use of Th would be in a salt, to feed

MSRs. Conceptual studies and related experimental programs are underway.

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Overview of the Thorium Programme in India

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Abstract

The use of thorium is necessary from a long-term objective of sustainability of nuclear energy resources in India. Its use requires reprocessing to separate the fissile ^{233}U for sustained operation using ^{232}Th–^{233}U. Over the years, owing to sustained technology developmental initiatives, India has gained significant experience in all aspects of the thorium fuel cycle. In addition to briefly explaining advantages and challenges of thorium fuel cycle, this paper covers the Indian experiences in this direction. It also briefly describes the designs of Indian reactors based on thorium fuel cycle, for example, the Advanced Heavy Water Reactor (AHWR), as well as conceptual designs of proposed future reactor systems.

Introduction

The third stage of the Indian nuclear power programme envisages use of ^{233}U-fuelled reactors with thorium as the fertile material. The ^{233}U required for this purpose would be obtained from operation of Pu/Th-based fast reactors in the later part of the second stage of the Indian nuclear power programme. Although the vision for extensively using thorium-fuelled reactors in the third stage was based on significant thorium reserves in India, thorium as a nuclear material also has significant advantages in its own right. In view of its advantages, India is developing technologies for thorium-based reactors in many configurations, from light water cooled designs to high temperature liquid metal and molten salt cooled options. One of these options is the Advanced Heavy Water Reactor (AHWR), which has a number of passive safety systems that facilitate achieving a core damage frequency that is one order of magnitude lower than the value for current light water reactors (LWRs). A high level of safety thus addresses the concerns of large-scale deployment. The high temperature options hold promise for producing hydrogen as an alternate energy carrier for transport applications, thus ensuring long-term energy security. Another option, which is being studied, is the molten salt-fuelled reactor, which would be self-sustaining thorium based reactor. The Indian Molten Salt Breeder Reactor (IMSBR) can be configured to give significant breeding ratios.

Advantages of Thorium

Thorium, being a fertile material, cannot directly fission and needs to be converted into fissile ^{233}U. The other important fertile material occurring naturally is ^{238}U, which is a major part of natural uranium. The physics parameter, which is a measure of the conversion of fertile to fissile isotopes, is the neutron capture cross section. In the thermal spectrum, the value for this measure in the case of ^{232}Th is typically 2.47 times that for ^{238}U (Fig. 1a). Thus, thorium offers greater competition for neutron capture and lower losses to structural and other parasitic materials, leading to an improvement in conversion of ^{232}Th to ^{233}U [1].

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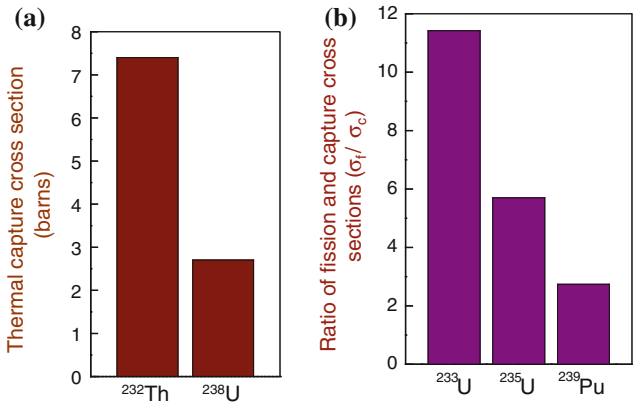

Fig. 1 **a** Comparison of thermal capture cross sections for thorium and ^{238}U; **b** fission to capture cross section ratio of various fissile isotopes

Fissile isotopes (^{233}U formed from ^{232}Th, ^{239}Pu formed from ^{238}U, and ^{235}U occurring naturally) can fission by absorption of incident neutrons. However, not all neutrons absorbed by a fissile nucleus can cause fission. The parameter that defines this wasteful neutron absorption is the capture cross section for the fissile isotope. Another parameter, the fission cross section, defines the useful absorption that causes fission. The capture cross section of ^{233}U is much smaller than for ^{235}U and ^{239}Pu, whereas the fission cross section is of the same order; this implies lower non-fission absorption, leading to higher isotopes (Fig. 1b). This favours the feasibility of multiple recycling of ^{233}U compared with plutonium.

Not only is the capture cross section of ^{233}U lower than other fissile isotopes, but ^{233}U also releases more neutrons per thermal neutron absorbed (η value) over a wider range of incident neutron energies, that is, in both the thermal and epithermal regions (Fig. 2), unlike ^{235}U and ^{239}Pu. This facilitates high conversion ratios with thorium utilisation in reactors operating in the thermal/epithermal spectrum. In addition, it can be shown that for higher discharge burnup (and hence the extraction of more energy from a given mass of fuel), the amount of initial fissile material needed is less [2] in case of thorium than for ^{238}U (Fig. 3). Also, by virtue of being lower in the periodic table than uranium, the long-lived minor actinides resulting from burnup are produced in much lower quantities with the thorium fuel cycle. Further, the thermal conductivity of ThO_2 is higher than that of UO_2 (Fig. 4), and the coefficient of thermal expansion of ThO_2 is less than that of UO_2, resulting in lower fuel temperature and less strain on the clad, which in turn favours higher burnups. Long-lived minor actinides resulting from the burnup chain are produced in much lower quantities with thorium fuel cycles, especially if the reactor operates purely in the ^{233}U–Th cycle. Actinides having masses beyond 237 are produced in negligible quantities. This is an important advantage, as the burden for management of long-lived radioactive waste is significantly reduced. Also, ThO_2 is relatively stable compared with UO_2 and retains fission gases within the fuel matrix. The fission gas release rate for ThO_2 is one order of magnitude lower than that of UO_2. In addition, it has good radiation resistance, dimensional stability, and is more tolerant to clad failures than UO_2-based fuels. These are important attributes during fuel failures. A comparison of failed UO_2- and ThO_2-based fuels is shown in Fig. 5.

Challenges of the Thorium Fuel Cycle

An important isotope of some concern in the thorium fuel cycle is ^{232}U. It is formed through (n, 2n) reactions from ^{233}Pa, and ^{233}U. The half-life of ^{232}U is about 69 years. The daughter products of ^{232}U are hard gamma emitters such as ^{208}Tl (2.6 MeV) with very small half-lives. As a result, the radioactivity increases with time for bred uranium isotopes.

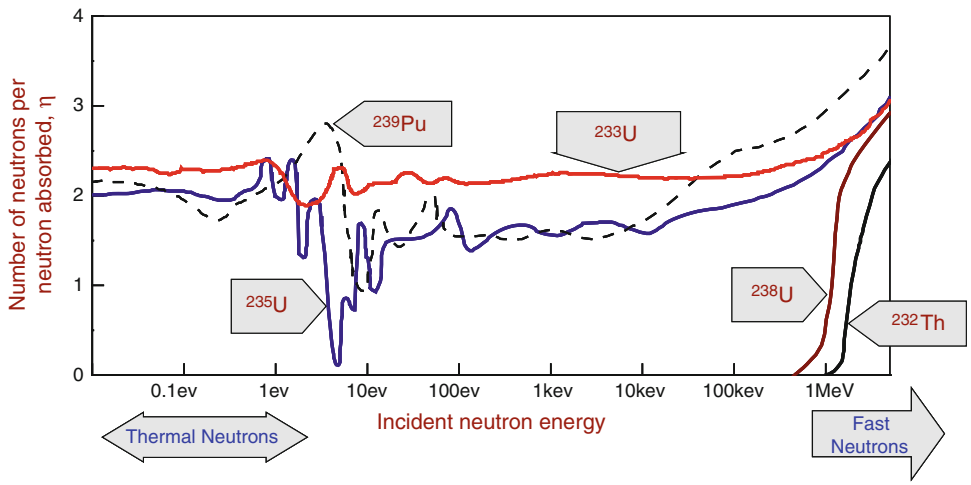

Fig. 2 Variation of η value for various fissile isotopes. ^{233}U shows a high value of η over a wide range of incident neutron energies

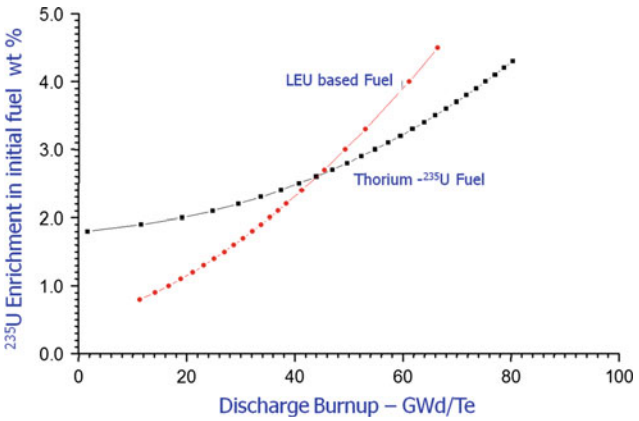

Fig. 3 Performance potential versus fissile topping in the Pressurised Heavy Water Reactor (PHWR)

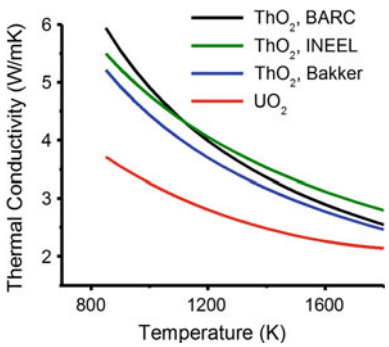

Fig. 4 Thermal conductivity of ThO_2 compared with UO_2, showing the superior performance potential of ThO_2

This feature makes thorium-based fuels inherently proliferation resistant. This also presents several technological challenges in the reprocessing and recycling of bred ^{233}U [1]. India is developing a laser-based separation technique, which can be used to clean ^{233}U by separating out the ^{232}U. The back end of the fuel cycle has special challenges owing to the inert nature of thoria and the major challenge is to make it dissolve during the spent fuel reprocessing operations.

Thorium Utilisation in India [3]

India has developed expertise in all aspects of thorium utilisation, starting from mining, metal extraction, fuel fabrication, irradiation in reactors, reprocessing, fuel fabrication using the recovered ^{233}U, and using it in reactors.

Thorium Irradiation in Research Reactors

(a) CIRUS reactor: Thoria rods (J-rods) have been continuously irradiated in the CIRUS reactor and ^{233}U has been accumulated. These rods have then been reprocessed. These J-rods have approximately 2–3 ppm of ^{232}U in uranium (being irradiated in the low flux reflector region).

(b) PURNIMA-II (1984–1986): India initiated thorium studies as early as the seventies and the first research reactor built with ^{233}U fuel generated in J-rods in the CIRUS reactor was PURNIMA-II. Experiments were performed at the PURNIMA-II facility with uranyl nitrate solution containing ^{233}U with BeO blocks as reflectors. Variations of the critical mass with different $H/^{233}U$ ratios were measured. Measurements of various reactivity parameters and neutron lifetimes were also performed. These experiments also gave confidence in handling ^{233}U.

PURNIMA-III (1990–1993) experiments with ^{233}U–Al dispersion fuel: Experiments were performed with ^{233}U–Al dispersion fuel in the form of plates in PURNIMA-III. BeO canned with zircaloy was used as reflector. Void coefficients and temperature coefficients of reactivity were also measured. These measurements helped in finalising the core of the KAMINI reactor. The data also helped in validating the nuclear cross section data of ^{233}U and Be.

(d) KAMINI reactor: A research reactor based on ^{233}U fuel in the form of U–Al alloy was commissioned at the Indira Gandhi Centre for Atomic Research (IGCAR) in

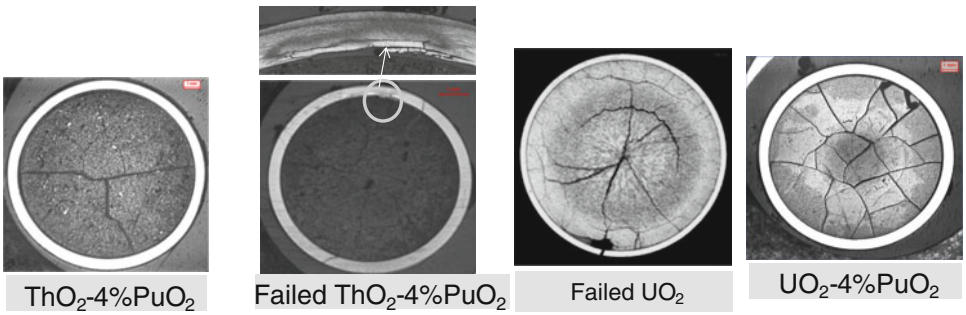

Fig. 5 Comparison of failed ThO_2–4 % PuO_2 (18,500 MWd/tonne), UO_2 (400 MWd/tonne), and UO_2–4 % PuO_2 (16,000 MWd/tonne) fuels at indicated burnups

Kalpakkam in 1996. Currently, it is the only operating reactor in the world with ^{233}U as fuel. The reactor power is 30 kW and it has a very high flux-to-power ratio. It has three beam holes used for activation analysis and a facility for neutron radiography of the Fast Breeder Test Reactor (FBTR) fuel.

(e) Irradiations in the pressurised water loop of the CIRUS Reactor: Advanced fuels using thorium were irradiated in dedicated engineering loops in CIRUS in a power reactor environment. The experimental fuel cluster BC-8 was irradiated to a nominal burnup of 10,000 MWd/tonne (HM). This cluster consisted of two pins each of ThO_2–6.75 % PuO_2, UO_2–3 % PuO_2, and UO_2 fuel and six pins containing ThO_2 fuel. Collapsible Zircaloy-2 cladding was used for these fuels. Non-destructive, post-irradiation examinations such as visual examination, diameter measurements, leak tests, ultrasonic tests, eddy current tests, gamma scanning, and gamma spectrometry have been completed on these fuel elements.

(f) Thoria bundles irradiated in the blanket zone of FBTR: 54 thoria subassemblies (717 kg) have been loaded into the 9th ring of the FBTR core as a radial blanket. The ^{233}U produced from this will have low content ($\sim$5 ppm) of ^{232}U.

(g) ^{233}U-based Prototype Fast Breeder Reactor (PFBR) fuel irradiation testing in FBTR: A 37 fuel pin PFBR type MOX experimental subassembly has been fabricated and has undergone irradiation test in the FBTR. The composition of fuel is (71 % UO_2 + 29 % PuO_2) with UO_2 containing 53.5 % ^{233}U. The testing was carried out for peak rating of 450 W/cm and to a burn up of 112 GWd/tonne.

Thorium in Power Reactors

Flux Flattening in Pressurised Heavy Water Reactors (PHWR) Using Thorium

One major step in thorium utilisation in India was achieving initial flux flattening with thoria bundles. The maximum power achievable in the fresh core of 220 MW_e PHWRs is only 70 % of full power (FP) if it is loaded with all natural uranium fuel bundles, and the full power can be achieved only after about 100 full power days (FPD) of operation. However, the maximum operating power can be improved by flattening of the flux. For the equilibrium core, the flux flattening is achieved by adopting a differential on-line refuelling scheme for the two burnup zones of the core. For the fresh core, the flux flattening was achieved by using differential enrichment, that is, by loading depleted bundles in the central region of the core and natural uranium in the remaining part of the core. This practice was followed in the Madras Atomic Power Station (MAPS) and the Narora Atomic Power Station (NAPS). With the differential enrichment fuel loading scheme, the maximum power achievable was about 90 % of FP from the fresh core and full power could be achieved after 50–60 FPD of operation. Another alternative for flux flattening in the fresh core was the use of thorium in PHWRs. Hence, 35 locations were optimised in the core for one thorium bundle in each channel such that it provides the required flux flattening without compromising the worth of the shut-down systems. Such a loading was tested for the first time in 1991 at the Kakrapar Atomic Power Station (KAPS-1). The 35 thorium bundles were loaded in subsequent reactors including KAPS-2, Kaiga Generating Station (KGS), and Rajasthan Atomic Power Station (RAPS-3&4). Moreover, after reconditioning of RAPS-2, the same flux flattening in the fresh core was achieved by using only 18 thorium bundles in symmetrical positions. In total, three stations (with two reactors at each station), KAPS, KGS, and RAPS-3&4, have irradiated a total of 210 thorium bundles, with 35 bundles in each reactor. All the 210 bundles have been discharged. The performance with regard to the irradiation of these bundles was found to be excellent. The maximum burnup achieved for a thorium bundle was about 14,000 MWd/tonne, and the power produced by the bundle just before discharge was about 400 kW. Moreover, it remained in the core in a high flux region for about 600 FPDs. The ^{233}U content was estimated for this bundle and was about 150 g. However, the average ^{233}U content in the 210 irradiated thorium bundles was about 100 g per bundle. The fuel integrity of these bundles was very good and the fuel performed well in the operating conditions.

Post-irradiation Examination of the Thoria Bundles

Post-irradiation examinations of some of these bundles were carried out and a few samples from the bundles were also analysed. The measured results indicated some variations in the measured isotopic composition of uranium with respect to the predicted one. The samples taken for analyses were from the ends of the bundle, whereas the predictions were for the whole bundle on an average basis. Therefore, a few more samples were analysed to confirm the results.

Fabrication of Thorium Fuel

To support the fuel cycle of the AHWR, several technologies have been brought to a level of maturity, and some new ones are being developed. It is expected that such a fuel cycle will not only cater to the special challenges associated with

re-fabrication of the radioactive thorium-based fuel, but will also use all the fissionable materials in the fuel cycle very efficiently, while minimising the radiological toxicity of the waste stream. A part of the fresh fuel for AHWR, bearing ^{233}U, will be required to be produced remotely behind thick shields owing to the presence of ^{232}U. Schemes for laser isotopic clean-up of ^{233}U, as well as several alternative advanced technologies for remote fabrication of this fuel on a large scale, are in progress [4]. Notable among the latter are sol–gel microsphere pelletisation, vibropacking, pellet impregnation, and advanced agglomeration technologies [5, 6]. Facilities have been set up for characterisation of thoria-based fuels [7].

Reprocessing of Thorium Fuel

Extensive research and development studies have been carried out in the field of thoria-based spent fuel reprocessing [7, 8] and waste management fields for more than a decade, including research on issues related to the closing of the AHWR fuel cycle. The major areas addressed are optimisation of solvent extraction and scrubbing stages to improve the product purity of ^{233}U, tail-end purification of ^{233}U, studies on the dissolution of thoria, development of a matrix to immobilise radioactive waste, a study on the compatibility of materials for construction in reprocessing and waste management operations, and matrix development for vitrification. Regarding the technology development for reprocessing of thoria fuel, encouraging results were obtained. Programmes were launched as a part of the third stage activities to reach a desirable goal of using all the fissionable materials in the fuel cycle very efficiently while minimising the radiological toxicity of the waste stream. A facility named Power Reactor Thoria Reprocessing Facility (PRTRF) has been constructed at Trombay [8, 9] for processing the irradiated Zircaloy-clad thoria bundles from PHWRs, to separate ^{233}U. Thoria subassemblies from power reactors such as KAPS-1&2, MAPS-1, RAPS-3&4, KAIGA-1&2, will be reprocessed at PRTRF. This facility will provide some rich, new experiences as several new technologies are being adopted in the plant. The process flow sheet involves dismantling of the spent fuel bundle end plates by laser cutting for the first time by using a 150 W Nd: YAG laser mounted on a computer based numerical control system. Individual pins from the bundle will be chopped into pieces in one stroke and fed into the dissolver for leaching with nitric acid in the presence of sodium fluoride–aluminium nitrate. ^{233}U will then be selectively extracted and further purified by ion exchange prior to conversion to oxide. The thorium stream containing the bulk of the fission products will be treated to separate thorium from fission products. The separated thorium will be stored as thorium nitrate and the high level liquid waste containing fission products will be immobilized in a suitable matrix.

Waste Management in the Thoria Fuel Cycle

High level radioactive liquid waste generated during reprocessing of thoria-based spent fuels is expected to contain thorium, aluminium, and fluoride in addition to the host of fission products, corrosion products, and actinides. The presence of these elements adds a few additional challenges with respect to vitrification in view of the refractory nature of thoria and the corrosion behaviour of fluoride. Modifications are needed in the sodium borosilicate-based matrix to improve the solubility of ThO_2 in the glass. The presence of aluminium in the waste necessitates higher pouring temperatures owing to the increased viscosity. Extensive R&D has been carried out on the development of melter technology [8, 9] for achieving temperatures exceeding 1200 °C so as to realize higher waste loading in the matrix. In view of the reactive nature of fluoride, material issues during vitrification of High Level Waste (HLW) were also studied extensively. Investigations on matrix development for waste immobilization also were carried out. Typically, borosilicate glass matrix has been adopted worldwide for management of HLW. The solubility of actinides in the silicate glass matrix is low; therefore, attempts were made to try various modifiers in a sodium borosilicate glass matrix to improve the solubility of thorium without impairing the desirable properties of the conditioned product. Based on the studies [10], it was inferred that about 16 wt% of ThO_2 can be incorporated in a barium borosilicate glass matrix without any adverse effects on its thermophysical properties.

Thorium-Based Innovative Reactors

The significant advantages of thorium-based fuels, as elaborated in the previous sections, make it a favourable choice in many innovative and advanced reactor designs for the Indian nuclear programme [11].

Advanced Heavy Water Reactor (AHWR)

A 300 MW_e thorium-based AHWR [12] is being developed at the Bhabha Atomic Research Centre (BARC). It is a vertical pressure tube type, boiling light water cooled, and heavy water moderated reactor. The reactor is fuelled with a dual MOX (mixed oxide) fuel consisting of $(^{233}U–Th)O_2$ and $(Pu–Th)O_2$. AHWR can adopt various fuel cycles including fuel cycles based on [Th–low-enriched uranium (LEU)] MOX, among others. The AHWR provides a platform for development of thorium fuel cycle technologies on an industrial scale. The design of AHWR is fine-tuned towards deriving nearly two thirds of its power from thorium-based fuel. Another version of AHWR, known as

AHWR-LEU, which is fuelled with (LEU–Th)O_2, has also been designed. AHWR is a first-of-a-kind (FOAK) reactor in several respects: it is based on the thorium–uranium fuel cycle, it has heat removal through two-phase natural circulation, passive injection of cooling water by an emergency core cooling system (ECCS) directly inside the fuel cluster, passive decay heat removal paths, and several inherent safety features. The physics safety design challenges included achieving a negative coolant void coefficient of reactivity in a pressure tube system, a uniform core power distribution, and tailoring the axial flux distribution to have improved thermal margins. The fuel cycle is based on plutonium as the external fissile feed and has been optimised to achieve self-sufficiency in ^{233}U, which was also a challenging task. Additionally, AHWR will facilitate development of all technologies required for the third stage of India's nuclear power programme, enabling large-scale utilisation of indigenous thorium resources to meet Indian energy needs.

AHWR will nearly eliminate the need for emergency planning in the public domain following any credible accident scenario in the plant, thereby allowing it to be located near population centres. The safety goal for AHWR is to achieve the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) directives of eliminating severe accident, meltdown, and radioactivity release. This has been achieved by introducing several FOAK passive safety features as a part of the defence-in-depth strategy. These FOAK features have been validated in integral test facilities simulating AHWR conditions. These facilities also helped to test several passive devices required to achieve safety without operator intervention. The safety analysis of AHWR was carried out for an exhaustive list of postulated initiating events (PIEs) by considering deviations in operating parameters as well as equipment malfunctions or postulated failures. The analyses of these PIEs and their consequences were assessed against the acceptance criteria to deterministically evaluate safety of the AHWR. The estimated probability of unacceptable radioactivity release beyond the plant boundaries is less than 1×10^{-7}, which is lower by an order of magnitude than present nuclear reactors. Incorporation of inherently safe and passive features has resulted in a robust design to mitigate the consequences of extreme natural events such as the recent Fukushima event. Although AHWR design claims only a grace period of seven days, analysis of Fukushima-type station blackouts revealed that the reactor can survive safely for a period of more than three months without any operator action.

Commercial viability of this reactor has been demonstrated by technology development so as to achieve a design lifespan of 100 years. Assurance of structural integrity and the long life of 100 years of the primary system components [13] of the AHWR has been demonstrated by improvements in manufacturing technology, accelerated ageing studies on materials/welds, and full-scale piping component tests [14] under different combinations of cyclic loading that are anticipated during operation and accidental conditions. Accelerated tests were also carried out on the pre-stressed concrete structures to determine concrete shrinkage and tendon relaxations. From these studies, it was concluded that structural integrity would not be impaired for 100 years' of operation.

AHWR incorporates several passive safety systems to enhance safety, reliability, and public acceptance in addition to improving the economic competitiveness. Passive systems are incorporated for core cooling under normal operation, shutdown, and accident scenarios. Additionally, AHWR adopts passive systems for containment cooling following a loss of coolant accident (LOCA). Besides, innovative passive safety systems have been designed for several non-cooling applications, such as reactor trip in case of wired shut-down system failure, containment isolation, and automatic depressurisation.

AHWR basic design and experimental development in areas required to establish feasibility have been completed. Several major experimental facilities have been set up to produce additional data. The designs of the major systems, structures, and components have been completed. The reactor physics design of the core has been completed. The AHWR general arrangement is shown in Fig. 6.

A critical facility, a low power research reactor, built for conducting physics experiments for validation of the physics design parameters of AHWR, was made critical in April, 2008. Peer review of the design of AHWR has been completed. Pre-licensing safety appraisal of AHWR was carried out by the Indian regulating agency, Atomic Energy Regulatory Board (AERB). Presently, detailed engineering of AHWR is being carried out in consultancy mode. The site selection process is underway. AHWR is expected to attain first criticality in the mid-2020s.

Indian High Temperature Reactors

The current Indian high temperature nuclear reactor programme [15] is mainly based on requirements related to the production of hydrogen by splitting water using high efficiency thermochemical processes. BARC is currently developing concepts of high temperature nuclear reactors capable of supplying process heat at a temperature around 1000 °C. These nuclear reactors are being developed with the objective of providing energy to facilitate combined production of hydrogen, electricity, and drinking water. The reject and waste heat in the overall energy scheme are proposed to be utilised for electricity generation and desalination, respectively. Presently, technology development for a

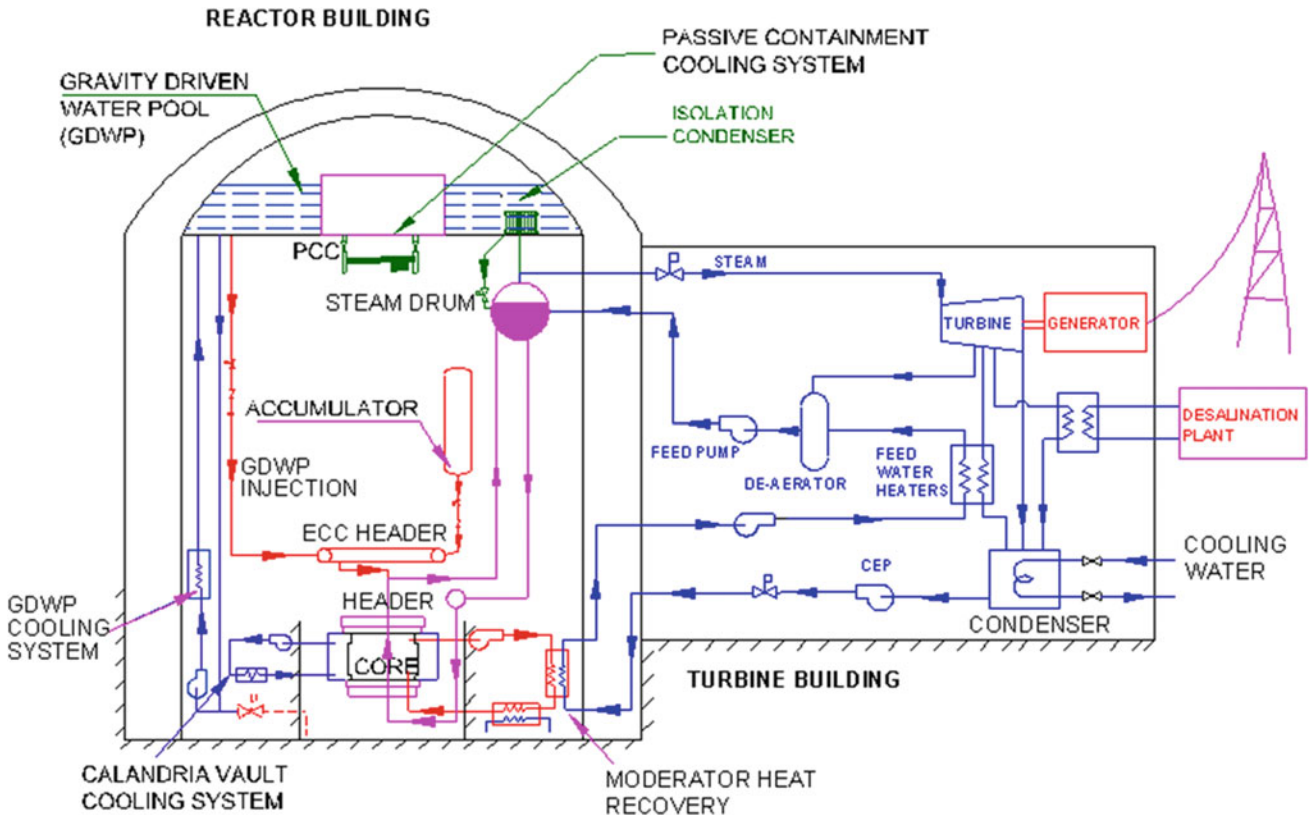

Fig. 6 AHWR general arrangement

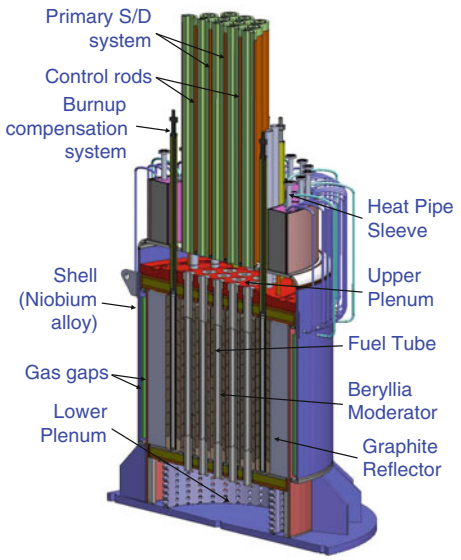

Fig. 7 Cross-sectional view of CHTR

low power compact high temperature reactor (CHTR, Fig. 7) capable of supplying high temperature process heat at 1000 °C is being carried out. In addition, conceptual design of a 600 MW_{th} innovative high temperature reactor (IHTR, Fig. 8), capable of supplying heat at 1000 °C for large-scale hydrogen production, is also being carried out. CHTR is a ^{233}U–thorium fuelled, lead–bismuth cooled, and beryllium oxide moderated reactor. This reactor will have a core life of around 15 years and will have several advanced passive safety features to enable its operation as a compact power pack in remote areas not connected to the electrical grid. An alternate design with a ^{235}U-based fuel has also been worked out. Important technologies related to the development of TRISO-coated particle fuel [16]; special materials such as high-density isotropic graphite and carbon–carbon composites [17], high-density beryllium oxide, oxidation-resistant coatings [18, 19], and niobium alloys [20] are in the advanced stages of development. The reactor heat is removed by passive natural circulation of coolant [21–23]. Analytical and experimental studies have been carried out to establish various aspects of the design [24–26]. A lead–bismuth natural circulation loop, operating at 550 °C has been in operation for the last five years. Another loop to facilitate operation at 1000 °C has been recently commissioned (Fig. 9).

The reactor possesses the following inherent safety features:

(a) A strong negative Doppler coefficient of the fuel for any operating condition results in reactor power

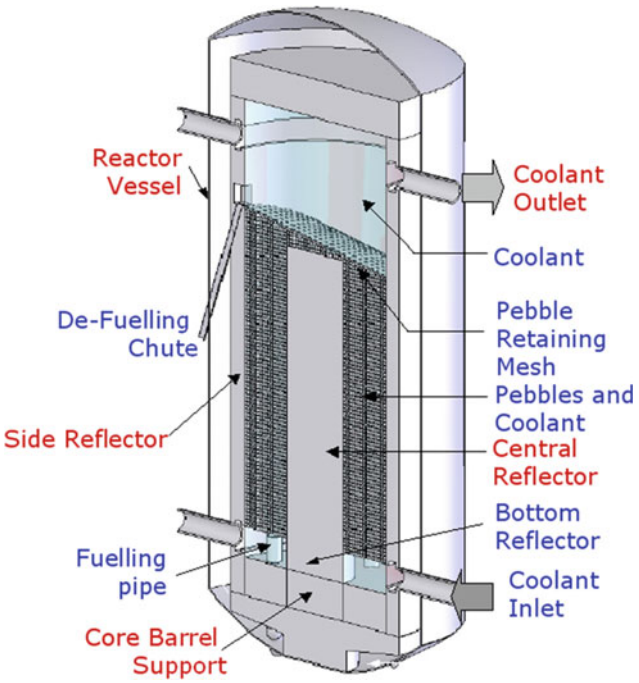

Fig. 8 Schematic of 600 MW_{th} IHTR

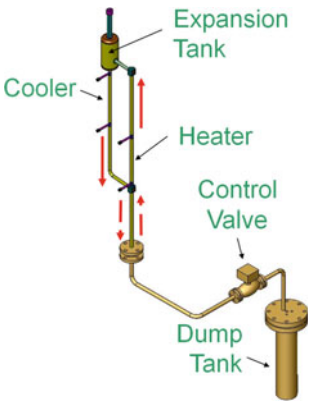

Fig. 9 Schematic of the LBE natural circulation test facility operating at 1000 °C

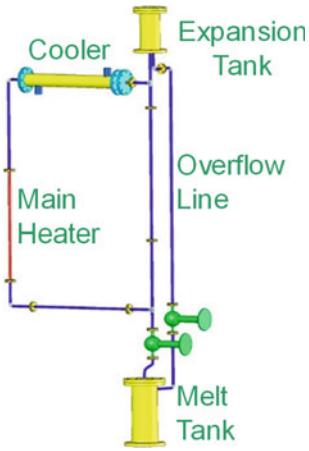

Fig. 10 Schematic of the molten salt natural circulation test facility

Fig. 11 Molten salt corrosion test facility

reduction in case of fuel temperature rise during any postulated accident scenario;

(b) High thermal inertia of the all-ceramic core and low core power density (~0.5 W/cc) results in very slow temperature rise of the reactor core components as well as fuel during conditions when all heat sinks are lost;

(c) A large margin between the normal operating temperature of the fuel (around 1100 °C) and the allowable limit for the TRISO-coated particle fuel (1600 °C) enables retention of fission products and gases during normal operating conditions. This also provides a healthy margin to take care of any unwanted global or local power excursions;

(d) A negative moderator temperature coefficient results in lowering of the reactor power in case of an increase in moderator temperature as a result of any postulated accident conditions;

(e) Owing to the use of liquid lead–bismuth eutectic (LBE) coolant with a very high boiling point (1670 °C), there is a very large thermal margin to its boiling at the normal operating temperature of 1000 °C. This eliminates the possibility of heat exchange crisis and

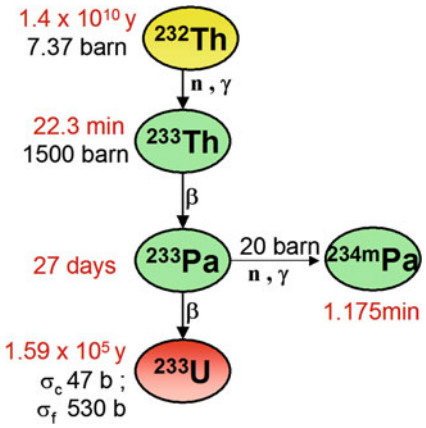

Fig. 12 Conversion chain of ^{232}Th to ^{233}U

increases the reliability of heat removal from the core. The coolant operates at low pressure; thus, there is no over pressurisation and, hence, no chance of any adverse conditions caused by coolant overheating;

(f) The LBE coolant, which is maintained in an inert gas atmosphere, is itself chemically inert. Even in the eventuality of accidental contact with air or water, it does not react violently;

(g) Owing to the above-atmospheric-temperature coolant melting point (123 °C), even in case of a primary system leakage, it solidifies and prevents further leakage;

(h) The thermal energy stored in the coolant that is available for release in the event of a leak or accident is small;

(i) The very low pressure in the coolant allows use of a graphite/carbon-based coolant tube with a low neutron absorption cross section; thus, improving the neutronics of the reactor;

(j) A low induced long-lived gamma activity of the coolant so that, in the case of a leakage, the coolant retains iodine and other radionuclides;

(k) For the LBE coolant, the reactivity effects (void, power, temperature, etc.) are negative; thus, reducing the reactor power in case of any inadvertent power or temperature increase.

IHTR is a 600 MW_{th} molten salt cooled reactor with pebble-based fuel. All core materials are made of nuclear-grade high-density isotropic graphite. To study the thermal hydraulics behaviour of the molten salt coolant, a molten salt natural circulation loop (Fig. 10) has been set up. In addition, an experimental facility to study the corrosion behaviour of molten salts on structural materials has also been set up (Fig. 11). Experimental set-ups as well as other developments related to fuel and materials have been initiated.

Indian Molten Salt Breeder Reactor (IMSBR)

Despite its other advantages, thorium, if used in conventional reactors, suffers from one disadvantage: ^{233}Pa is an intermediate isotope in the chain of nuclear reactions (Fig. 12), which results from the conversion of ^{232}Th to ^{233}U. The conversion of ^{233}Pa to ^{233}U is possible through beta decay. However, in a reactor core, ^{233}Pa is always surrounded by fission neutrons, which if absorbed, neither leads to the formation of ^{233}U, nor to fission, and hence is a parasitic loss. This is unavoidable in the case of solid-fuelled reactors, as the fuel pins need to be exposed to certain burnups before being extracted for reprocessing. This can be avoided if the fuel is in fluid form, so that the ^{233}Pa is separated out as soon as it is produced, and allowed to decay to ^{233}U outside of the core. A feasible way of doing this is the molten salt reactor.

The fuel in a molten salt reactor is in the form of a continuously circulating molten salt. Fluoride-based salts have been proposed. A crucial part for achieving reasonable breeding in the thermal/epithermal spectrum in such reactors is the need to reprocess the salt continuously and on-line. This constitutes a major technological challenge for this type of reactor. India has recently started carrying out fundamental studies so as to arrive at a conceptual design for the Indian molten salt breeder reactor (IMSBR) [27]. Presently, various design options and possibilities are being studied from the point of view of reactor physics and thermal hydraulic design. In parallel, studies on various molten salts have also been initiated for their characterisation, purification, thermal hydraulic behaviour, etc.

Developmental Issues for Molten Salt Breeder Reactors

The MSBR technology is quite different from the currently existing reactor systems in India. Hence, design and development related activities are being initiated. A partial list has been identified for initial development and includes the following:

(a) Development of closely coupled neutron transport and computational fluid dynamics (CFD) codes with capability to account for on-line reprocessing;

(b) Material compatibility studies for core and structural materials;

(c) Characterisation and purification of salts;
(d) Studies on thermal hydraulics of molten salts;
(e) Studies and development of on-line reprocessing systems;
(f) Evolution of safety philosophy;
(g) Development of instrumentation sensors;
(h) High efficiency energy conversion systems.

Conclusion

Availability of significant resources of thorium forms the basis of the Indian three-stage nuclear programme. Thorium offers a sustainable and proliferation-resistant fuel option with a lower minor actinides burden. This, coupled with the other significant advantages that a ^{233}U–Th system offers, makes it a preferred choice for the third stage. Over a period of time, indigenous technologies have been developed in India for all aspects of the thorium fuel cycle, viz., mining, fuel fabrication, fuel irradiation, reprocessing, and waste management. Design of various reactor systems have been worked out. The design of the Advanced Heavy Water Reactor (AHWR), for which site evaluation is in progress, makes use of a thorium-based fuel and has many enhanced safety features. It is being developed as a technology demonstrator for an industrial-scale thorium fuel cycle. Thorium is also planned to be used in the 600 MW_{th} Innovative High Temperature Reactor (IHTR), which is aimed at producing hydrogen by splitting water; thus, reducing our dependence on imported crude oil. Large-scale deployment of molten salt breeder reactors (MSBRs) is envisaged during the third stage of the Indian nuclear power programme. This reactor type, with ^{233}U–Th fuel in the form of molten salts, with provision of on-line reprocessing, can be configured to be self-sustaining, if not as a breeder, and provides an attractive option for the third stage of the Indian nuclear programme.

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The Role of Thorium in Nuclear R&D in the UK

Robert Arnold

Abstract

The UK has recently revised its nuclear strategy to allow for possibly significant expansion of nuclear energy generation during the 21st century. Key to this is the need for a fuel cycle that is sustainable over the long term and the maintenance and development of the skills, technologies and infrastructure to deploy it. In support of this, a nuclear research and development roadmap has been created and the fuel cycles of possible future nuclear generation scenarios have been modelled. One such scenario covers the deployment of molten salt fast reactors (MSFRs) based on the neutronic modelling of the EVOL FP7 project. The potential speed of deployment of MSFRs is considered under different scenarios and the requirements for R&D involvement for both these and wider thorium technologies are examined in the light of the overall UK nuclear strategy.

UK Nuclear R&D Strategy

History of Nuclear R&D Need in the UK

The UK is committed to reducing greenhouse gas emissions by at least 80 % relative to 1990 levels by 2050. The 2050 Pathways Analysis Tools were developed to analyse possible technology trajectories to de-carbonisation based on current consumption. The study reported here and subsequent studies suggested that up to around 75 GW nuclear power generation may be needed during the 21st century.

Decisions taken between 2008 and 2013 make new nuclear builds and a significant share of UK electricity generation from nuclear seem increasingly likely. These decisions include policy statements, approval of new reactor designs, consortia bidding to build, etc.

In December 2011, the House of Lords (the UK's 2nd Parliamentary chamber) drew attention to risks over UK nuclear R&D and skills retention. As a consequence, the government initiated a review of strategy and an analysis of skills and technology needs for the near- and long-term future.

In March 2013, an extended nuclear strategy, including a nuclear R&D roadmap was published [1].

Roadmap for Nuclear R&D in the UK

The Roadmap is the skills and technology trajectory analysis for the UK nuclear industry, devised under the principle of keeping technology options open. Many scenarios were considered, but all their needs fall within the bounds of three scenarios, representing limits of likelihood, which are those considered for the Roadmap:

- No new nuclear power plant (not even those currently proposed);
- Up to 75 GW open cycle by the mid-21st century;
- Up to 75 GW closed cycle by the mid-21st century.

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The Role of Fuel Cycle Modelling and the Role of Thorium

Fuel Cycle Modelling with ORION

All scenarios used the ORION fuel cycle modelling code, developed by the UK's National Nuclear Laboratory. The code runs radioisotope inventory analysis to determine the throughput of material throughout a number of facilities in the nuclear fuel cycle, including storage (e.g. at the mine, mill or final disposal facility), fuel fabrication facilities, reactors, and reprocessing plant. This allows complete mining-to-disposal modelling of fuel cycle scenarios by predicting the evolution of fuels and wastes as they progress through nuclear facilities.

In particular:

- Feasible futures require feasible fuel cycles. ORION ensures a feasible fuel cycle;
- Modelling extends to the shutdown of the fuel cycle or to an equilibrium in the late 22nd century;
- Descriptions of many variants of individual facilities are included in the model;
- ORION requires as an input a description of the neutronics of any reactors being considered.

For modelling the isotopic inventories within a reactor, ORION requires burn-up-dependent, shielded cross-sections produced by post-processing the results from deterministic or Monte-Carlo-based neutronic analyses of the reactor core. ORION has the capability to calculate the radiotoxicity, toxic potential, activity, spontaneous neutron emission rate and decay heat throughout the fuel cycle, as around 2500 isotopes, including fission products and actinides, are tracked. For radiotoxicity calculations, doses are evaluated using ingestion conversion coefficients provided in International Commission on Radiological Protection (1996). For decay heat and neutron emission rates, data from the JEF-2.2 Nuclear Data Library (OECD-NEA, 2000) are used

ORION Scenarios

ORION models were generated for three electrical power scenarios (16, 40 and 75 GWe) with many variants. A typical output includes: decay heat key factors (governing repository capacity), radiotoxicity, and volumes of waste. An example 75 GWe variant assumes a new build light water reactor (LWR) fleet followed by a sodium fast reactor fleet to provide long-term sustainability. A thermal fleet is required to generate a sufficient amount of plutonium to fuel the fast reactor fleet. Figure 1 shows the level of power generated over time and the type of reactors contributing to that power.

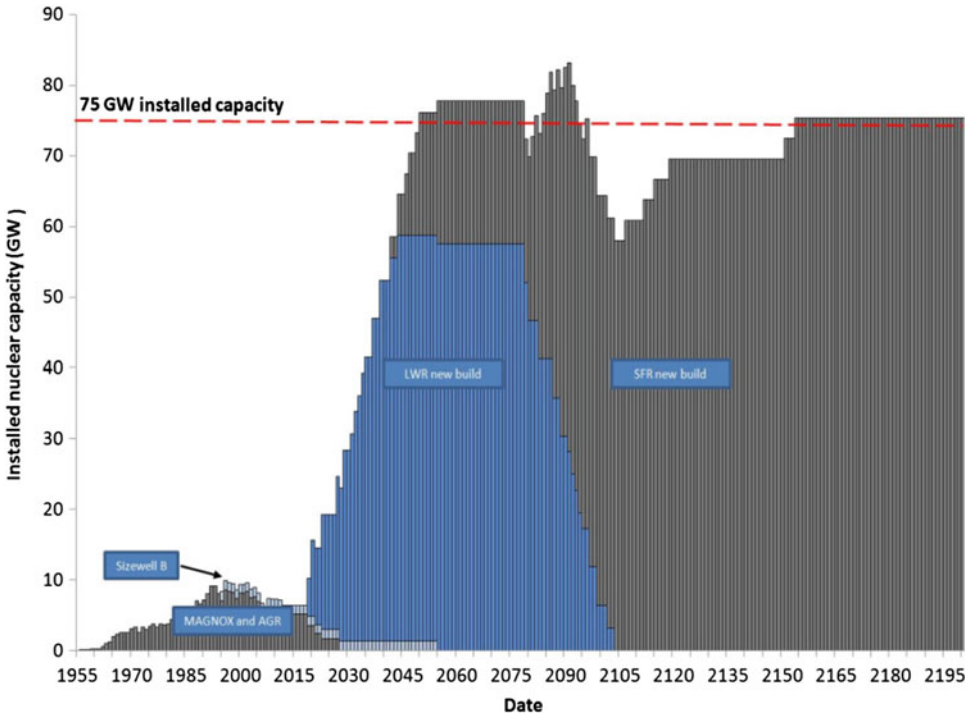

Fig. 1 The source of power generated against time for a 75 GW_e closed fuel cycle. The fuel cycle modelling has assumed new reactor build contributions from (i) The UK European Pressurized Reactor (EPR) design developed by AREVA and Electricité de France (EDF) and (ii) The AP1000 design developed by Westinghouse Electric Company (WEC). Taken from [1]

Table 1 R&D programme synergies: comparison of the current baseline pathway with the closed and open fuel cycle pathways with 16 to 75 GW_e nuclear generation, in terms of skills and technologies (Adapted from [1])

	Next generation reactors/fuels	Reprocessing/recycle	Advanced thermal reactors/fuels	Current thermal reactors/fuels	Material and components	Construction and installation
Closed fuel cycle pathway	√	√	√	√	√	√
Open fuel cycle pathway			√	√	√	√
Baseline pathway				√	√	√
	Operations	Spent fuel and nuclear material	Decommissioning and clean-up	Waste management	Geological disposal	Fusions reactors/fuels
Closed fuel cycle pathway	√	√	√	√	√	√
Open fuel cycle pathway	√	√	√	√	√	√
Baseline pathway	√	√	√	√	√	√

Roadmap Findings

The main findings of the Roadmap are the following:

- A closed fuel cycle, recycling fuel into fast reactors has significantly lower demands on fuel and final repository thermal management than an open one;
- Most technologies and skills are needed in all scenarios (Table 1).
- Assessing thorium-fuelled reactors and understanding the implications for the attainable rates of expansion of nuclear capacity will be important for understanding the potential role of thorium in a UK fuel cycle;
- Further analysis and fuel cycle modelling will be necessary to understand the implications for waste management and disposal of using thorium fuels.

In the wider UK context, decisions on which fuel to use are down to the developer and, of course, the regulator.

Modelling of a Fast Spectrum Thorium-Fuelled Molten Salt Reactor (MSR)

Scenarios involving thorium-fuelled molten salt fast reactors were included early on in the NNL's fuel cycle modelling for two main reasons:

- A model was already available—the neutronic description developed by Dr. E. Merle-Lucotte [2] and used as part of the EVOL project [3];
- MSFR require fewer key constraints than conventional fast reactors:
  - There is no need for a cooling-off period for spent fuel—immediate recycle reduces cycling time for fuel;
  - The highest degree of recycling for fast reactors is potentially possible;
  - A mechanical structure of solid fuels and associated neutron loss is avoided.

The design parameters for the core were chosen to be the same as those used by Merle-Lucotte et al. [1] (Fig. 2). The

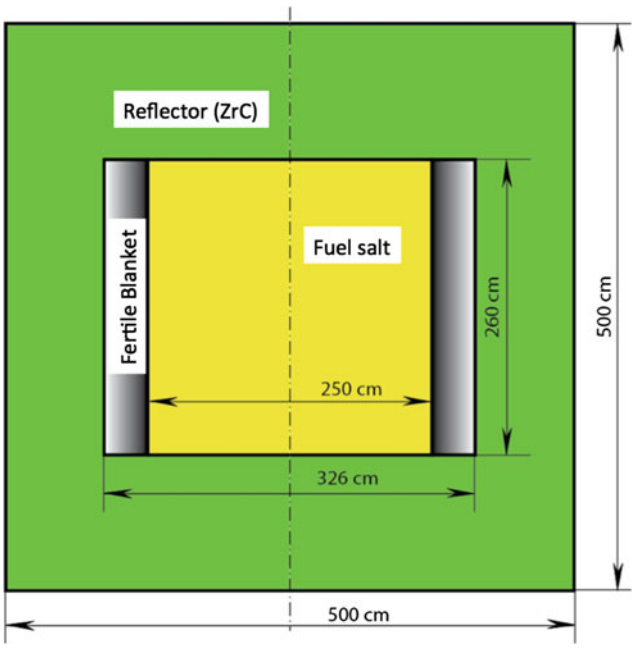

Fig. 2 Schematic of the MSR core used in the NNL study

same two start-up scenarios were also used for the initial fissile load: U-233 and Pu + minor actinides (PuMA). Both fuel loadings were assumed to start with a heavy nuclide initial molar concentration of about 17.5 %. However, the NNL study used a different neutronic modelling code. Whereas Merle-Lucotte et al. used MCNP [4], NNL [5] used ERANOS [6]. The NNL modelling assumed that U-233 is extracted on recycle.

In the end, the output of the simulation is an estimate of the rates of roll-out resulting from breeding alone.

The comparison between the two startup assumptions is illustrated in Fig. 3. Excessive reactivity in the core started with PuMA implies greater U-233 production (and extraction to manage the core) and a shorter doubling time for the fissile inventory. However, the core started with U-233 has a much lower inventory of transuranic elements (and hence lower very-long-term radiotoxicity) early on in the fuel cycle (Fig. 4).

Eventually, the heavy nuclide inventories from the two start-up scenarios do converge. The fissile inventory dou-

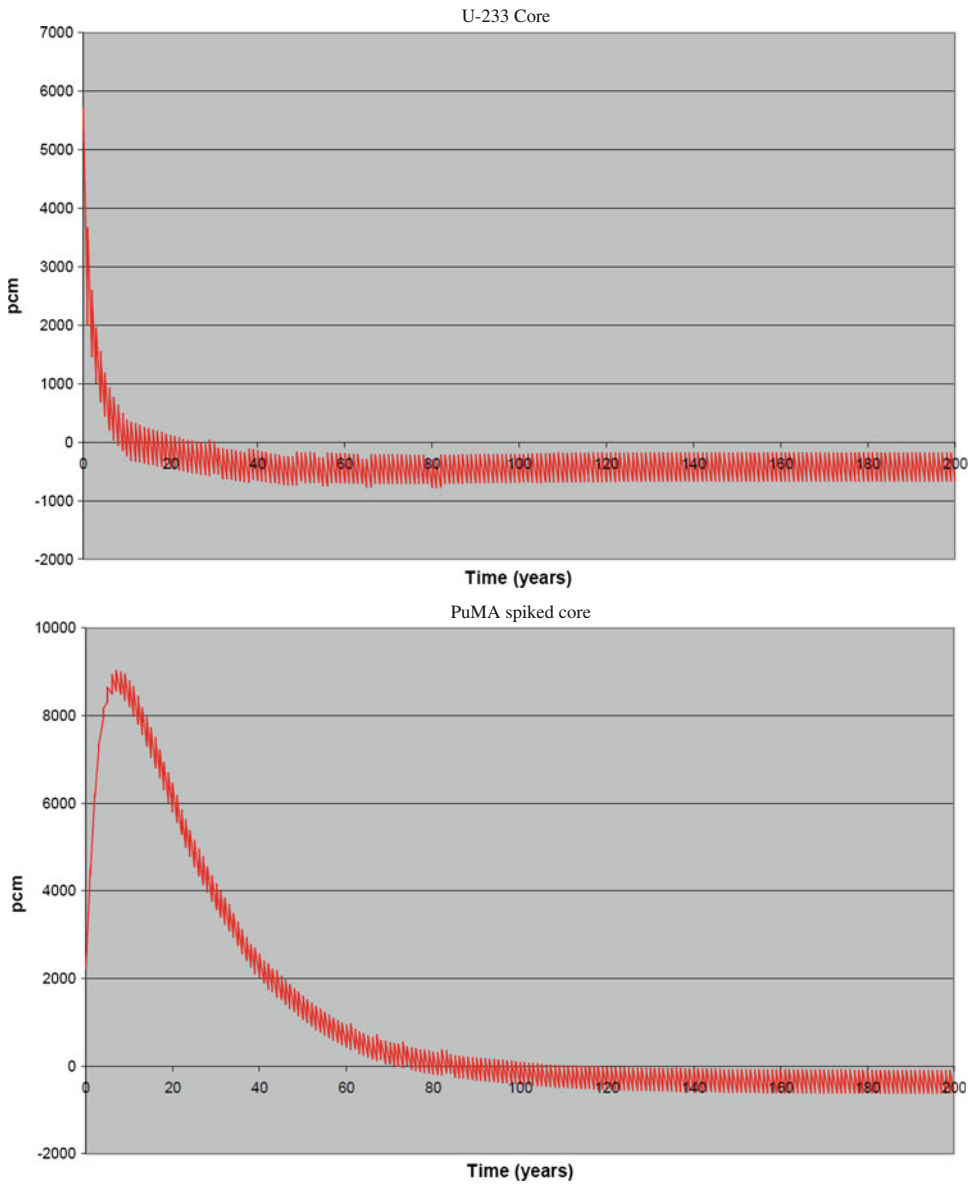

Fig. 3 Time evolution of the reactivity expressed in pcm for the core started with U-233 (*upper*) and the core started with PuMA (*lower*)

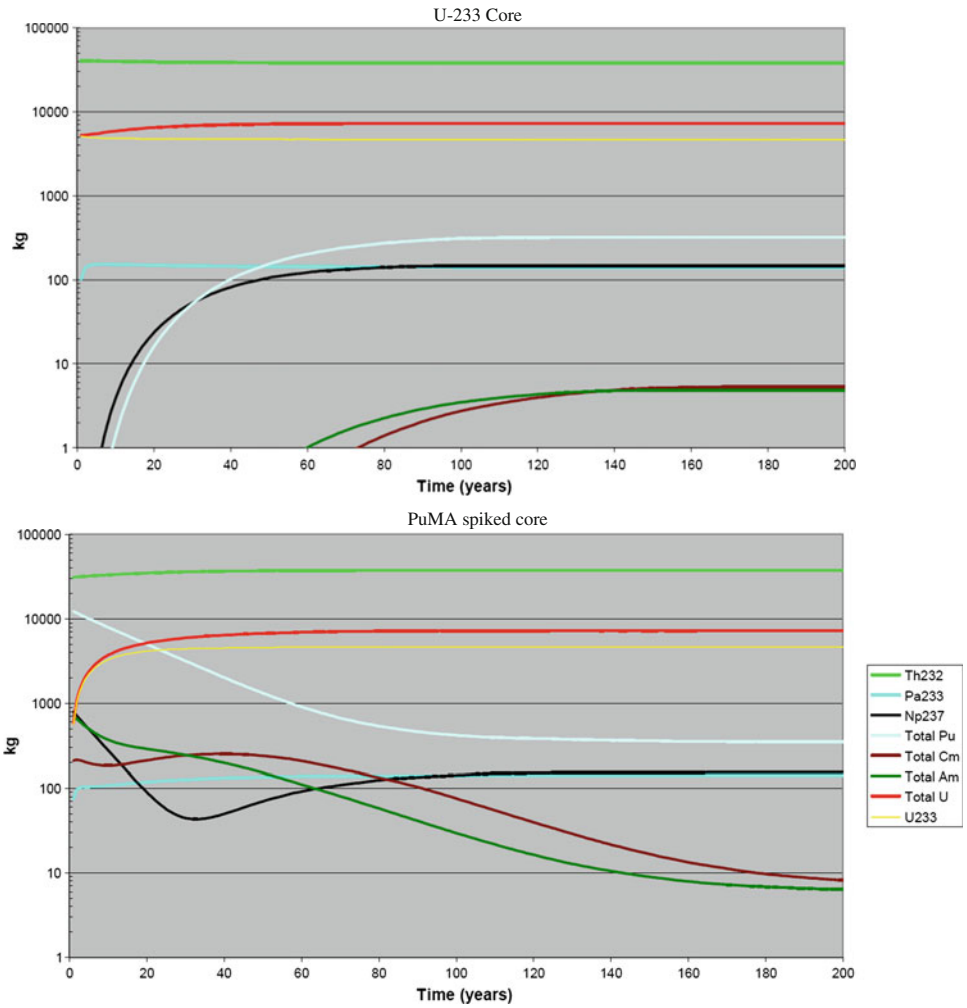

Fig. 4 Time evolution of the inventory of heavy elements in the core (in kg) for the core started with U-233 (*upper*) and the core started with PuMA (*lower*)

bling time for core startup with U-233 is approximately 53 years, and for core start-up with PuMA it is approximately 38 years. These NNL results yield similar values to those found by Merle-Lucotte et al.

Conclusion

If relying on fuel breeding only for the roll-out of thorium-fuelled MSFRs (if they are developed), the speed of roll-out is likely to be improved by the use of plutonium and minor actinides as the initial fissile charge rather than U-233. However this entails higher long-term radiotoxicity of the core inventory for at least the first 100 years of the fuel cycle.

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Feasibility and Desirability of Employing the Thorium Fuel Cycle for Power Generation

Bal Raj Sehgal

Abstract

Use of the thorium fuel cycle for nuclear power generation has been considered since the very start of the nuclear power era. In spite of a large amount of research, experimentation, pilot-scale and prototypic-scale installations, thorium fuel has not been adopted for large-scale power generation (Loewenstein and Sehgal in Trans Am Nuclear Soc, 27:312, 1977 [1]; Sehgal in The application of thorium in fast breeder reactors and in cross-progeny fuel cycles, NARA, Japan, 1982 [2]). This paper reviews the developments over the years on the front and the back ends of the thorium fuel cycle and describes the pros and cons of employing the thorium fuel cycle for large-scale generation of nuclear power. It examines the feasibility and desirability of employing the thorium fuel cycle in concert with the uranium fuel cycle for power generation.

Introduction

Natural thorium, unlike natural uranium, does not contain a fissile element. The predominant isotope, ^{232}Th, is a fertile material like ^{238}U, the predominant isotope in natural uranium. ^{232}Th has a fast fission cross section, but much smaller than that of ^{238}U and at higher neutron energy. It also has a much higher neutron capture cross section for thermal neutrons and a much lower resonance integral [3] than those for ^{238}U. Thus, replacing ^{238}U with ^{232}Th in a thermal reactor would require a greater amount of fissile material (^{239}Pu or ^{235}U) to bring system to criticality [4] and to achieve the equivalent length of operation. This is a significant disadvantage for using thorium in thermal reactors [5].

However, the capture of a neutron by thorium provides a significant advantage for the use of thorium in thermal reactors, as the ^{233}U produced by neutron capture with ^{232}Th is arguably the best fissile fuel for thermal reactors. It has a much greater value for η, the neutron yield in fission, per neutron absorption, than both ^{235}U and ^{239}Pu. This fact has been known since the 1950s and was the driver for the initiation of the first large-scale application of the thorium fuel cycle in nuclear power generation, through the light water breeder reactor (LWBR) project in the USA, which produced power in the Shippingport light water reactor (LWR) plant. The core for the LWBR employed ^{233}U–Th oxide fuel and an innovative design pioneered by Alvin Radkowsky, called the seed blanket design in which neutron economy was the watch word.

The LWBR core was reprocessed and it was demonstrated that indeed a small breeding gain was obtained. The reprocessing campaign was a one-time endeavor at the uranium fuel reprocessing plant at West Valley, New York, which was adapted for the reprocessing of the thorium–uranium irradiated fuel. There was, however, much activity in the USA at the pilot scale level on thorium fuel reprocessing and refabrication, the two elements of great importance if the complete advantages of using the thorium fuel cycle are to be realized. A brief review of the pilot-scale development work will be provided in this review paper. The review will be selective in assessing the main positive and negative features to give an objective technical view point on the desirability and feasibility of establishing the thorium fuel cycle for nuclear power generation in future.

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Table 1 Reactors operated with substantial amounts of thorium

Reactor	Type	Location	Operating period (with thorium)	Thermal power (MW_{th})	Electric power (MW_e)
Elk River	BWR	USA	1962–1968 [9]	58 [9]	15[a] [11]
Indian Point	PWR	USA	1962–1964 [9]	615 [9]	151[a] [11]
Peach Bottom 1	HTGR-prismatic	USA	1966–1972 [8]	115 [9]	40 [12]
Shippingport	LWBR[b]	USA	1977–1982 [8]	236 [9]	60 [5]
Fort St Vrain	HTGR-prismatic	USA	1974–1989 [9]	842 [9]	330 [12]
AVR	HTGR-pebble	Germany	1967–1988 [8]	45 [10]	15 [8]
THTR	HTGR-pebble	Germany	1985–1989 [8]	750 [10]	300 [8]
Various	PHWR	India	Ongoing	–	–
Dragon	AGR	England	1966–1973	20 [8]	
FBTR	LMFBR	India	Ongoing	40 [8]	

[a]Plus additional from fossil-fired super heaters
[b]Data represents the LWBR core, not the original Shippingport core, which did not use thorium or ^{233}U

Front End of the Thorium Fuel Cycle

A history of the reactors that operated with substantial amount of thorium is provided in Table 1. A short description of the experience achieved is provided in the following paragraphs:

Experience with Water Reactors

The Elk River boiling water reactor in Elk River, Minnesota, USA, was designed by Allis-Chalmers. It was much smaller than, but similar in many ways to today's BWRs. It utilized fuel consisting of UO_2–ThO_2 as cylindrical, ceramic pellets contained in stainless steel cladding. The reactor was shut down after only six years of operation because of "extensive stress-corrosion cracking" in the "primary coolant system", a reason that had nothing to do with the thorium fuel [6].

The Indian Point 1 pressurized water reactor (PWR) started in 1962 using the first core fuel containing both UO_2–ThO_2 and pure UO_2 as cylindrical, ceramic pellets, and like Elk River, it used stainless steel cladding. The uranium was enriched to about 93 % ^{235}U. Low-enriched uranium, without thorium, was used after the first cycle. The reactor was shut down in 1974, primarily because of the lack of an emergency core cooling system [6].

The final core of the Shippingport PWR plant was started up with thorium and ^{233}U that had been produced in classified US Department of Energy reactors. The LWBR was a non-conventional design that included hexagonal fuel assemblies with a moveable, central "seed" containing the ^{233}U, as shown in Fig. 1. The part of the assembly that contained only thorium was stationary. Reactor control was affected by moving the seed downwards out of the core; thus, avoiding the use of conventional, neutron-absorbing control rods.

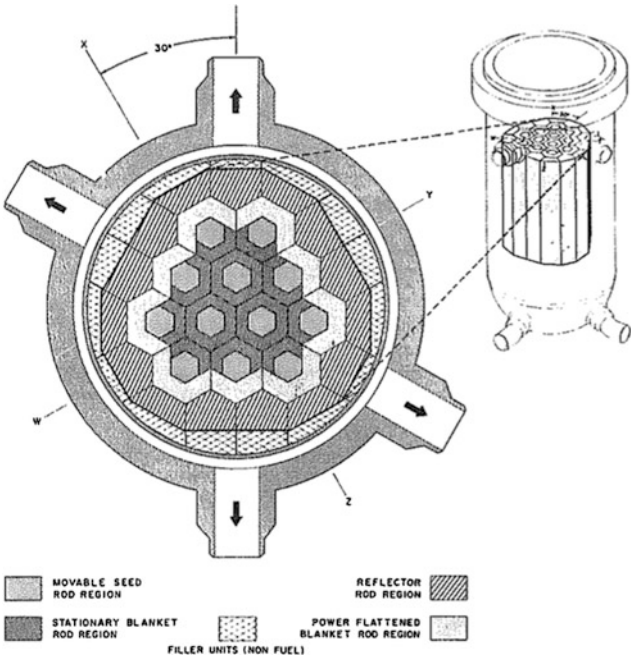

Fig. 1 Shippingport LWBR core with hexagonal seed-blanket assemblies

The fuel consisted of two kinds of ceramic fuel pellets; "duplex" pellets of ThO_2–UO_2 were used in the seed and pure ThO_2 in the blanket. Both types of pellets were clad with Zircaloy. The reactor was shut down in 1982, having obtained the objective of showing that thermal breeding could be achieved [6].

This plant was not licensed by the United States Nuclear Regulatory Commission (USNRC). It was designed and run by US Naval Reactors to their own standards, which,

however, are believed to be at least as stringent as those of the NRC. A later report issued after the LWBR was shutdown [7] did not mention any problem with the smaller delayed neutron fraction of ^{233}U than that of ^{235}U and stated that there were no safety or licensing issues experienced with this core.

Experience with Other Reactors

India has irradiated a number of thorium oxide bundles in at least three operating pressurized heavy water reactors (PHWRs) with no reported incidents. These were essentially CANada Deuterium Uranium (CANDU)-type assemblies consisting of ceramic pellets encased in cylindrical cladding. There is no indication of any safety or licensing issues associated with these thorium bundles [8].

The Peach Bottom 1 reactor in Peach Bottom, Pennsylvania, USA, was a prismatic block high-temperature gas-cooled reactor (HTGR) designed by General Atomics. It started operation in 1966. The Peach Bottom 1 reactor was permanently shut down in 1974, in part because it did not have a permanent license [6]. There is no indication that thorium use contributed to this decision.

The Fort St Vrain Reactor in Platteville, Colorado, USA, was a prismatic HTGR designed by General Atomics. It began operation in 1974. The fuel used two kinds of kernels—thorium–uranium carbide and pure thorium carbide. There are no reports of safety or licensing problems with the fuel. The plant was shut down in 1989 because of problems with the helium circulators [6, 9, 10].

DRAGON was an English HTGR that was used as a test bed for the OECD high-temperature reactor project. Among its functions was to test coated-particle thorium fuels. The fuel was deliberately driven to failure in many cases, so its safety behavior seems not pertinent in the present context [11].

The AVR and THTR were pebble bed HTGRs, both situated in Germany. Operation of the AVR revealed some pertinent safety-related characteristics of the kernel fuel, which presumably would be corrected in modern designs [12].

In summary, there have been no significant safety issues in operating water reactors or high-temperature gas-cooled reactors with thorium fuel. However, the total extent of thorium reactor experience is relatively small.

Back End of the Thorium Fuel Cycle

Introduction

It has been amply stated that the full benefit of employing thorium fuel in a power reactor is obtained only if the back end (reprocessing and refabrication) of the thorium fuel cycle is established and the ^{233}U used. Pilot- to relatively large-scale facilities were built to develop the processes needed for the reprocessing of the irradiated thorium, the separation of the ^{233}U, and the refabrication of fuel rods containing thorium and ^{233}U. Large-scale reprocessing was performed successfully at the reprocessing plant of the Nuclear Fuel Services (NFS) Company in New York State for the LWBR core and the thorium fueled core employed in the Indian Point Reactor Plant. Obtained ^{233}U is stored at the Oak Ridge National Laboratory (ORNL), Tennessee, USA.

A considerable amount of pilot-scale work was performed for thorium fuel processing and refabrication of fresh fuel at the ORNL and the laboratories of the Babcock and Wilcox (B&W) Company. Most of this work was performed in the 1960s and some literature is available from these investigations and will be described briefly in this paper.

An essential part of the thorium fuel cycle is the reliability of the good performance of the thorium fuel, enriched with uranium or plutonium, to the burnup level that is desired. If a once-through fuel cycle is employed for thorium-fueled reactors, it is beneficial to employ a very long burn-up ($\sim$100,000 MWd/tonne of thorium) in order to burn as much as possible of the ^{233}U formed in situ to compete successfully with the uranium once-through fuel cycle. Considerable data on irradiation performance of small pins and samples were obtained in the USA in the 1960s. The information available from some of the published literature is described briefly here.

There are several issues of concern that are particularly relevant to thorium fuel cycle facilities. These include, but are not limited to:

- radiation protection;
- shielded versus unshielded refabrication facilities for fuel manufacture;
- material safeguards and non-proliferation.

The radiation protection issue assumes a very large position for the back end of the thorium fuel cycle and, in particular, for the refabrication of thorium–^{233}U fuel. The major problem is the build-up of ^{232}U, which decays with the release of a hard gamma-ray. Thus, the refabrication of fuel, which takes places after the reprocessing of the irradiated thorium-based fuel and the separation of the formed ^{233}U, has to be performed either very quickly (within a few days) or it has to be performed in shielded facilities with remote operations.

The material safeguards and non-proliferation issues are of concern as ^{233}U is almost as potent a material for weapons as ^{235}U. There is, however, the mitigating factor of high personnel radiation exposure that would be incurred in working with ^{233}U owing to the content of ^{232}U.

Experience Base in the USA in the 1960s

In the following sections, we will briefly describe the experience base that was accumulated for the reprocessing, refabrication, and irradiation of thorium-based fuels. In particular, the experience gained in the refabrication of fuel rods in unshielded and shielded facilities will be described.

Reprocessing of Irradiated Thorium Fuel

The Acid–Thorex process with solvent extraction [13, 14] was employed to reprocess the irradiated thorium fuel and recover thorium and uranium. A head-end process was developed [13] at ORNL for the metal-clad ThO_2–UO_2 fuels and it was claimed to be close to being fully developed for industrial application. A shear–leach process was employed, which was also used for reprocessing the thorium fuel from the Indian Point reactor and the LWBR at the NFS facility in New York State. The published NFS price [15] is quoted as \$78/kg for reprocessing the Indian Point reactor ThO_2 core, which is the same cost as that at ORNL. A closely coupled small-volume pilot plant for reprocessing and refabrication was constructed at the ORNL. The reprocessing product is a Th–U sol–gel, which can be directly employed for the refabrication of rods. The estimated cost for both reprocessing and refabrication processes was \$156/kg of U–Th. This would decrease for the industrial-size plant. The costs of reprocessing ThO_2–UO_2 fuel was estimated to be $\sim$30 % greater than the cost of reprocessing the low-enrichment uranium oxide fuel. The reprocessing of thorium–uranium oxide fuel is somewhat more difficult than that of uranium oxide fuel owing to the slower dissolution rate of thoria and corrosion of the stainless steel equipment by the acidic fluoride needed to dissolve thoria.

An interesting set of pilot-scale tests [16, 17] were performed at the Savannah River and Hanford reprocessing plants to produce ^{233}U with less than 5 ppm concentration of ^{232}U. The successful tests produced about 160 kg of ^{233}U with 5 ppm ^{232}U content at a cost of $\sim$50 % more than that obtained for ^{239}Pu. However, the main advantage was that the refabrication of the Th–^{233}U fuel could be performed in unshielded facilities due to the very low radiation level from the decay of ^{232}U. It was found that by using thorium ore with a low content of ^{230}Th helped to reduce the formation of ^{232}U.

Another interesting process developed to reduce the radiation hazard while working with the thorium–^{233}U fuel reprocessing and refabrication processes is to immediately separate the daughter ^{228}Th from the ^{233}U, as that represents a substantial and early radiation source, which leads to other daughters that are also radioactive.

Refabrication of ThO_2–$^{233}UO_2$ Fuel

The ORNL and B&W Co. performed the major development work in this area. The B&W Co. designed and operated an unshielded direct-fabrication glove box line for the preparation of ThO_2–$^{233}UO_2$ reactor feed materials [18]. The objective was to perform rapid operations immediately after removal of the daughter ^{228}Th from ^{233}U. Thoria with 3 % $^{233}UO_2$ was prepared by the sol–gel process for fabrication of fuel elements [19]. The pilot plant processed several hundred kilograms of irradiated thoria and ^{233}U fuel. The typical sol–gel run was 2 days in the pilot plant and with this form of operation, the dose received by the personnel was found to be below the accepted limits. The B&W pilot plant [18] manufactured a typical BWR ThO_2–3 % $^{233}UO_2$ fuel assembly as a demonstration (Fig. 2). The full-size fuel rods were assembled under water with the sol–gel fuel vibratory-compacted in clad tubes and welded on top. The

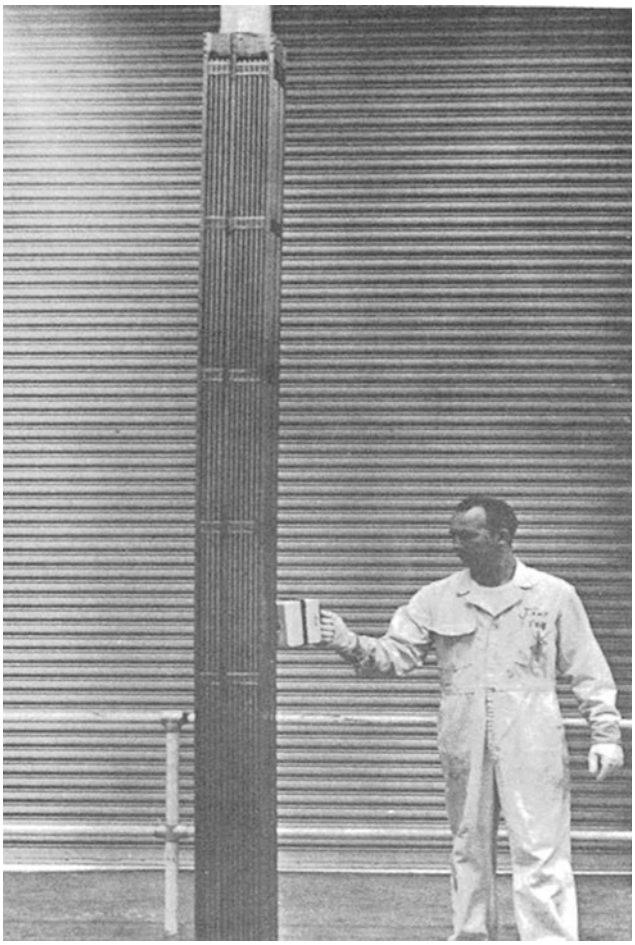

Fig. 2 Finished fuel assembly produced during demonstration program (withdrawn rapidly from pool for quick survey of gamma activity)

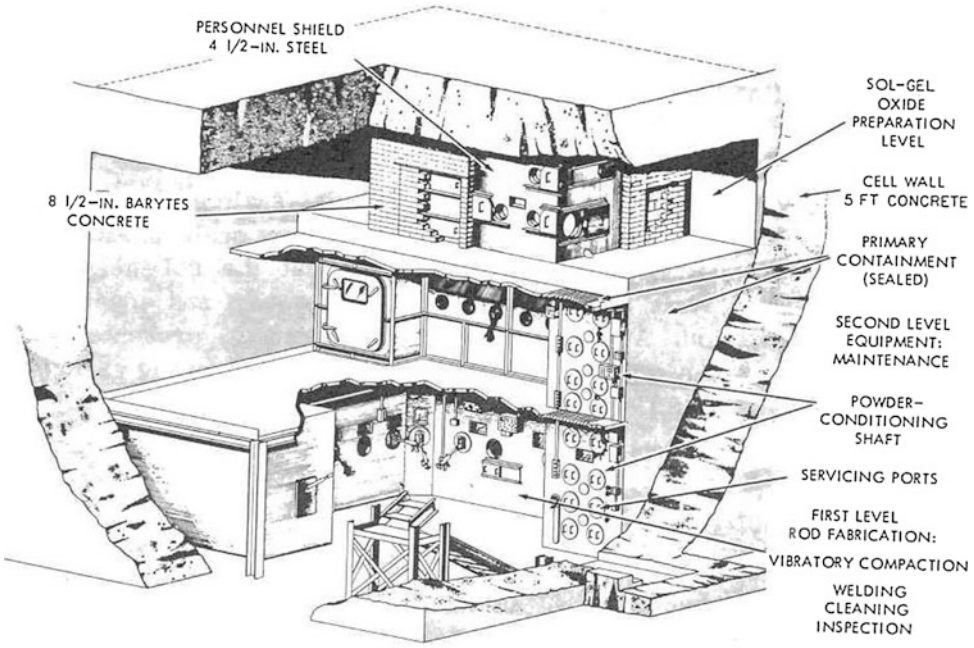

Fig. 3 The KILOROD facility at Oak Ridge National Laboratory

assemblies were stored under water. The contamination level of ^{232}U in ^{233}U was ~42 ppm. It was estimated that with removal of ^{228}Th and with rapid fabrication processes, the limit of toleration for ^{232}U in ^{233}U could be up to 1000 ppm for the pilot plant operation within the acceptable personnel exposures for a 5-day operation at a capacity of 50 kg of ThO_2–3 % $^{233}UO_2$ per week. Of course, the acceptable limits for personnel exposure may have been much higher then than current recommendations.

The ORNL constructed a shielded semi-remote-operation pilot plant called the KILOROD facility [20, 21], where 1000 fuel rods containing ThO_2–3 % $^{233}UO_2$ were manufactured in an 8-month-long operation using the reprocessed ^{233}U and thorium oxides after removal of ^{228}Th. The KILOROD facility employed the same techniques as those employed by the B&W Co., that is, (1) employing the sol–gel process to create particles and (2) using the vibratory-compaction method to manufacture the fuel rods. The 1000 rods were later employed at the Brookhaven National Laboratory (BNL), Upton, New York, USA, to perform exponential lattice experiments to obtain the criticality and other neutron physics data for validation of the reactor physics codes employing the then-available cross section data for ^{232}Th and ^{233}U. The rods were handled in a regular manner at BNL, that is, without any special protective measures against exposure or contamination.

The shielded cell of the KILOROD facility [21] is shown in Fig. 3. ORNL also performed a scaling analysis [20] on the data they obtained from the KILOROD facility to determine design and cost parameters for conceptual ThO_2–$^{233}UO_2$ shielded [22] and remote-operation fuel fabrication plants [23] having capacities of 60–3700 kg of heavy metal/day. If the plant is kept simple and the number of operations are minimized, the costs for 930 and 3700 kg/day plants, respectively, were estimated to be ~12 and 8 % larger than those for an equivalent unshielded plant.

Irradiation Experiences with Thorium-Based Fuels

ORNL conducted a long campaign of irradiating samples of sol–gel-derived pure and mixed components of thorium, uranium, and plutonium as oxides and carbides. The samples were assembled with vibratory compaction. The performance characteristics of the vibratory-compacted fuel were found to be as good as those of compressed and sintered pellets and burn-ups of greater than 100,000 MWd/tonne of heavy metal were achieved. Table 2 [24] shows a summary of the irradiations performed; some ThO_2–PuO_2 pins were also irradiated. The BNL sol–gel is the same fuel as that used for manufacturing the 1000 rods, that is, 3 % $^{233}UO_2$ mixed with 97 % ThO_2. Some rods were carried to 100,000 MWd/tonne burnup.

ThO_2–PuO_2 irradiations [25] were performed at the Pacific Northwest Laboratory, Richland, Washington, USA, and at the Institute for Transuranium Elements of the EU in Karlsruhe, Germany. Zircaloy-clad samples of ThO_2–PuO_2 were irradiated. These samples were not prepared by vibratory compaction, but by wet ball milling of the ThO_2 and PuO_2 powders and later sintering in hydrogen at 1600 °C. Very stringent irradiation testing was performed, generating power levels of 1 kW/cm (30 kW/ft or

Table 2 Summary of thorium fuel cycle program irradiation of powder-packed rods[a]

Designation	No. of rods	Type of oxide	Fuel density (% theoretical)	Linear heat rating (W/cm)	Peak burnup (MWd/ton of metal)	Objective
MTR group I	7	Arc-fused Sol–gel E	86–87	390	15,000–100,000	To provide base-line data to use in comparing sol–gel and arc-fused oxide
MTR group II[b]	2	Sol–gel S	88–89	600	100,000	To obtain higher heat rating by increasing enrichment
MTR group III[b]	6	Sol–gel 35	86–89	820	100,000	To compare oxide calcining atmospheres and higher heat ratings obtained by increasing diameter
ETR group I	4	Sol–gel 35	86–89	960	22,000	Same as for MRT group III
NRX group I	8	Sol–gel A & B Arc-fused	86–87	160	16,000	To provide base-line data
NRX group II	4	Sol–gel C	83–86	210	5000	To study effect of increased length
NRX group III	6	Sol–gel S	88–89	270	23,000	To study effect of increased length
NRX group III	3	Sol–gel ThO_2–PuO_2	74–76[c]	260	22,000	To study ThO_2–PuO_2 oxide and lower packed density
ORR loop	3	Sol–gel 26	84–85	500	2100	To study in pressurized water at 260 °C and 1750 psi
ORR poolside	2	Sol–gel D	85[c]	340	5000	To measure effective thermal conductivity by using a central thermocouple in Na–K at 540 and 750 °C and 315 psi
ETR group II[b]	6	BNL sol–gel	90	630	30,000–100,000	To study effects of remote fabrication and oxide recalcining
ETR group III[b]	7	Sol–gel ThO_2	88	770	10,000–70,000	To study ThO_2 blanket material with gradually increasing heat rating and provide high protactinium low-fission-product material for chemical processing

[a]All rods were clad with type 304 stainless steel except for ETR groups II and III and ORR loop, which were clad with Zircaloy-2
[b]Currently under irradiation
[c]Tamp-packed; all others vibratory compacted

100 kW/m) and maximum burn-ups of ~3 × 1020 fissions/cm^3 (~110,000 MWd/tonne). Some of the samples experienced center melting.

The experience with irradiation testing of the ThO_2–UO_2 and ThO_2–PuO_2 fuel samples was in general similar to that with UO_2, UO_2–PuO_2, and ZrO_2–PuO_2 fuels. The structures formed in the fuel were also found to be quite similar, that is, there was nothing unusual and, in fact, ThO_2 fuel could undergo burn-ups of 100,000 MWd/tonne.

First-of-a-Kind and Licensability Issues for the Thorium Fuel Cycle

Front-End Issues of the Thorium Fuel Cycle

Light Water Reactors with Uranium

The use of thorium together with uranium enriched to less than 20 wt% in light water reactors has been studied in some detail. For the breed and burn (no reprocessing) approach, heterogeneous designs have been studied more than homogeneous designs. One paper phrased the conclusion as "heterogeneous U–Th fuel provides higher neutronics potential than a homogeneous fuel" [26]. Another paper reported that a homogeneous mixture of uranium and thorium only becomes comparable to pure uranium at about 120 MWd/kg discharge burnup [27]. This is double the limiting discharge burnups presently allowed by western licensing authorities [28].

For homogeneous designs where thorium is mixed with enriched uranium in the fuel rods, thorium requires an increase in ^{235}U enrichment to maintain the same length of operating cycle. For the ^{235}U-enrichment level to stay below the proliferation limit of 20 %, no more than ¾ of the fuel can be thorium. To keep the ^{235}U-enrichment level below 10 %, no more than ½ the fuel can be thorium [27]. The reactivity coefficients and control worths can change significantly from those of the typical PWR depending on the thorium content and the desired discharge burnup [28]. It may be possible to design around some of these issues.

Most heterogeneous fuel concepts for thorium use are based on a design called the Radkowsky Thorium Fuel

(RTF), developed by Alvin Radkowsky and his associates [29, 30], which derives ultimately from the US Navy's LWBR seed-blanket concept [6]. In the original RTF design, the seed and blanket comprised a single PWR assembly, the removable seed being the center part and the blanket the outer part (cf. Fig. 1). A variant called the Whole Assembly Seed Blanket (WASB) uses separate seed and blanket assemblies, but is functionally similar. The WASB design (see Fig. 4) may raise fewer safety and licensing questions related to the mechanical operation of the seed and blanket components.

In either case, the seed would be replaceable at normal PWR intervals of 4–6 years, but the blanket would remain in-core for 8–12 years, allowing time to fission much of the ^{233}U bred there. Reactivity coefficients for typical seed–blanket cores show modest differences from those of typical PWRs.

Either variant is subject to the same licensing issues [26]:

- Both seed and blanket have to achieve burnups well beyond those justifiable from present experience and would undoubtedly require irradiation tests before use to ensure adequate fuel integrity, adequate fission gas retention, and limited clad corrosion;
- Because the blanket is essentially subcritical and the seed is supercritical, the seed power density is much higher than that of the blanket. This requires a non-standard design for the seed fuel pins (both trefoil and annular have been proposed), which would have to be irradiation tested and licensed;
- Because the seed power density is much higher than that of the blanket, the design has to ensure greater water flow through the seed to cool it properly. Although studies have shown how such designs might be achieved, their

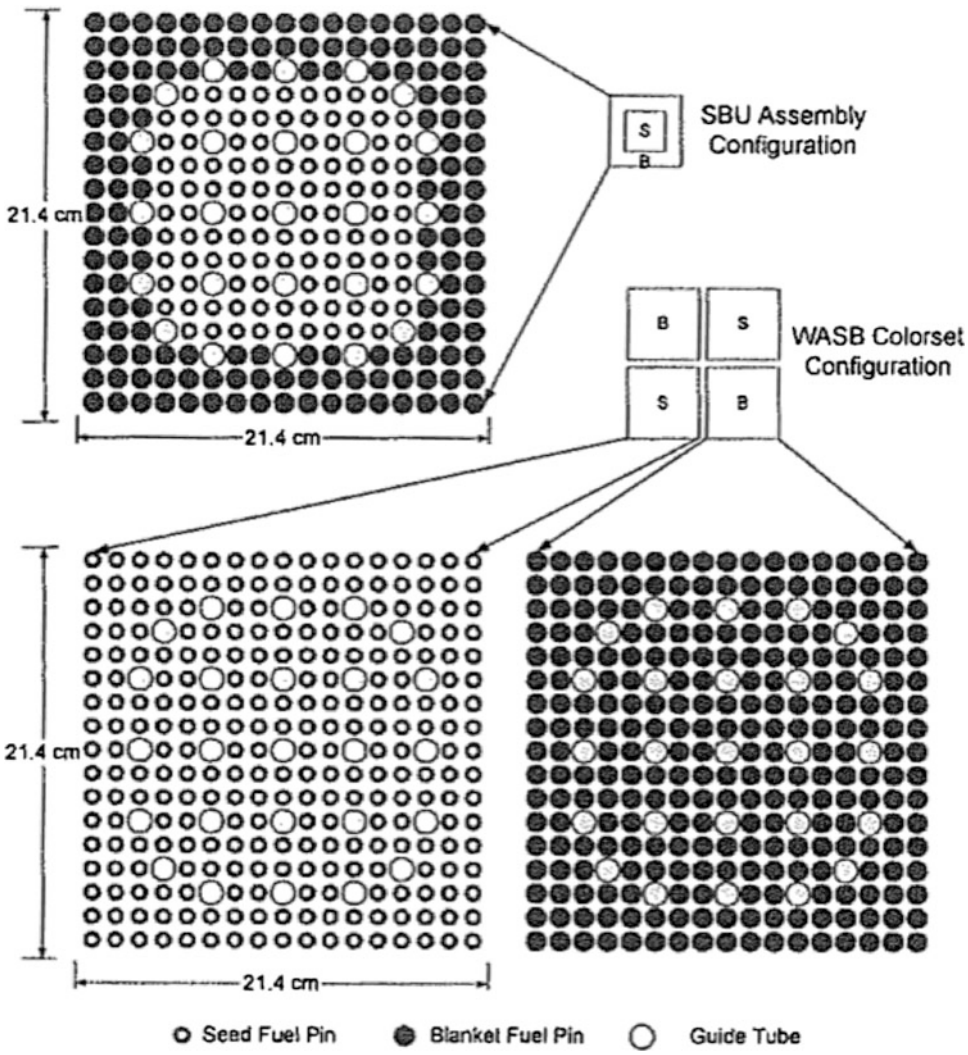

Fig. 4 Layout of seed–blanket unit (SBU) and WASB fuel assembly designs

use in the open lattice core of a PWR leads to questions about how cross flow might affect the core response to off-normal and transient conditions;
- Compared with typical PWRs, seed–blanket reactors reduce the effectiveness of control rods and of soluble boron used to control reactivity and to ensure that the reactor can be shut down.

Light Water Reactors with Plutonium

Either weapons-grade (WGP) or reactor-grade plutonium may be utilized in place of medium-enriched uranium as the fissile material in a thorium reactor. WGP is derived from surplus nuclear weapons, and therefore consists almost entirely of the ^{239}Pu isotope, which has a larger fission cross section than ^{235}U. Reactor-grade plutonium arises from reprocessed fuel obtained from any kind of power reactor that uses low-enriched uranium (as most do), and typically contains only 50–70 % of the ^{239}Pu isotope, the remainder consisting of diminishing fractions of the higher plutonium isotopes 240, 241, and 242.

Homogeneous thorium–uranium PWRs with reactor-grade [31] or weapons-grade plutonium [32] would contain largely thorium oxide, with up to 15 % plutonium oxide, and in some cases up to 15 % natural uranium to denature the resulting ^{233}U. The reactivity coefficients of these cores have the same sign as do those of typical PWRs, but differ in magnitude by as much as a factor of two. The effectiveness of control poisons may be reduced by a factor of two or three. Using either thorium or plutonium tends to decrease the worth of reactor control materials. Using both at once exacerbates the problem. There have been some suggestions that a heterogeneous seed–blanket arrangement can mitigate this effect [26, 33, 34]. Most importantly, the delayed neutron fraction is less than 0.3 % [35–37], compared with a minimum of just under 0.5 % in the typical PWR.

Heavy Water Reactors

For thorium-use CANDU reactors, fuel designs have been developed for breed and burn cycles (known in heavy water reactors as the OTT, once-through thorium) and for cycles that recycle ^{233}U after reprocessing. Early CANDU designs had a positive void coefficient, which has been eliminated by adding a neutron-absorber rod to the center of each bundle. Use of thorium rods in the bundle tends to have the same effect owing to the greater thermal absorption of thorium compared to that of ^{238}U. Plutonium addition to the bundle tends to make the coefficient more positive, as the hardening of the spectrum results in greater contribution of the plutonium fission resonances. Thus, the combination of thorium with the plutonium is particularly appropriate.

As usual, thorium leads to generation of ^{233}U, the subsequent fissioning of which tends to reduce the delayed neutron fraction. When both recycled ^{233}U and plutonium are used, the resulting delayed fraction is about 0.3 % [33, 36, 37].

In present-day CANDU reactors, typical discharge burnups are less than 20 MWd/kgHM. When using thorium, either in the breed and burn or recycle mode, burnups of 30–40 MWd/kgHM are envisaged, and some designs propose burnups on the order of 70 MWd/kgHM. These are all substantially beyond present-day experience with uranium fuels. A few thorium bundles have been tested for CANDU reactors.

Back-End Issues of the Thorium Fuel Cycle

Clearly, the knowledge base on the thorium fuel cycle is very old and not all documentation may be available. The question to ask is, “is the available documentation sufficient to envisage a cautious plunge into developing thorium fuel cycle facilities?” An ancillary question is, “how much development will be needed for preparing the case for licensing the fuel reprocessing and fuel refabrication facilities?”

It should be recalled that the knowledge base on the uranium–plutonium fuel cycle was non-existent when the first reprocessing facilities were constructed in the USA during the Second World War and later in other countries. The fuel refabrication facilities were built later, however, at present, plutonium recycling is being employed regularly in France and some other countries as well. Thus, the state of knowledge on thorium fuel reprocessing and refabrication is not so limited that development cannot proceed as it did for the uranium fuel cycle.

It is not imperative to have the thorium reprocessing and fuel refabrication facilities for the initial introduction of the thorium fuel cycle, just as in the uranium fuel cycle, where so far most countries have not reprocessed their spent fuel. Only a handful of nuclear-power countries have started to employ recycled plutonium fuel in their LWRs. The penalty associated with the once-through thorium fuel cycle is much larger than that for the uranium fuel cycle; however, it would be outweighed by the benefits of reprocessing and fuel refabrication once the thorium fuel cycle is closed, as ^{233}U is a more valuable fuel for use in LWRs than ^{239}Pu.

We believe that the first-of-a-kind technical issues associated with developing the thorium fuel cycle are surmountable. There is sufficient information available to remain on the right track. The thorium fuel cycle is more difficult and complicated than the uranium fuel cycle and will require shielded facilities and remote or semi-remote operation. Thus, it would be more expensive. However, developing the fuel cycle facilities would be tantamount to a Generation IV type effort, with similar time spans for development and deployment. The related major issue is that

of development costs, which would have to be justified on economic and political grounds.

Feasibility and Desirability of Employing the Thorium Fuel Cycle for Nuclear Power Generation

Feasibility

The previous sections have described the experience gained with thorium fuel in its use in reactors and in reprocessing and refabrication operations. Most of the experience base is in the USA and is very old. Several technical advantages and disadvantages were identified for the use of Th-based fuels for nuclear power generation.

Advantages:

- The ^{233}U converted from thorium is the best thermal spectrum fuel. It is also better than ^{235}U in the fast spectrum, but not as good as ^{239}Pu;
- ThO_2 is a better compound than UO_2, with better conductivity and greater stability. Perhaps it can go routinely to a burnup of 100 GWd/tonne;
- ^{232}Th has six less neutrons (and protons) than ^{238}U. It will produce shorter-lived actinides than ^{238}U. Thus, waste volumes will be smaller and not as long-lasting;
- Thorium is an excellent fuel for burning Pu in thermal and fast reactors. In fast reactors it helps to provide a negative void coefficient;
- The disadvantage of producing ^{232}U, which emits hard gamma rays, along with ^{233}U, becomes an advantage in terms of resistance to proliferation;
- The disadvantage of high decay heat in the Pu produced, if medium-enriched uranium is employed to start the Th cycle, also becomes an advantage owing to the detrimental effect of decay heat on a weapon;
- Much ^{233}U formed can be burned in situ if burnup lengths of 75–100 GWd/tonne can be employed in thermal reactors;
- Thorium is more abundant and more wide-spread than uranium. The price of ThO_2 may not escalate as much as that of U_3O_8.

Disadvantages:

- Natural thorium has no fissile isotope. It also has a very high threshold for fast fission and low value of fast fission cross section;
- Thorium irradiation produces ^{232}U, which emits a hard gamma ray with its decay. Fabrication of new fuel after reprocessing of ^{233}U becomes difficult;
- Thorium enriched with medium-enriched uranium will produce Pu containing a greater fraction of ^{238}Pu and ^{242}Pu, which embody higher decay heat. It is a handicap for further use in reactor system or for disposal;
- Thorium in the reactor produces protactinium, which decays to ^{233}U with a half-life of 27 days. Thus, the ^{233}U build up after reactor shut down has to be watched and greater shut down requirements will be needed for almost 3 months;
- ^{233}U and ^{239}Pu provide a Beta of approximately 0.3 % versus approximately 0.5 % provided by ^{235}U. Thus, the reactivity transients will be faster;
- The control rods and soluble boron worths are decreased with thorium in the thermal system as the fuel is blacker and competes better with the control rods and the dissolved boron;
- The seed–blanket configuration, which may impart better exploitation of the advantages of thorium in the thermal (or even in fast) systems, is also prone to the thermal hydraulic disadvantage of requiring flow orificing and different rod geometry or configuration;
- The use of medium-enriched uranium ($\sim$20 % ^{235}U) to mix with thorium has been criticized on proliferation grounds as the separative work needed to convert it to weapons-grade ^{235}U is rather small.

All of the above advantages and disadvantages have been known for many years. The disadvantages can be designed around, as has been demonstrated. The disadvantage of ^{232}U in the back end of the thorium fuel cycle has also been accounted for by the use of rapid operations or remotely controlled shielded facilities. Clearly, the cost of reprocessing and refabrication will be greater than those for the uranium fuel cycle that allows the recycle of plutonium in thermal reactors. A compensation for these costs may be possible due the greater reactivity and burnup that may be achieved with ^{233}U–Th fueled LWRs or PHWRs. One could make the argument that the feasibility of thorium as a reactor fuel has been demonstrated on the reactor scale and the feasibility of reprocessing the Th-based fuel and refabricating it with the formed ^{233}U has been demonstrated on the pilot scale. It may be difficult, but there are no roadblocks.

Desirability

Past Situation

It is clear that the desirability argument for employing the thorium fuel cycle for large-scale nuclear power generation is not very compelling; otherwise, this fuel cycle would not have been abandoned in the 1980s. At that time, it did not make economic sense to consider the thorium fuel cycle for

LWRs and PHWRs with its need for greater ^{235}U enrichment. There was also the firm belief at that time that the LWR fuel will be reprocessed and the Pu produced would be used in fast reactors, essentially allowing an unbounded nuclear power growth scenario, and that there would be no pressure on the supply of uranium.

Current Situation

The current situation is that LWRs are the 'King'—they are the main-stay of providing the large-scale nuclear power that is needed in the world due to the demand in the emerging economies and due to global warming concerns. The increase in nuclear power deployment and the need to assure supplies for each plant for 60–80 years will, undoubtedly, put pressure on uranium supply and prices. The predominant use of the once-through fuel cycle, and the deployment and use of only limited amounts of Pu, are not helpful either. Employing the thorium fuel cycle, which can provide a back-up to the large-scale consumption of uranium, may be prudent for the long-term economy of nuclear power. The reduced waste burden (if reprocessing becomes common) of the thorium fuel cycle will also be a long-term gain.

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MYRRHA: A Flexible and Fast-Spectrum Irradiation Facility

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Abstract

MYRRHA (Multipurpose hYbrid Research Reactor for High-tech Applications) is a multipurpose research facility currently being developed at SCK•CEN. MYRRHA is based on the ADS (accelerator-driven system) concept, where a proton accelerator, a spallation target, and a subcritical reactor are coupled. MYRRHA will be able to work in both critical and subcritical modes, enabling the facility to target a broad range of R&D applications. In this paper, the most recent developments of the design of the MYRRHA facility are presented, as well as the irradiation performance of MYRRHA.

Introduction

MYRRHA (Multipurpose hYbrid Research Reactor for High-tech Applications) is the flexible experimental accelerator-driven system (ADS) in development at SCK•CEN. MYRRHA is able to work both in subcritical (ADS) and in critical modes. In this way, MYRRHA aims to target the following applications:

- to demonstrate the full ADS concept by coupling the three components (accelerator, spallation target, and subcritical reactor) at reasonable power levels to allow operation feedback and scalability to an industrial demonstrator;
- to allow the study of the efficient technological transmutation of high-level nuclear waste; in particular, minor actinides that require high and fast flux intensity;
- to be operated as a flexible fast-spectrum irradiation facility allowing for:
  - fuel developments for innovative reactor systems;
  - material developments for GEN IV systems and fusion reactors;
  - radioisotope production for medical and industrial applications;
  - industrial applications, such as Si-doping.

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MYRRHA started from the ADONIS project (1995–1997), which was the first project at SCK•CEN that studied the coupling between an accelerator, a spallation target, and a subcritical core. ADONIS was a small irradiation facility, having the production of ^{99}Mo as its single objective. In 1998, the ad hoc scientific advisory committee recommended extending the purpose of the ADONIS machine to become a material testing reactor for materials and fuel research in order to study the feasibility of transmutation of minor actinides and to demonstrate the principle of the ADS at a reasonable power scale. Since 1998, this project has been called MYRRHA.

In 2005, MYRRHA consisted of a proton accelerator delivering 350 MeV × 5 mA to a windowless spallation target coupled to a subcritical fast core of 50 MW_{th}. This 2005 version is the "MYRRHA–draft 2" design [1]. This 2005 design was used as the starting point for the FP6 EUROTRANS integrated project [2], which resulted in the XT-ADS [3] (the eXperimental demonstration of the technical feasibility of Transmutation in an Accelerator-Driven System) design, where a linear proton accelerator delivers a 600 MeV × 3.2 mA beam into the spallation target. The reactor power of XT-ADS was 57 MW_{th}.

The XT-ADS design became the starting point for the work performed in the FP7 CDT project [4], which resulted in the MYRRHA-FASTEF (MYRRHA Fast-Spectrum Transmutation Experimental Facility) design. The MYRRHA design has now entered into the Front End Engineering

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Design (FEED) phase, where a consortium of engineering companies has been selected to support the FEED. The current design of MYRRHA-FASTEF is described later in this paper, as well as the irradiation performances of the facility.

The MYRRHA ADS

The MYRRHA Accelerator

The accelerator is the driver of MYRRHA as it provides the high-energy protons that are used in the spallation target to create primary neutrons, which in turn feed the subcritical core. In the current design of MYRRHA, the machine must be able to provide a proton beam with an energy of 600 MeV and an average beam current of 3.2 mA. The beam is delivered in continuous wave (CW) mode. Once a second, the beam is shut off for 200 μs so that accurate on-line measurements and monitoring of the subcriticality of the reactor can take place. The beam is delivered to the core from above through a beam window.

Accelerator reliability is a crucial issue for the operation of an ADS. A high reliability is expressed by a long Mean Time Between Failure (MTBF), which is commonly obtained by a combination of over-design and redundancy. On top of these two strategies, fault tolerance in the high-energy section of the linear accelerator (above 17 MeV) must be implemented to obtain the required MTBF. Fault tolerance allows the accelerator to recover the beam within a beam-trip-duration tolerance after failure of a single cavity. In the MYRRHA case, the beam–trip-duration tolerance is 3 s. Within an operational period of 3 months, the number of allowed beam trips exceeding 3 s must remain under ten; shorter beam trips are allowed without limitations. The combination of redundancy and fault tolerance should allow a MTBF value in excess of 250 h to be obtained so as to meet the required number of beam trips per 3-month operation cycle.

At present, proton accelerators with megawatt-level beam power in CW mode only exist in two basic concepts: sector-focused cyclotrons and linear accelerators (linacs). Cyclotrons are an attractive option with respect to construction costs, but they lack any modularity, which means that a fault tolerance scheme cannot be implemented. Also, upgrading the beam energy is not a realistic option. A linear accelerator, especially if superconducting, has the potential for implementing a fault tolerance scheme and offers high modularity, resulting in the possibility to recover the beam within a short time and to increase the beam energy.

The Core and Primary System

The main components/systems of the current MYRRHA-FASTEF design are of the same MYRRHA/XT-ADS type defined in the EUROTRANS project, only increased in size. The primary and secondary systems have been designed to evacuate a maximum core power of 100 MW_{th}. All the MYRRHA-FASTEF components are optimized for extensive use of the remote handling system during component replacement, inspection, and handling.

As MYRRHA-FASTEF is a pool-type ADS, the reactor vessel houses all the primary systems. In previous designs of MYRRHA, an outer vessel served as secondary containment in case the reactor vessel leaked or broke. In the current design, the reactor pit fulfils this function, improving the capabilities of the reactor vault air cooling system. The vessel is closed by the reactor cover, which supports all the in-vessel components. A diaphragm inside the vessel acts to separate the hot and cold lead–bismuth eutectic (LBE) plenums, to support the In-Vessel Fuel Storage (IVFS), and to provide a pressure separation. The core is held in place by the core support structure, consisting of a core barrel and a core support plate. Figure 1 shows vertical cut sections of the MYRRHA-FASTEF reactor with its main internal components.

In the present state of design, the reactor core (Fig. 2) consists of mixed oxide (MOX) fuel pins, typical for fast reactors. A major change with respect to the previous version of the core is the switch from a windowless loop-type spallation target to a window beam tube-type spallation

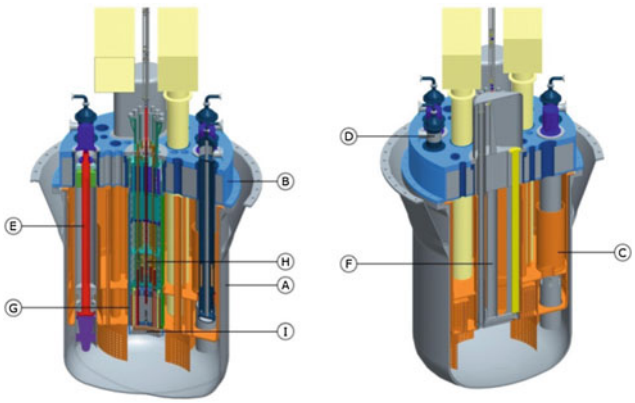

Fig. 1 Section of the MYRRHA-FASTEF reactor. **a** Reactor Vessel. **b** Reactor Cover. **c** Diaphragm. **d** Primary heat exchanger. **e** Primary pump. **f** In-vessel fuel handling machine. **g** Core. **h** Above core structure. **i** Core restraint system

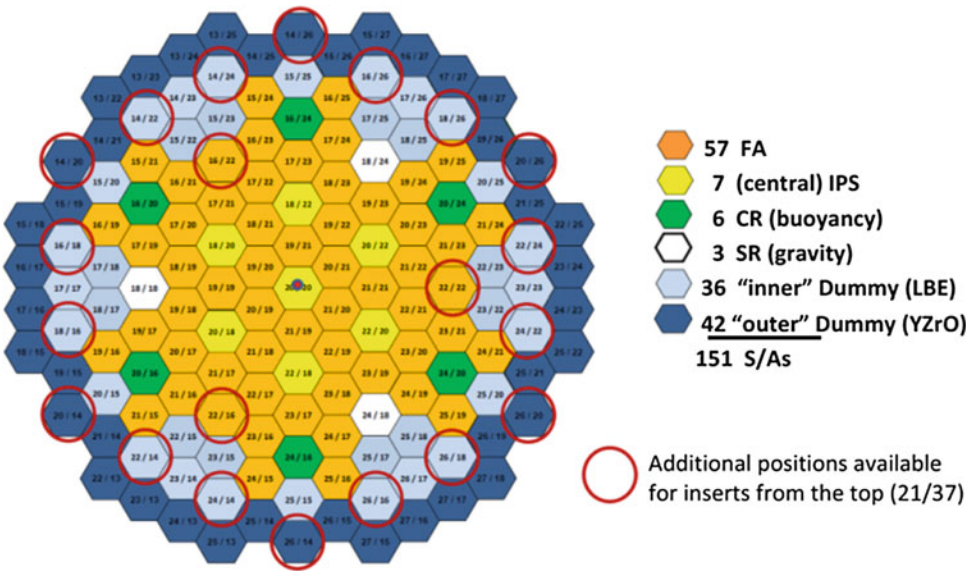

Fig. 2 Section of the MYRRHA-FASTEF core

target. The previous version needed three central hexagons to house the spallation target, whereas the present design only needs one central hexagon. To better accommodate this central target, the fuel assembly's size is slightly increased compared with the MYRRHA/XT-ADS design. Consequently, the In-Pile test Sections (IPS), which are located in dedicated core positions, (Figs. 2 and 3), are larger in diameter, which results in more flexibility for experiments. Thirty-seven positions can be occupied by the IPS, by the spallation target (the central one of the core in the subcritical configuration), or by control and shutdown rods (in the core critical configuration). This gives significant flexibility in the choice of the most suitable position (neutron flux) for each experiment.

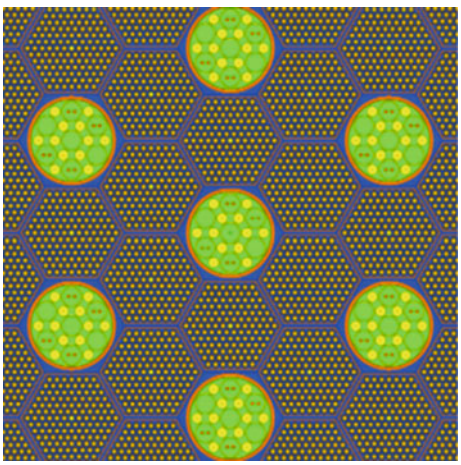

Fig. 3 Horizontal cross-section of the critical core, focusing on the seven IPS positions

The required high, fast flux intensity has been obtained by optimizing the core configuration geometry (fuel rod diameter and pitch) and maximizing the power density. We will be using, for the first core loadings, 15-15Ti-stabilized stainless steel as the cladding material instead of T91 ferrite-martensitic steel, which will be qualified progressively further on during MYRRHA operation for a later use. The use of LBE as coolant permits the lowering of the core inlet operating temperature (down to 270 °C), decreasing the risk of corrosion and allowing the core ΔT to be increased. This, together with the adoption of reliable and passive shutdown systems, will enable the high, fast flux intensity target to be met.

As depicted in Fig. 2, which shows a critical core layout (with seven central IPS) at the equilibrium of the fuel cycle, 37 positions are available for Multi-Functional Channels (MFC) that can host equally:

- fuel assembly and dummy, loaded from the bottom (in all 151 positions);
- IPS, control and scram rods, loaded from the top.

In subcritical mode, the accelerator (as described in the previous section) is the driver of the system. It provides the high-energy protons that are used in the spallation target to create neutrons, which in turn feed the subcritical core. The accelerator is able to provide a proton beam with energy of 600 MeV and a maximum current of 4 mA.

In subcritical mode, the spallation target assembly, located in the central position of the core, brings the proton beam via the beam tube into the central core region. The spallation heat deposit is dissipated to the reactor primary circuit.

The spallation module guarantees the barrier between the reactor LBE and the reactor hall and insures optimal conditions for the spallation reaction. The spallation module assembly is conceived as an IPS and is easily removed or replaced.

The primary, secondary, and tertiary cooling systems have been designed to evacuate a maximum thermal core power of 110 MW. The 10 MW over the nominal core power account for the power deposited by the protons, the power of the in-vessel fuel, and for the power deposited in the structures by γ-heating. The average coolant temperature increase in the core under nominal conditions is 140 °C with a coolant velocity of 2 m/s. The primary cooling system consists of two pumps and four primary heat exchangers (PHX).

The primary pumps deliver the LBE to the core with a mass flow rate of 4750 kg/s (453 L/s per pump). The working pressure of the pump is 300 kPa. The pump is fixed at the top of the reactor cover, which is designed to be the only supporting and guiding element of the pump assembly.

The secondary cooling system is a water cooling system and the tertiary system is an air cooling system. These systems function in active mode during normal operation and in passive mode under emergency conditions for decay heat removal.

The main thermal connection between the primary and secondary cooling systems is provided by the primary heat exchangers. These heat exchangers are shell and tube, single-pass, and counter-current heat exchangers. Pressurized water at 200 °C is used as the secondary coolant, flowing through the feed-water pipe in the center of the PHX to the lower dome. All the walls separating the LBE and water plena (feed-water tube, lower dome, and upper annular space) are double walled to avoid pre-heating of the secondary coolant and to prevent water leaking into the LBE in case of tube rupture.

Irradiation Performance of MYRRHA

As MYRRHA was conceived as a flexible fast-spectrum irradiation facility, many and diverse possibilities exist to irradiate materials in MYRRHA. Material irradiations can be performed in In-Pile Sections (IPS) positioned around the central core or at the central core position below the spallation target when operating in subcritical mode.

Figure 2 shows a typical MYRRHA reactor core layout with the different fuel and dummy assemblies, control and safety rods, seven central IPS (yellow assemblies), and additional IPS positions. In total, thirty-seven positions can be occupied by IPS. All IPS positions are dedicated irradiation positions that have a penetration through the reactor cover, so they are accessible from the top. Each IPS has a usable irradiation volume of about 4900 cm^3.

As a fast spectrum is available throughout the entire core, standard fuel assemblies can be replaced by experimental assemblies for material irradiations under experimental conditions determined mainly by the reactor conditions. Given that all safety constraints are met, such experimental assemblies can replace, in principle, any fuel assembly inside the core. In this situation, however, irradiation temperature conditions are linked to the reactor conditions.

When working in subcritical mode, a special irradiation position below the spallation target inside of the central dedicated "beam window" IPS is present. Due to the spallation process of 600 MeV protons on the LBE fluid target, a more energetic spectrum than the standard fast spectrum is created and allows for material irradiation conditions particularly suited for fusion materials research. The irradiation temperature conditions inside this dedicated IPS and their control are determined by the design of the IPS.

Material Irradiation Performance in the IPS

A preliminary design has been made for Charpy (impact resistance) samples. In critical mode, seven IPS of this type are loaded to estimate their irradiation conditions (Fig. 3). In subcritical mode, the central IPS is replaced by the spallation target, resulting in six occupied IPS positions.

The basic parameters that characterize the irradiation conditions are the thermal power of the core, the proton beam current when operating in subcritical mode, the neutron flux, and induced displacements per atom (dpa) in the IPS. These basic parameters can be found in Table 1.

Irradiation Conditions Below the Beam Window

When MYRRHA is working in subcritical mode, a higher energy spectrum can be found below the spallation target due to the spallation reaction. This higher energy spectrum will induce higher dpa and appm He/dpa levels closer to the irradiation conditions for some components in fusion applications.

The spallation target is housed in the central position of the core. The beam window is located in the top half of the assembly. The volume available for irradiation can be found under the beam window and is about 3 L.

The dpa and He production for the volume below the beam window have been calculated separately for both neutron- and proton-induced damage. Finally, the combined appm He/dpa ratio has been calculated (see Fig. 4).

Figure 4 indicates that total dpa values of 50 dpa/EFPY can be reached in a volume of about 0.4 L, whereas peak values of more than 70 dpa/EFPY can be obtained in a volume of about 0.15 L. The volume available for

Table 1 Main core results for critical and subcritical modes

	Critical	Subcritical
#IPS	1 + 6	0 + 6
Core power (MW_{th})	100	94
Neutron flux (10^{15} n/cm^2 s)		
Φ_{tot} (central pos.)	2.93	4.63
Φ_{tot} (6 IPS)	2.62	3.33
dpa (central position)		
dpa/EFPY	23.5	–
appm He/EFPY	14.1	–
appm He/dpa	0.6	–
dpa (6 IPS)		
dpa/EFPY	20.7	28.0
appm He/EFPY	12.4	10.7
appm He/dpa	0.6	0.4

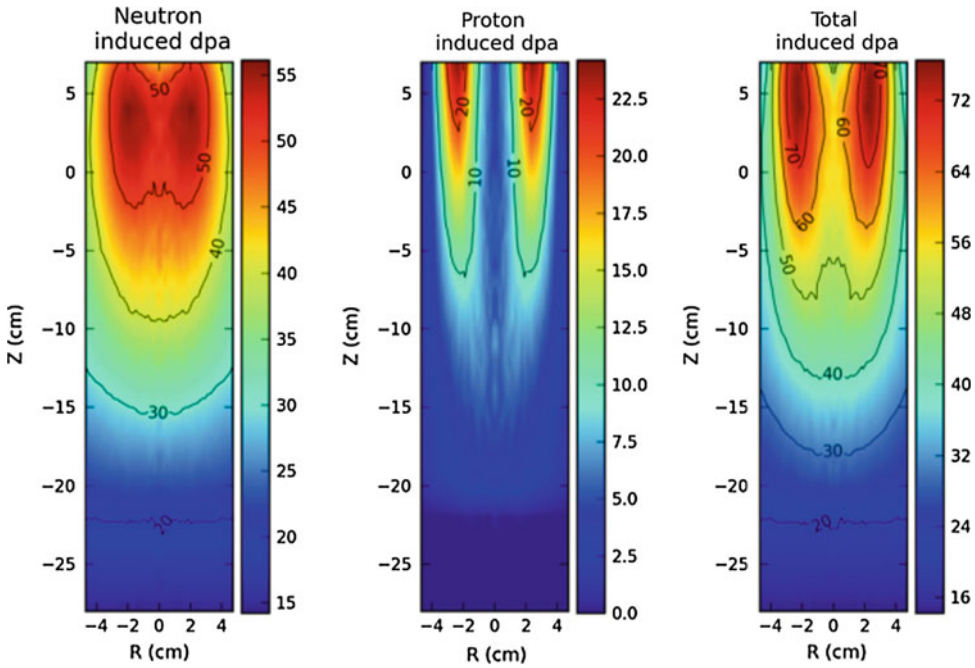

Fig. 4 dpa/EFPY below the beam window

irradiations of at least 20 dpa amounts to about 2.3 L. The figure clearly shows the impact of the Gaussian shape of the proton beam profile.

The same dependence is even more evident in Fig. 5, which shows He production. This He production is largely dominated by proton-induced He production.

Finally, Fig. 6 shows the appm He/dpa ratio, important for fusion irradiations. From the figure, one can see that we can reach a minimum level of 12 appm He/dpa in a volume of 1 L.

Based on these irradiation performances, SCK•CEN developed a module that can be inserted in the central position of the core, below the proton beam window, in the spallation zone to allow material experiments under these representative conditions. As a result, the sample specifications that can be met simultaneously during irradiation are:

- sample surface temperature: 100–650 °C;
- ΔT over the sample: <30 °C;
- dpa levels: up to 30 dpa/y;
- appm He/dpa: up to 20 appm He/dpa;
- volume: about 50 samples, typically 3–4 cm length and with a diameter of 7 mm.

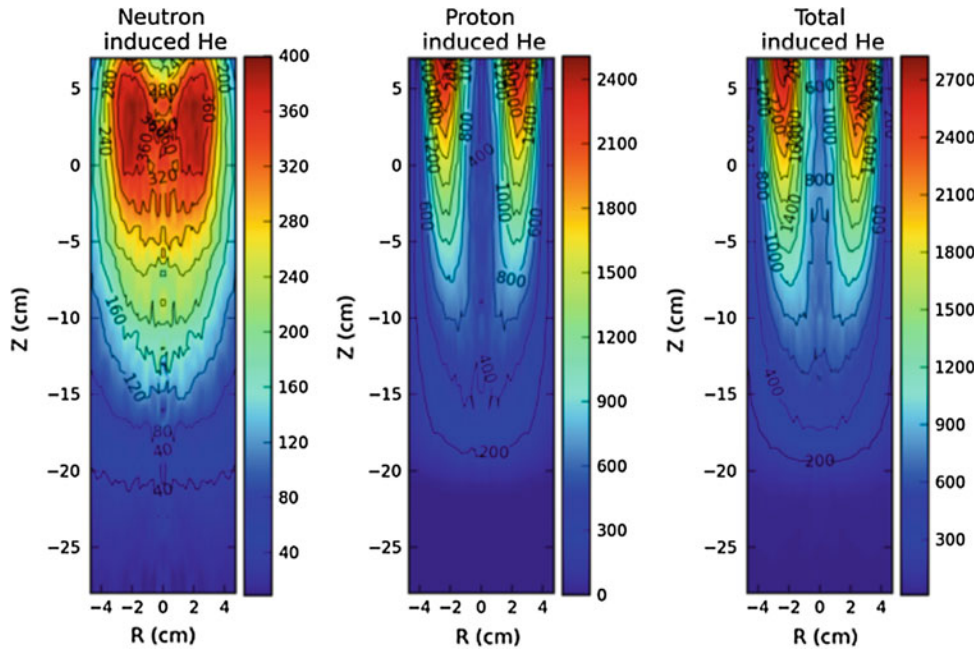

Fig. 5 He production (appm He/EFPY) below the beam window

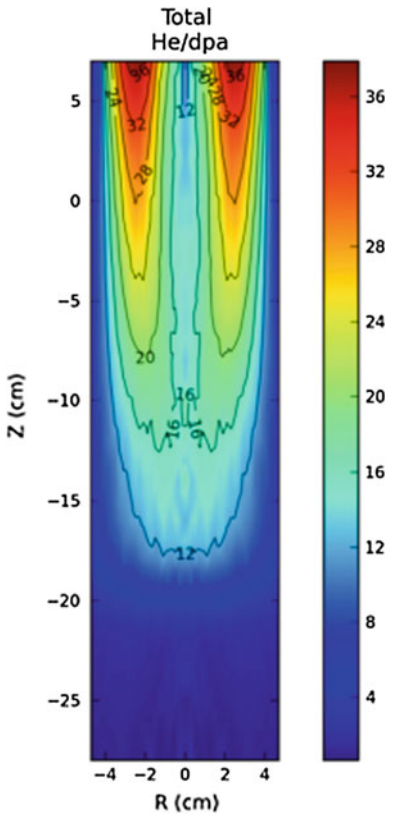

Fig. 6 appm He/dpa ratio below the beam window

Use of MYRRHA for the Development of Fusion Materials

The MYRRHA-IMIFF facility will be able to provide support for many tasks specified in the R&D programs on fusion technologies [5], including:

- screening tests for development of radiation-resistant materials;
- qualification of materials under a broad range of temperature and irradiation conditions—temperature-controlled conditions can be achieved in the In-Pile Sections in the core and for the irradiation module below the beam window;
- generation of experimental data for the development of Codes and Standard procedures for the selected materials.

MYRRHA should be able to help investigate and resolve the following issues:

- swelling of various materials, including the baseline candidate FM steel under fusion representative conditions (irradiation damages of the material matrix accomplished by formation of transmutation helium and hydrogen);

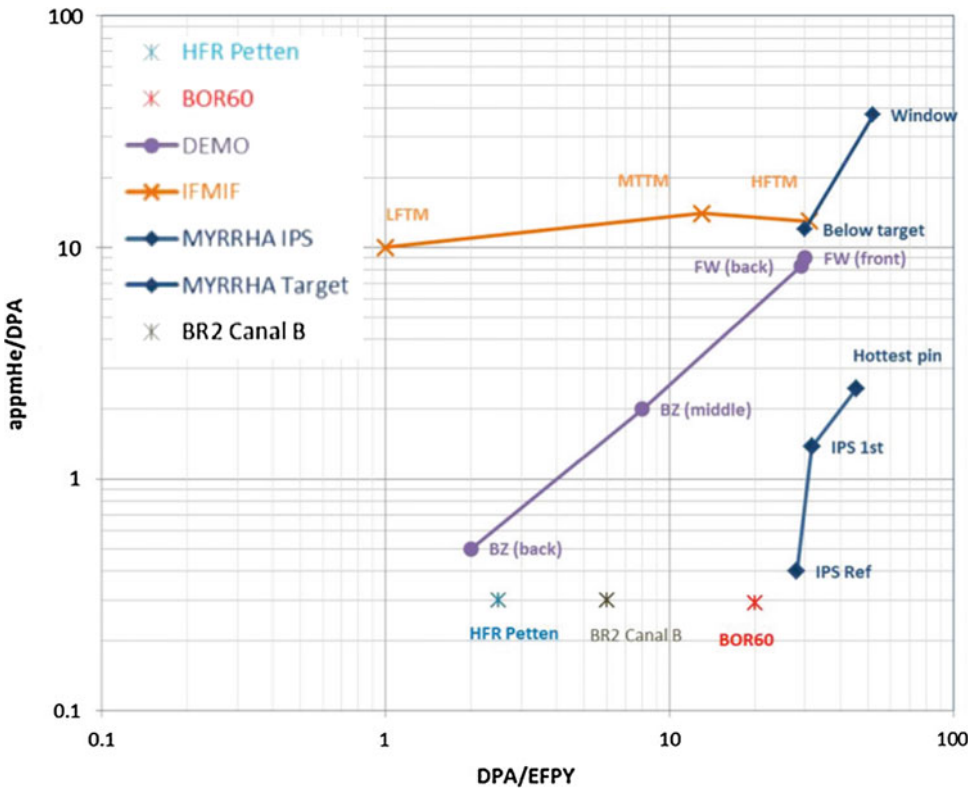

Fig. 7 DPA and He production in DEMO, IFMIF, and MYRRHA: Estimated damage induced in DEMO and proposed irradiation conditions in IFMIF and MYRRHA

- low-temperature irradiation embrittlement of the candidate materials;
- high-temperature helium embrittlement and effects of transmutation helium on creep properties;
- corrosion under irradiation;
- other irradiation effects, which might be identified at a later stage.

Due to these advantages it will be possible, in a reasonable time, to irradiate specimens with dimensions sufficient for reliable mechanical characterization, which is required both for screening experiments and for the production of material properties for design codes. Flexibility in the helium production rate should help with the investigation and understanding of both the combined effect of dpa and helium in the low-temperature range and the helium embrittlement at high temperature and help to develop viable mitigation strategies.

The graph in Fig. 7 shows the different irradiation damage expected in DEMO together with the irradiation conditions reached in IFMIF, MYRRHA-IMIFF, HFR, BR2, and BOR-60. All non-MYRRHA data can be found in [2].

The figure clearly shows that MYRRHA, by combining irradiations in the IPS positions and below the target, provides good irradiation conditions to simulate the damage induced in DEMO.

Conclusion

As MYRRHA was conceived as a flexible fast-spectrum irradiation facility, many and diverse possibilities exist to irradiate materials in MYRRHA. Material irradiations can be performed in In-Pile Sections, in experimental assemblies, and below the spallation target in subcritical mode.

MYRRHA as an irradiation facility has the advantages of the high flux of fast neutrons and thus a high ratio of damage dose accumulation, a wide range of appm He/dpa ratios under the spallation target, and big irradiation volumes. Due to these advantages, it is possible to irradiate specimens with dimensions sufficient for reliable mechanical characterization that is required both for screening experiments and for the production of material properties for design codes during a reasonable timescale.

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Part III

Invited Speakers: Innovative Thorium-Reactor Concepts

The Thorium Fuel Cycle: Past Achievements and Future Prospects

Dominique Greneche

Abstract

This presentation aims to provide a broad overview of the thorium-based nuclear fuel cycle, which can be considered as a complement or even a substitute for the classical uranium nuclear fuel cycle in the operation of nuclear reactors. We first examine the incentives that led to numerous studies in the past and that still justify the continuation of work on this topic around the world. For that purpose, we describe and explain the main physical and nuclear properties of thorium as well as those of the fissile isotope that is generated by the thorium, uranium-233, as this is its major advantage. We also give a summary of the available data on terrestrial thorium reserves. We present an overview of the history of the development of the thorium cycle, including the acquired experience of its use in nuclear reactors. From this basis, we explain the various problems that may arise from using the thorium cycle on an industrial scale, from the front end of the fuel cycle to the final stage of spent fuel reprocessing and recycling of materials and, vis-à-vis, the waste associated with this cycle. Finally, we discuss some generic issues related to implementation of the thorium cycle, in particular, the issues of non-proliferation and economy. A conclusion and summary of the advantages and disadvantages of the thorium cycle and its development prospects is also presented.

Reasons for Considering Thorium as a Fuel for Nuclear Reactors

Historical Perspective

From the early days in the development of nuclear energy, thorium has been considered a potential fuel that could possibly supplement or even replace the natural uranium that was feared to be very scarce in the Earth's crust. Indeed, the "New Pile Committee" created in April 1944 in the USA to explore a variety of reactor concepts recommended that "more work should be done on the nuclear development of thorium because of its greater availability" and the Committee also suggested experiments to develop reactors that would convert thorium to uranium-233. It is worthwhile to mention that members of this Committee numbered some of the most eminent physicists and chemists working in the area of nuclear reactors, and in particular, three Nobel Prize Laureates in physics. Among them, the initial idea of "thorium converters" was submitted in early 1944 by Eugène Wigner (Nobel Prize Laureate 1963) as an option for making the atomic bomb using uranium-233 as a fissile material. The proposal of using thorium for nuclear reactors was then further developed by Phil Morrison, another distinguished scientist, in the framework of the New Pile Committee. It is important to remember that at that time, nuclear reactors were an entirely new technology; the first man-made chain reaction had occurred less than two years before, on December 2, 1942, in Chicago. Knowing this, it is perfectly legitimate to say that the use of thorium was considered as an option for the fuel of nuclear reactors since the birth of nuclear energy.

However, thorium cannot be a real alternative to uranium, at least in the short term, because unlike the latter, it has no "fissile" isotope with slow neutrons (also called "thermal"

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neutrons), but it does allow a way of simply generating uranium-233 (U-233) by neutron capture in a nuclear reactor. This artificial uranium isotope, thus created, is relatively stable (half-life of 160,000 years) and is an excellent fissile isotope for thermal neutron reactors. It is even better than uranium-235 (U-235) or plutonium-239 (Pu-239), and this is why the thorium cycle has always been of significant interest and has been studied worldwide.

The Place of the Thorium Cycle with Regard to the Uranium Cycle

Unlike natural uranium, thorium has only one isotope in its natural state, thorium-232 (Th-232), which is not fissionable with slow neutrons. However, it is a fertile isotope, because when it absorbs a neutron, it generates uranium-233 (U-233) via Th-233 (half-life = 22 min) and Pa-233 (half-life = 27 days).

A chain reaction can clearly not be sustained with only Th-232, unlike with natural uranium, which contains 0.711 % fissile U-235. Therefore, a fissile material (U-235, U-233 or plutonium) must be added to thorium to make a fuel for a nuclear reactor. In this case only, the thorium material can generate U-233, which, like plutonium, can be "burned" in situ or recycled. This is what is called the thorium cycle.

So we see that this cycle is not really an alternative to the classic uranium cycle, but a possible supplement to this cycle, at least in the foreseeable future. Indeed, it is first necessary that sufficient quantities of fissile material are available for this generation process of U-233 from thorium to be initiated and developed on a large scale, provided that the residual U-233 content in spent fuel is recycled (as is done in France for plutonium in the uranium cycle). However, the process can be accelerated by the design of reactors called "breeder reactors", which produce globally more fissile material (U-233) than is consumed during an irradiation cycle in the reactor. It is only by meeting this condition that one could imagine an almost total replacement of the uranium cycle by the thorium cycle in the long term.

Uranium-233 (U-233)

For fission induced by thermal neutrons, U-233 has a higher neutron yield per neutron absorbed compared with U-235 or Pu-239. Indeed, the average number of fission neutrons produced by the absorption of a thermal neutron (called the "eta" factor, usually denoted η) is typically 2.27 for U-233 in a pressurized water reactor neutron spectrum compared with 2.07 for U-235 and 2.11 for Pu-239. This is a major advantage of the thorium cycle: the high value of η makes U-233 the best fissile isotope in the range of thermal neutrons of all existing fissile isotopes. With such a value, it is theoretically possible to achieve breeding in today's thermal reactors with a thorium–U-233 cycle. Indeed, having η neutrons available after one fission, one of them must necessarily be absorbed in another fissile nucleus to maintain the chain reaction. Thus, there remains $(\eta - 1)$ neutrons available to eventually be absorbed into a fertile nucleus and produce in situ a new fissile nucleus. The breeder process (that is to say, the ability to produce an excess of fissile nuclei compared to those consumed) is possible only if $(\eta - 1)$ is greater than 1 (or $\eta - 2 > 0$). However, in nuclear reactor cores, one loses neutrons in various sterile captures or leakages and it is therefore necessary that the margin between $(\eta - 1)$ and 1 be sufficient. We see here that for slow neutrons, this margin is not sufficient for U-235 or Pu-239, but it is more comfortable for U-233. We note in passing that the situation is quite different for fast neutrons where plutonium is most suitable for enabling the fast neutron breeding process, which also explains why plutonium is the preferred fuel for fast neutron reactors. Let's recall here that the major advantage of being able to achieve breeding in thermal neutron reactors lies in the fact that these reactors have a much lower fissile inventory than that required for fast reactors.

It is only after the spent fuel is unloaded from the reactor that one can recover the residual U-233 formed in the reactor for possible recycling. This so-called "reprocessing–recycling" process is sometimes also called the "treatment–recycling" process and is similar to that which can be implemented for the uranium–plutonium cycle. Thorium, which has a very low consumption in reactors, can also be recovered and recycled in reactors.

However, the recycling operation of U-233 raises difficult technical problems related to the unavoidable presence of a certain amount of the isotope uranium-232 (U-232) mixed with U-233. U-232 is a radioactive isotope with a half-life of 72 years and has some descendants that are very strong gamma-emitters (and thus noxious). The main formation pathway of U-232 is a (n, 2n) reaction of U-233.

The decay chain of U-232 comprises Th-228, an α-emitter with a half-life of 1.9 years, followed by Ra-224, also an α-emitter with a half-life of 3.6 days. At this point, the chain joins that of Th-232 and ends with the stable Pb-208.

However, if uranium is chemically purified so that its radioactive decay products are removed, and if it is then rapidly manipulated to manufacture the U-233 bearing fuel, this operation can be performed in simple glove boxes lightly shielded without excessive occupational exposure to workers.

Physical Properties of Thorium

Thorium is a relatively soft and ductile metal, gray-white to silver when pure, but it oxidizes very rapidly in air to form ThO_2, which is dark in color. It has a density of 11.72 g/cm^3, significantly lower than that of uranium (19.1 g/cm^3). Conversely, its melting point is much higher than that of uranium metal: 1750 °C compared with 1135 °C for uranium. It is the same for the oxide, ThO_2, which has a melting point of over 3300 °C, significantly higher than that of 2800 °C for uranium oxide, making ThO_2 one of the most refractory materials that exist. The thermal conductivity of the metal is greater than that of uranium: 54 W/m K for thorium compared with 27.6 W/m K for uranium, which is almost double (these values are given at room temperature, but increase significantly with temperature). Thorium oxide also has a slightly higher thermal conductivity than that of UO_2. In both cases, the conductivity decreases with temperature. At 800 °C, it is about 5 W/m K for ThO_2 compared with 3–4 W/m K for UO_2 (values given in the literature for this parameter are quite disparate as they depend mainly on its intrinsic porosity, which itself depends on the fabrication process of the oxide). From the point of view of thermal properties, we see that the thorium cycle occurs more favorably than uranium, either as metal or oxide form (which is by far the most common form used for nuclear fuels). It is worthwhile to mention here that thermal properties are of particular importance for the choice of a fuel for nuclear reactors. Indeed, designers usually look for a fuel matrix that has a melting point as high as possible in order to ensure a sufficient margin between the maximum operating temperature at the center of the fuel element and the melting temperature of the matrix, especially in accident situations. In addition, the thermal conductivity determines the difference between the maximum temperature at the center of the fuel and its external border, close to the clad (or even in contact with it), which is essentially a function of the temperature of the coolant, which "wets" the clad.

Natural Abundance and Reserves of Thorium

Natural thorium is a relatively abundant element on Earth, with an average concentration of 7.2 ppm in the Earth's crust. This value is significantly higher than for uranium (2.5–3 ppm), which is the result of the very long half-life of Th-232 (1.41×10^{10} years) compared with 4.47×10^9 years for U-238. However, this does not mean that the exploitable reserves of thorium are two or three times higher than those of uranium, as many people say in the literature. In fact, because its use so far has been limited to the specific applications mentioned above, there has never been a comprehensive survey of thorium, so reliable estimates of world reserves of thorium are not currently available. The well-known "Red Book" of the International Atomic Energy Agency (IAEA)–OECD [6], which is updated periodically, included fairly detailed information on thorium resources until its 1981 edition. Since then, however, only aggregate data has been provided. For example, the edition published in 2009 shows a value of 3.616 million tons for the total global resource of thorium. This value comes from the addition of the so-called "identified" resources, the extraction of which would cost less than 80$/kg (US), estimated at 2.229 million tons, and these resources "prognosticated" without specifying the cost extraction, estimated at 1.387 million tons. There is also a more recent assessment by the US geological survey [7] that states "reserves" worldwide are estimated at 1.4 million tons of ThO_2.

To sum up, quantities of thorium that could be extracted from the ground under reasonably commercially viable conditions certainly amount to several million tons and are probably at least of the same order of magnitude as those of uranium. In any event, if a closed thorium cycle was deployed one day on a large industrial scale, thorium reserves would not in fact be a real problem because Th-232 is a fertile isotope, the transmuted portion of which can be easily recycled. Thus, a thorium cycle deployed with recycling of U-233 would be able to support the development of nuclear energy for hundreds or even thousands of years in a breeding mode.

Past Developments of the Thorium Cycle

Feedback Experience of Thorium Utilization in Nuclear Reactors

During the early development of nuclear energy for civilian purposes, in the 1950s and 1960s, a large number of ways to use thorium were studied, not only in the United States and the former USSR, but also in Europe and, to some extent, in Asia [8]. To illustrate the interest aroused by thorium in the early years of the development of nuclear energy, we should mention that the first international symposium dedicated to the thorium cycle took place in 1962 in the United States. The "proceedings" of this symposium are unfortunately no longer available, but those of the second symposium held in Gatlinburg, Tennessee, in 1966 can be obtained from the US Department of Energy (DOE) under the reference CONF–66025 or from the Library of Congress under reference 67-62083. In particular, in this document one can find detailed studies on the use of thorium fuel in different types of nuclear reactors.

The initial impetus for studies on the thorium cycle essentially came from the development of molten salt reactors (MSRs) in the late 1950s in the United States. These

reactors are indeed particularly suitable for the thorium cycle owing to the opportunity they offer to reprocess the fuel "online", which opens the way for continuous extraction of Pa-233 and gives rise to U-233 free of U-232, and enables recycling of U-233 as an always "online" process. This potential, combined with other favorable properties of MSRs, enables breeding with a "thermal" reactor, which is an attractive prospect in many ways (safety, economy, technology, sustainability), at least at the conceptual stage.

The most remarkable point to note, however, is that the first commercial light water reactors (LWR) developed in the United States (almost all nuclear power reactors are LWRs today), initially operated with a thorium-based fuel. These are the Elk River boiling water reactor (BWR), which started in 1963 just after the prototype BWR in Dresden (started in 1960) and the Peach Bottom pressurized water reactor (PWR), which started in 1967 just after the prototype of Shippingport (started in 1957). It is also remarkable that demonstration of the possibility of a LWR breeder was carried out precisely at the Shippingport reactor in the 1970s and 1980s using a U-233–thorium cycle [8]: the conversion factor reached was 1.0139 [9]. This was the only power reactor that used U-233 as the fissile material. It was thus demonstrated experimentally that it is possible to produce more U-233 than is consumed in a LWR. However, this performance was achieved at the cost of a sophisticated design of the reactor core, which would be difficult to extrapolate to commercial reactors, and to the detriment of other performances. From that time, many attempts to use thorium fuel in research reactors and power has been made, and significant experience has been accumulated on this type of fuel.

In the frame of this article, it is not possible to discuss all this experience and we refer the reader who wishes to deepen their knowledge of this aspect to [8–11].

Analysis of Incentives and Hindrances in the Historical Development of the Thorium Cycle

The main motivation for the development of thorium fuel was initially to provide a fuel cycle that can replace the uranium fuel cycle in anticipation of the expected rapid growth of nuclear power and the possibility of a natural uranium shortage. An additional incentive was the supposed abundance of thorium resources in nature, based on the fact that the average concentration in the Earth's crust is about three times that of uranium. In addition to that, in the mid-1970s, the price of uranium had reached a high level (around 40$/lb for U_3O_8) and this was then translated into a fear of a uranium shortage, which was accentuated by the fact that a large nuclear plant supplier was unable to meet its commitments to supply uranium to its customers. In addition to the abundance of thorium in nature and the prospect of fast breeder reactors that produce U-233, a number of other reasons advocate for the development of a thorium cycle, including:

- The absence of uranium resources in some countries with ambitious nuclear programs, such as India, which has large thorium resources identified in their soil, and whose access to imported uranium was limited for political reasons;
- The good neutronic properties and very good behavior under irradiation of thorium-based fuels.
- A lower initial excess reactivity (thus, reduced control means) with higher conversion factors (see below) with a uranium fuel, with designs tailored to meet these high conversion factors (gains on the order of 20–30 %).

It is useful to recall here that the conversion factor is a fundamental parameter that characterizes the ability of a nuclear reactor to produce fissile materials at the same time as it consumes it to produce energy. More specifically, this is the ratio between the *total* amount of fissile material produced in the reactor during a given period (for example, during an irradiation cycle at equilibrium) and the *total* amount consumed in the reactor during the same period.

In the early 1980s, several factors dimmed the enthusiasm for alternative cycles to uranium. First, interest in the nuclear option has weakened significantly, particularly in the United States, where public support for nuclear power has declined considerably after the accident at Three Mile Island in April 1979. This mistrust was then intensified in Europe after the Chernobyl disaster seven years later. Also in the early 1980s, and in conjunction with these events, the price of uranium fell to very low levels so the need to search for an alternative fuel became virtually redundant, despite the fact that use of thorium fuel has other benefits (which are discussed in this introduction). A third event that occurred in the late 1970s, was the banning of commercial processing of irradiated fuel by the administrations of US Presidents Ford and Carter for political reasons related to the risk of nuclear proliferation from fissile material separated during these operations. By the same logic, the use of uranium enriched beyond 20 % in civilian reactors was practically forbidden anywhere in the world. These decisions hindered the possible implementation of a thorium cycle for two reasons: firstly, promoters of the thorium cycle had to give up the reference cycle based on the use of highly enriched uranium (HEU) and replace it with uranium enriched to 20 % maximum (mid-enriched uranium, MEU), which complicates the implementation of the thorium cycle and reduces its overall performance; secondly, the ban on reprocessing the spent fuel negates the

possibility of recovering the U-233 content in spent fuel for recycling, at least in the United States.

In the last decade, however, there has been renewed interest in thorium-based fuels. This seems to have been initially motivated by the development of a fuel cycle deemed "proliferation-resistant" that was developed in the late 1990s by an American team led by Alvin Radkowsky [12]. We will not analyze, at this point, the significance of this concept in terms of non-proliferation, which appears questionable and is discussed further below. We will merely point out that it uses a reactor core that combines fissile zones loaded with low-enriched uranium (called "seed zones") and fertile zones containing a mixture of MEU (20 %-enriched uranium) and thorium (called the "blanket zone").

Another factor that boosted (slightly) the interest in the thorium cycle is the impetus for the development of nuclear energy in the world during the years 2000–2010, however, this has been somewhat reduced since the accident in Fukushima in 2011. In the wake of the expected growth of nuclear power worldwide, new questions have emerged here and there on the long-term availability of cheap uranium. Finally, new strengths emerged in favor of the thorium fuel cycle, including:

- The low production of plutonium and minor actinides by using thorium-based fuels, which is simply due to the position of the thorium element in the periodic table of Mendeleev, coming before that of uranium;
- The ability to "burn" excess plutonium in thermal reactors by implementing a thorium–plutonium cycle. Such concepts can also be imagined in very innovative reactors such as molten salt reactors (MSRs), accelerator driven systems (ADS reactors, also called "hybrid" reactors), and even as blankets in fusion-based energy systems [8];
- The transmutation of minor actinides;
- The possibility to reach a conversion factor greater than one, or at least close to one, with the use of thorium in some innovative thermal reactors such as advanced heavy water reactors (HWRs) or in more ground-breaking reactors such as MSRs, which is one of the concepts under study in the framework of fourth-generation reactors.

All these reasons means that there is currently a resurgence of interest in thorium in several academic and R&D institutions and also from certain industrial reactor designers and/or fuel vendors. One can add that in Japan, the high-temperature test reactor (HTTR) could be used in the future with thorium, as well as the HTR-10 in China. In addition, India is still considering thorium as fuel for industrial use in the not too distant future.

Present Status of Thorium Fuel Cycle Developments

Studies on the thorium cycle continue today in several countries, including the United States, Russia, China, Canada, Sweden, Norway, Japan, France, and especially India. The European Union is also active on the subject, but at a modest level. In fact, many of these works are carried out in the broader context of research on the molten salt reactors already mentioned above. This is particularly the case in France, where the CNRS (Centre National de la Recherche Scientifique) carries out research in the framework of European programs (EURATOM) and in collaboration with several countries (USA, Japan, Korea, Canada, Russia). A recent review of these studies can be found in [13].

However, most of these programs are mainly restricted to academic works that have limited use of the experiences industrially and that generate virtually no technological developments. The only exception is India, which has been pursuing a significant R&D program since the 1950s, which is described elsewhere in the ThEC13 proceedings.

From this brief overview of past and present studies on the thorium cycle, we can now summarize their main findings and we can analyze the advantages and disadvantages of this cycle, referring in particular to the industrial challenges of its implementation on a large scale.

Technical Characteristics and Industrial Challenges of the Thorium Fuel Cycle

The Front End of the Fuel Cycle

We distinguish two major phases of the front end of the fuel cycle, which are recovering natural thorium (ore mining, milling, and thorium concentration) and manufacture of thorium-based fuel for reactors.

Mining, Milling, and Thorium Concentration

We have seen that thorium has never been used on a massive scale for nuclear power reactors fuels. Only a few hundred tons were necessary for the production of fuels for the prototype reactors that we mentioned above. The regular supply needs of thorium are those that correspond to the specific non-nuclear applications, which are very limited. These requirements amount to a few tens of tons per year and they are declining. Under these conditions, there has never been any mining activities designed exclusively to recover thorium, and it is almost exclusively obtained as a by-product of mining rare-earth metals or possibly uranium when these two elements are present in the same deposit. Therefore, the

experience gained in this domain is incomplete. However, it allows us to draw the following conclusions.

The primary source of thorium mineral is a rare-earth phosphate of thorium called monazite, which is contained in some beach sand or alluvial deposits. If there was no need for the rare-earth industry, monazite would probably not be recovered for its thorium content. There are indeed other thorium ores with higher content than monazite itself, such as thorite, which are therefore more likely to be a mineable resource.

It is important to note here that the tailings resulting from ore processing are relatively less troublesome than those from the extraction of uranium in terms of their long-term management and their potential impact on the biosphere. This stems from the fact that the radioactive descendants of thorium are much shorter lived than those of the uranium isotopes. For example, Ra-228 from the decay of thorium-232 has a half-life of 5.74 years, whereas several descendants of U-238 have very long half-lives: 250,000 years for U-234, 75,000 years for Th-230, and 1600 years for Ra-226. In addition, potential problems from expelling radon gas in the case of uranium tailings are almost non-existent for thorium. Indeed, in the case of uranium, it is the Rn-222 isotope that has a half-life long enough (3.8 days) for it to remain radioactive even after migration through the soil to the surrounding atmosphere, whereas in the case of thorium, its counterpart, the Rn-220 isotope (also called thoron), has a half-life of 56 s and can be stopped by thin layers of soil (its descendants are solids).

In contrast, the treatment of thorium ore itself raises problems of radiation protection as the radiation is more difficult to control than in the processing of uranium ores. This is primarily due to a descendant of thorium-232, Tl-208, which is a powerful gamma-emitter (as discussed when we presented the decay chain of U-232). However, once the "pure" thorium is fully separated from its descendants, these problems no longer arise. One disadvantage is that the pure thorium returns to equilibrium with its descendants after a few decades, so it quickly becomes a radiating material and must be handled with some care.

One can find in [15] a detailed discussion of these issues of radiological protection and radioactive waste related to mining activities.

Let us note finally that there is already a globally equivalent of about 25,000 tons of ThO_2 contained in the extraction of rare-earth residues ([8], p. 46). Such a stock would enable the supply of tens of thorium-powered reactors if a thorium cycle were to develop industrially.

Thorium-Based Fuel Fabrication

The fabrication of thorium-based fuel on an industrial scale should not raise any major technical obstacles because enough experience exists on this subject, dating back to the 1960s. This experience covers a wide variety of fuels that have been made for different types of reactors, as it has been presented above. Note, however, that radiation protection precautions must be taken when handling large amounts of thorium, especially when it has been separated for several years (see [10] for more on the accumulation of radioactive descendants). Moreover, thorium in powder form is pyrophoric and can cause explosions when dispersed as dust.

Some thorium fuel irradiation programs continue today, in particular for mixed oxide thorium–plutonium fuels (MOX) in light water reactors. This type of experimental fuels were manufactured and irradiated in the BWR of Lingen (270 MW_e gross) in Germany and in the PWR of Obrigheim (350 MW_e gross), also in Germany, within the European framework research programme on the recycling of plutonium in nuclear power reactors. In addition, these experiments showed very good behavior of these fuels under irradiation, even better than that of uranium oxide fuels. However, the burnup achieved in these experiments was relatively small (<40 GWd/ton) and new qualification programs of these fuels would be needed to reach the much higher burnup of today's water-cooled reactors (that is, 50–60 GWd/ton and beyond for some options).

To conclude on this point, it is clear that the basic technical knowledge exists to develop industrial processes for the fabrication of thorium-based fuels. However, complementary R&D programs would be required to qualify such processes for routine industrial manufacturing.

Characteristics and Behavior of Thorium-Based Fuels in Nuclear Reactors

General Neutronic Characteristics

From a neutronic point of view, thorium-based fuels have a number of advantages over uranium-based fuels, in particular regarding the following features:

- Reactivity drop of the reactor core during one cycle (that is, between the two fuel reloads). It is slower for the thorium cycle due to the fact that U-233, which is gradually formed during the cycle, has a fission yield (η factor) significantly higher than the plutonium isotopes (taken together) formed by U-238 in the uranium cycle. This results in less requirements for reactivity control means for a thorium-based fuel cycle than for a uranium-based fuel cycle;
- Heterogeneity of power in the core. The absorption cross sections of the three main isotopes of plutonium (Pu-239, Pu-240, and Pu-241) have large resonances at very low energy, which makes the neutronic behavior of fuels containing significant quantities of plutonium more complicated. Consequently, plutonium, which builds up

with irradiation, strongly changes the relative absorption of different heavy nuclei as the fuel depletes, in part because of the presence of these plutonium resonances. This leads to significant spatial variations in power (peak power) in adjacent fuel that does not have the same burnup, which is usually the case when refueling is fractionated (batch reloads). This is even truer when plutonium comes to coexist in the same core as uranium fuels or MOX fuels. All these complications are significantly attenuated with thorium fuels as they generate U-233, the neutron absorption cross sections of which are much less erratic for slow neutrons (energy <1 eV) due to the absence of large resonances in this energy range (the first major resonance for U-233 is at 1.8 eV);

- Temperature coefficients. Detailed analysis of the effects of temperature on the reactivity (that is, the neutron-multiplication factor) is fairly complex. The term-to-term comparison of each of these effects between a thorium-based fuel and a uranium-based fuel is beyond the scope of this article, the more so, as the magnitude and even the sign of some of these effects can vary widely with the type of reactor and with the fuel composition and its evolution, not to mention the fuel temperature itself. We therefore only indicate here that the main difference between a thorium fuel and uranium fuel relates to the neutron spectrum hardening effect, when the moderator temperature increases. This is because this effect strongly depends on cross-section changes for slow neutrons and, as we have seen, these variations are very different for the U-233 and plutonium isotopes due to resonances of the plutonium isotopes in this energy range. Let us simply point out that this effect appears more favorable for thorium-U233 fuels than for uranium–plutonium fuels, which facilitates the control of phenomena related to changes in power level and the mastering of some safety-related neutronic parameters;
- Yield of fission products. The yields of most of the high neutron capturing fission products, which contribute significantly to the neutron poisoning of the thermal neutron reactor core, are significantly lower for the fission of U-233 than for the fission of the other two important fissile isotopes of the uranium cycle, namely U-235 and Pu-239. This is particularly the case for xenon-135, which is by far the most troublesome fission product. The difference in fission yields with U-235 is about 20 % and it reaches almost 60 % for Sm-151 with Pu-239. This lower neutron capture by fission products is beneficial for several reasons, including lower loss of reactivity during fuel irradiation (and thus, the ability to increase the fuel burnup), better neutron economy, which favors the process of converting fertile nuclei into fissile nuclei, and reduction of constraints related to the xenon effect (poisoning balance management, power transients management, spatial deformation of neutron flux, etc.).

However, there is one drawback of thorium-based fuels that must be mentioned here. It arises from the fact that production of U-233 by neutron capture in Th-232 is through protactinium-233 (Pa-233), the half-life of which is relatively long at 27 days, compared with its counterpart in the uranium cycle, Np-239, the half-life of which is only 2.3 days (it produces Pu-239). This is a point to note when considering the possibility of using thorium reactors. Indeed, this relatively long period of decay leads to a relatively high concentration of Pa-233 in the reactor core, and thus the rate of neutron capture can be significant, especially as the capture cross section of Pa-233 itself is quite high; it is about 40 barns for thermal neutrons at the reference speed of 2200 m/s. This may be a penalty for thorium cycle reactors with high thermal neutron flux as the process of neutron capture by Pa-233 makes waste through the formation of a U-234 nucleus by the decay of Pa-234 (the Pa-233 nucleus disappears by capture before decaying into U-233). A simple calculation shows that it is only above a neutron flux of the order of 10^{14} n/cm^2 s that the penalty becomes significant.

Fuel Behavior Under Irradiation

Thorium is always used in the form of its oxide (ThO_2) or carbide (ThC_2) for nuclear reactor fuels (except, of course, for molten salt reactors, where it is in the form of a fluoride). These fuels have a better behavior under irradiation than uranium oxide and that allows higher burnup for thorium-based fuels. This is a consequence of the higher melting point and better thermal conductivity of ThO_2 or ThC_2 compared to UO_2.

Nuclear Material Management from Thorium Fuel

The once-through thorium cycle (that is, without recycling the residual fissile materials) would require, of course, an external supply of available fissile material, that is, either U-235 in the form of enriched uranium at an appropriate level or plutonium with adequate isotopic composition. In this case, one should seek the highest possible combustion rate (accompanied by an increase in the split of the core) to produce and "burn" the maximum in situ U-233 generated by the thorium, knowing, nevertheless, that the overall benefit of this process in terms of consumption of existing fissile material remains limited. However, one should especially aim for the highest possible conversion factors. In this regard, we have already noted that the conversion factors achieved with a thorium cycle are always higher than those obtained with a conventional uranium–plutonium cycle. This is, for example, the case in light water reactors for which the conversion factor may be around 0.7 (or even higher for

specific concepts) with thorium fuel although it is only 0.6 or so with uranium. In other reactors, such as heavy water reactors, it can be even higher with values close to 0.8 or even up to 0.9, or close to 1 for some concepts. With high-temperature reactors, one can achieve similar conversion factors by using thorium. Finally, we must mention MSRs, with which it is theoretically possible to achieve iso-generation or even breeding (conversion factor >1), owing to the online extraction of Pa-233 and to the presence of certain capturing fission products. This is a really attractive option for the implementation of a thorium cycle as it opens the way for a system of thermal breeder reactors that have a significantly lower fissile inventory than fast neutron reactors and in which the raw fissile material becomes practically inexhaustible.

Numerous studies have been devoted to the use of thorium in thermal reactors as various combinations of fuel cycles are possible by combining different types of reactors so that they act as symbiotic systems. It has been found that thorium can be mixed with four types of fissile material:

1. Highly enriched uranium (>90 % U-235), which gives a cycle referred to as "Th–HEU". This was the reference fuel for HTRs in the 1970s in the United States and Germany but, today, the use of this fuel is excluded due to the strict restrictions related to nuclear proliferation risks because HEU can be used directly and through a relatively simple process to make an atomic bomb;
2. Medium-enriched uranium (20 % U-235), which gives a cycle referred to as "Th–MEU". This leads to a cycle that is sometimes called "denatured" in the sense that the uranium cannot be used directly in the manufacture of an atomic bomb. Such an option might be considered as a "last resort" because it leads to complicated materials management insofar as it combines all the constraints associated with the materials management of the uranium and thorium cycles (natural uranium, thorium, enriched uranium, reprocessed uranium that still contains a high proportion of U-235 (about 10 %), plutonium, minor actinides, etc.)
3. Plutonium, whatever its origin and isotopic composition. This is simply called the "Th–Pu" cycle. This cycle is sometimes viewed as the best option for the use of thorium as it offers an interesting alternative to the MOX fuel system for the recycling of plutonium produced by standard uranium-based fuels. It is also considered as a means to "burn" excess plutonium stockpiles.
4. Uranium-233, where it is available in large quantities after processing of spent thorium-based fuel. This is the "Th–U-233" cycle.

The results of these studies show that the use of thorium in "conventional" thermal neutrons reactors allows a global economy of the use of uranium from a few tens of percent to a maximum of about 80 % when the equilibrium of the system is achieved. The results depend a lot on the combination of reactor type and recycling options used in the studies.

Regarding the use of thorium in fast neutron reactors (FNRs), several studies have been carried out, particularly in Russia for the BN-800 reactor, but also in France and Europe. They have demonstrated the ability to achieve iso-generation of fissile material or even breeding with a Th–U-233 cycle. However, the performances achieved in this area are lower than those obtained with a uranium–plutonium cycle, mainly for reasons related to the neutronic best qualities associated with plutonium from uranium-233 for fast neutron spectra. Another reason is that the fission cross section of thorium itself for fast neutrons is much smaller than that of U-238 (about three times lower). The use of the thorium cycle in FNRs is thus not very attractive, although some proponents of the thorium cycle see some benefits of such use, in particular, because the reactivity sodium void effect is much less positive than that of uranium–plutonium cores.

Overall, if thorium were to be heavily used in thermal neutron reactors with a closed cycle (that is, with recycling of U-233), it is hoped that the overall resources of fissile material could be increased by a factor of two or maybe more in the long term, provided that there are sufficient amounts of natural uranium to fuel the reactor park over a long period. Let us recall, however, that if fast neutron reactors were to be developed intensively with "classical" uranium–plutonium cycles, the energy potential of natural uranium resources would be multiplied by a factor of 50–100. In this case, the use of the thorium cycle as a complement to this system would again increase this already enormous energy potential by a factor of two or more, depending on the thorium resources available.

The Back End of the Fuel Cycle

Fuel Reprocessing

We have seen that to take full advantage of the assets of the thorium cycle, it is necessary to reprocess the spent fuel to extract the residual U-233 and then to recycle it in nuclear reactors. At the same time, this option allows recycling of thorium itself, the consumption of which in reactor cores is very low (typically a few percent depending on the type of reactor and associated fuel cycle).

Experience on the reprocessing of thorium-based fuels is very limited but it is not zero. In fact, it has existed since the late 1940s in the United States, which processed about 900 tons of thorium fuels, allowing for the separation of nearly 1500 kg of U-233. This was also the case for the fuel of the experimental reactor at Indian Point, which consisted of a mixed oxide of thorium and highly enriched uranium. The reprocessing of 16 tons of fuel enabled the recovery of 1.1 tons of a mixture of different isotopes of uranium, which in particular contain 7 % U-233 and 56 % U-235 [15]. Some other countries, such as India, have also conducted experiments in this field, but on very small amounts of fuel ([10], p. 220 and [16]).

Research and development works, as well as small-scale experiments, have shown that the treatment of thorium-based fuels is less technically simple to perform than treatment of uranium-based fuels; this is mainly due to the dissolution step. Thorium is much more difficult to dissolve than uranium, either in the form of the metal or its oxide. Historically, it was the Oak Ridge National Laboratory in the United States that developed a hydrometallurgical process called Thorex for thorium-based fuel reprocessing, which was derived from the Purex process still in use today for the reprocessing of uranium-based fuels. One can find in Sect. 6.2 of [8], a good description of this process, which is based on the addition of fluoride to the acid dissolution solution. We will only mention here that this acid is extremely aggressive towards materials (due to the fluoride ions). Therefore, a chemical inhibitor must be added to this solution to reduce the corrosion of stainless steel process equipment. Knowing that the use of fluorine (as HF) is incompatible with zirconium alloys, this is the material of choice for use in reprocessing facilities such as the La Hague plant in France. In addition to the complications resulting from the implementation of this type of process, it must be noted that it generates more secondary waste than the standard Purex process (+50–70 % as described in [8]).

To overcome these drawbacks, alternative non-aqueous processes to the Thorex process have been studied as part of the volatilization of fluorides or electro-refining processes, but these studies have not produced very good results and they were interrupted prematurely.

Once the dissolution has been performed, the steps of chemical separation and purification of materials for the thorium cycle should not be fundamentally different from that of a uranium–plutonium cycle. Only the management of an additional element, the thorium itself, could possibly add some difficulties, albeit without making the process too complicated.

In conclusion on this point, it appears that the development of reprocessing processes for thorium-based fuels that are commercially viable still need significant R&D efforts. It should be noted, however, that such efforts have been made in the past to improve reprocessing processes and technologies for uranium fuel and these efforts have allowed us to achieve considerable progress in the efficiency and effectiveness of these methods, including in terms of the management of waste generated in these operations.

U-233 Recycling

We have already discussed the problem of radiation that arises from the inevitable presence of U-232 mixed with U-233 because some descendants, such as Tl-208, are strong gamma-emitters (alone, Tl-208 contributes to 85 % of the total dose of radiation emitted after 2 years by the descendants of the U-232). This requires the U-233 to be handled remotely behind shielding after a few months, which greatly complicates the processes of the fabrication of fuels with U-233 in terms of technology. Although it is perfectly possible with the techniques now available, it is clear that this leads to significant additional manufacturing costs for these fuels.

One possibility to cope with this problem is to reduce the formation of U-232 in the nuclear reactor, which could be achieved through various specific adaptations for different types of reactors. For example, this is the case for the production of U-233 in thorium "blankets" for fast neutrons reactors, as envisaged in the second phase of the program in India. Indeed, gradually as one moves away from the active core by entering into the blankets, the main route of formation of U-232 by (n, 2n) reactions is much reduced.

Final Disposal of Radioactive Waste

For a given amount of "thermal" energy (that is, the energy produced in a reactor core), the amount of fission products is the same in all fuel cycles as one fission releases always the same energy whatever the fissile isotope is, and it gives birth to two fissions products. Only the spectra of these fission products, that is, the relative proportion of each of the radioisotopes, may be different depending on the origin of the fissions (U-235, Pu-239, Pu-241, U-233). Consequently, the evolution over time of the global radioactive inventory of these products is a little different between the two cycles, but it does not matter much as the radioactivity of these fission products virtually disappears after a few centuries. Consequently, there is almost no difference between thorium- and uranium-based fuels with regard to this part of the nuclear waste.

The real difference between the uranium and thorium fuel cycles actually comes from the quantities of "minor actinides" generated in each case. Indeed, in the case of the uranium cycle, significant amounts of three long-lived transuranic elements are generated: neptunium-237 (Np-237, with a half-life of 2.14 million years), americium (mainly as Am-241, with a half-life of 432 years, and Am-243, with a half-life of 7380 years), and curium (mainly as Cm-245, with a

half-life of 8530 years). However, these radioelements are particularly radiotoxic α-emitters and they contribute to almost all of the global radiotoxic inventory (GRI) of the ultimate radioactive waste beyond a few hundred years (excluding plutonium, which is supposed to be recycled in nuclear reactors). Nevertheless, they are poorly soluble in water and they have a very low mobility in geological media (provided that these media are under reducing conditions). This practically excludes the possibility that they can migrate to the biosphere, at least in a term during which they would still have a certain radioactive noxiousness for living species. Still, for the sake of minimizing the risk, the partitioning of some or all of these elements and then the transmutation of them in nuclear reactors is an option that is under consideration and study today. Research has been conducted in this direction for many years in several countries, such as the United States, Japan, and France. This has led to great strides in the knowledge in this area, which suggests that these operations could be carried out industrially today. However, this would be at the price of implementation of complex processes and sophisticated technologies, and therefore this would entail important, additional costs.

With uranium-233, almost none of these minor actinides are produced. Indeed, americium and curium mainly come from plutonium (via the decay of various radioactive isotopes), whereas Np-237 mainly comes from U-235 (via three successive neutron captures in U-235, U-236, and U-237, which decays in Np-237). However, a Th–U-233 fuel produces other long-lived radionuclides (excluding isotopes of thorium and uranium, which are supposed to be recycled). The chief of these is the minor actinide Pa-231, essentially formed from (n, 2n) reactions of Th-232. This isotope of protactinium (half-life 32,760 years) is therefore involved in the long-term GRI significantly. Nevertheless, in a Th–U-233 fuel, one can find also some minor actinides associated with the uranium–plutonium cycle, such as Pu-238 (half-life 88 years) and Np-237 (half-life 2.144 million years), but in very small amounts. For example, in a spent fuel Th–U-233 fuel irradiated at 60 GWd/t, there is about 30 times less Np-237 than in the spent uranium–plutonium fuel with the same irradiation. This report value is 60 for Pu-238 ([10], p. 52). In fact, we have seen that there are various ways to deploy the thorium cycle by using different fissile material (MEU, plutonium, U-233) in different types of reactors (light water, heavy water, fast neutron, molten salts, etc.), often in "symbiotic" systems that involve different types of reactors at the same time, with a proportion which varies with time. This makes comparisons difficult in terms of GRI for each scenario. However, all studies on this subject show that cycles based on thorium lead to GRIs well below those cycles based on uranium (see, for example, the study in [15]).

Non-proliferation Issues Regarding the Development of a Thorium Fuel Cycle

The non-proliferation of nuclear weapons from the use of nuclear energy has always been considered as one essential element to take into account in the deployment of any technology or process implemented in this field. This issue was addressed in particular in the framework of an extensive study conducted between 1978 and 1980 on the nuclear fuel cycle at large (International Nuclear Fuel Cycle Evaluation, INFCE, [17]). The general conclusion was that the technical obstacles to military use of thorium cycles with uranium enriched to less than 20 % are similar to those of the uranium–plutonium cycle. However, this was an exercise launched on the initiative of the President of the United States at the time, Jimmy Carter, and involved nearly fifty countries and more than 500 "experts" to give a final document that was 20,000 pages long, and as such the lessons learned from these studies were based on an avalanche of rather scattered technical data mixed with non-free commercial interests and combined with political or diplomatic considerations. Therefore, it is not sufficient to refer only to this general conclusion if one wants to compare more precisely the relative merits (or demerits) of uranium and thorium cycles in terms of non-proliferation.

For this, we must first recall some physical characteristics to take into account in assessing the difficulty to use fissile materials for atomic bombs. There are basically four points of interest:

1. The critical mass of a bare homogeneous sphere of fissile material, that is, without neutron reflector. This determines the amount of material to be manufactured, but also the weight and size of the explosive device. The critical mass values for U-233, U-235 and "weapons-grade" plutonium are, respectively, 16, 48 and 11 kg.
2. Spontaneous neutron emission, which should be as low as possible for the functioning of a "rudimentary" nuclear bomb in order to lower the probability of pre-detonation. Excessive spontaneous emission of neutrons requires designing complex devices to guard against the effects of these neutrons, and it even becomes practically impossible to design an "effective" and reliable bomb by using too highly neutron-emitting fissile materials. The emission values for U-233, U-235, and "weapons-grade" plutonium are, respectively, 1.23, 0.364, and 60,000 neutrons/s kg.
3. The heat generated by the intrinsic radiation emitted by the fissile material itself. Excessive heat can complicate the process, particularly for making a bomb, and even jeopardize its operation (except, again, with implementation

of sophisticated specific provisions to deal with this effect). The heat values for U-233, U-235, and "weapons-grade" plutonium are, respectively, 0.281, 0.00006, and 2 W/kg.

4. The irradiation power of the fissile material for the obvious reasons of radiation protection of the personnel who handle these materials, but also for reasons of potential damage to electronic components. For U-233, the radiation power depends on the U-232 content. For U235, it is very low and for "weapons-grade" plutonium it is low.

We notice that the most important difference between uranium (either U-233 or U-235) and plutonium comes from the neutron emission, which is mainly due to the isotope Pu-240 of the plutonium. Plutonium always contains a more or less significant proportion of Pu-240, depending on the origin of the plutonium and, particularly, depending on the time during which the fuel was irradiated (fuel burnup). Such a phenomenon does not exist for U-233, which is mixed with a substantial or small amount of U-234 (in proportions that vary depending on its origin), which is not a neutron source. Therefore, with U-233, it is possible, owing to this very low emission of neutrons, to produce a much simpler device called a "gun"-type bomb as was the Hiroshima bomb (ultra-rapid merge of two hemispheres). This type of device becomes almost impossible with plutonium, which requires the use of a much more complex implosion device to achieve an explosive chain reaction with a high yield (homogeneous material compression). This advantage can be mitigated, however, by the high α activity of U-232. Indeed, nuclear reactions of the type (α, n) can occur on light elements that may be present in trace amounts in the fissile material, a process that causes unwanted emission of neutrons. However, this process produces far fewer neutrons than Pu-240 and its effects can be minimized by reducing the levels of contamination of light elements.

All this shows that it is possible to make an atomic bomb with U-233, although the uranium contains a certain proportion of U-234 (typically 10–20 % for conventional reactors). Moreover, it is now established that at least one country, the United States, tested atomic bombs made with U-233, specifically during a test called "teapot" in 1957.

However, a difficult obstacle to overcome remains, which is that of radiation emission from the unavoidable presence of U-232 in U-233, as discussed in relation to the manufacture of fuel containing U-233. A detailed discussion of these issues can be found in [18]. That said, there are several ways to overcome these difficulties. One of them is to reduce the proportion of U-232 in U-233 produced in the reactor at the source. In this regard, there are several possibilities, which include the production of U-233 in blankets of fast neutron reactors, as we have already mentioned. It is then possible to produce U-233 with under 10 ppm U-232 contamination, that is, between 10 and 100 times less than that produced in a "classical" thermal reactor, but obviously in lesser amounts. One can find a detailed discussion on this topic cited in [19]. Let us just mention here that the United States was able to produce in the 1950s and 1960s about 130 kg of U-233 with a proportion of 40–50 ppm of U-232 and even 400 kg of U-233 with a proportion only 5–7 ppm of U-232.

In view of the aforementioned considerations, we find that the overall assessment of the degree of proliferation resistance of the thorium cycle over a uranium cycle is a multifaceted issue that requires careful analysis to avoid the hasty and sometimes simplistic conclusions that one can find in the literature. This type of analysis has been attempted by a specialist in these matters, Bruno Pellaud, former Deputy Director General of the IAEA and Head of Department of Safeguards, whose results were published in a non-public document [20]. In this study, a method of analysis called SAPRA [21] was used, which allows a comprehensive evaluation of various reactor systems and cycles vis-a-vis the risks of nuclear proliferation. This analysis focused on three types of reactors (light water, heavy water, high temperature) and six fuel cycles (uranium with or without recycling, thorium in association with uranium, plutonium, and U-233 with or without recycling). Suffice to say here, in conclusion on this issue, that this study showed that overall, the thorium cycle appears to be at least as resistant to proliferation as the uranium cycle, with a higher degree of resistance in some cases.

Conclusion

This overview shows that thorium offers some interesting prospects, especially in terms of natural uranium savings (if U-233 is recycled), but also with regard to the reduction of the total final waste radiotoxic inventory. Thorium fuels also present attractive features in terms of behavior under irradiation and neutronic behavior in the reactor. However, despite the existence of examples of use of thorium reactors in the past, industrial experience with this cycle is still very limited and almost non-existent on the back end of the cycle (reprocessing and recycling).

Most of the basic knowledge on thorium cycle is available, but it is clear that the deployment of this cycle on a large scale would require much R&D, especially in the field of reprocessing and fabrication of U-233-based fuels, as well as heavy industrial investments. It is unlikely that in the near

future, the conditions are met to justify the initiation of such efforts for a majority of countries. However, in few decades, the emergence of new constraints could change the current situation and lead to industrial deployment of fuel cycles based on thorium. To this end, one of the drivers could be the opportunity offered by these fuel cycles to approach or even to reach the iso-generation of fissile material in some types of thermal reactors. Another incentive would be the recycling of plutonium from LWR MOX fuels in a symbiotic system of nuclear reactors.

In that perspective, the thorium cycle certainly deserves further research and consideration. This report thus provides an update and a comprehensive overview of worldwide works on this important topic.

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Thorium Molten Salts: Theory and Practice

Paul A. Madden and Mathieu Salanne

Abstract

Molten salt reactors will involve multicomponent molten salt mixtures operating at high temperatures and the in-line chemical processing of the salt will be a crucial part of the operating conditions (Delpech et al., J Fluorine Chem 130:11–17, 2009, [1]). Furthermore, the reactors will operate under conditions of very fast pumping and extreme heat transfer. Because the rates of corrosion increase rapidly with increasing temperature, an important element in a successful design will be to work at as low a temperature as possible so as to allow a reasonable lifetime for the containment materials, pumps, and chemical processing elements. Finding the ideal range of salt compositions to minimise the corrosion problem, whilst maintaining a critical concentration of the fissile material in the core and avoiding precipitation of reaction products in the heat exchangers etc. is a very steep requirement.

Introduction

Molten salt reactors will involve multicomponent molten salt mixtures operating at high temperatures and the in-line chemical processing of the salt will be a crucial part of the operating conditions [1]. Furthermore, the reactors will operate under conditions of very fast pumping and extreme heat transfer. Because the rates of corrosion increase rapidly with increasing temperature, an important element in a successful design will be to work at as low a temperature as possible so as to allow a reasonable lifetime for the containment materials, pumps, and chemical processing elements. Finding the ideal range of salt compositions to minimise the corrosion problem, whilst maintaining a critical concentration of the fissile material in the core and avoiding precipitation of reaction products in the heat exchangers etc. is a very steep requirement.

To design a suitable salt, information is required on the phase diagrams, thermodynamic properties (heat capacity, activity coefficients, etc.), and the transport properties (viscosity, thermal conductivity, etc.) of these multicomponent mixtures. In general, progress on the phase behaviour can be made with thermodynamic modelling methods [2–5], where data on the pure materials or simple mixtures are combined to predict freezing points, vapour pressures, etc., of more complex mixtures. However, the fundamental databases on the elements of interest are very sparsely populated. Performing the experiments to determine properties such as the thermal conductivity with hot materials is virtually impossible. In these circumstances, first-principles-based atomistic computer simulation methods can play a vital role in providing values for thermodynamic and transport properties of interest and in supplying the insight into the relationship between these quantities and the underlying atomic structure of the fluids [6, 7].

In this article, the potential of the computer-simulation methodology for making a practical contribution (perhaps in concert with thermodynamic models) to the design process with salt mixtures of realistic complexity will be assessed and the underlying reasons for any limitations in applicability explained. The particular focus will be on mixtures of lanthanide and actinide fluorides and chlorides with fluorides

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and chlorides of alkali and alkaline-earth metals. Technical aspects of suitable simulation methods for such materials have been described in numerous publications and will not be repeated here [8, 9]. In brief, these involve atomistic molecular dynamics simulations where, for example, an LiF:ThF_4 system is treated as mixture of Li^+, Th^{4+}, and F^- ions [6]. The ionic interaction potentials are derived from condensed-phase electronic structure calculations [10, 11]. Applications of such simulations to calculate all the properties mentioned above for representative melts of the type of interest have also been published, and the details of the calculations will not be repeated here. Rather, we will focus on the extent to which such methods could be applied in general and highlight potential limitations.

The Materials of Interest

The chemistry of the fluorides and chlorides of the elements of interest is relatively simple, and this certainly facilitates the construction of realistic simulation models. The different elements in the actinide and lanthanide series are distinguished by different numbers of electrons in the f-orbitals. However, in the most common oxidation states of these elements, these f-electrons do not play a significant role in interactions with halide ions—they behave as if part of the electronic "core". Consequently representing them as simple, spherical "closed-shell" ions, and neglecting any covalent interactions with the halides, works well in practice and is supported by electronic structure calculations. The same simplicity is evident in the systematic trends observed in the chemical and physical properties of the melts [12, 13]. The cations of different elements differ mostly through their different sizes.

This ionic picture has several important consequences for the overall endeavour of dealing with complex mixtures. Firstly, the interaction potential for any given pair of ions is, to a good approximation, transferable—that is the potential derived for (say) a pure LaF_3 melt will describe the interactions between La^{3+} and F^- ions in a LiF:LaF_3 mixture very well—and the same will apply to more complex multicomponent mixtures. Furthermore, it will transfer between the liquid and solid phases, and may therefore be used to study melting. Indeed, the transferability is the principle that underpins the success of the thermodynamic modelling methods, as in [2–5]. Secondly, the interaction potentials vary systematically with the elements involved, with the size of the cation being its key distinguishing feature, as indicated above [12–14]. This enables any differences in the physical behaviour exhibited by different elements to be rationalised and also allows the identification of elements that can be expected to have very similar physical properties: La^{3+} and U^{3+} have very similar ionic sizes and similar physical properties in $LaCl_3$ and UCl_3, for example. Recognition of such similarities can greatly reduce the effort required to set up a database of properties for the whole lanthanide and lighter actinide series as suitable proxies for particularly difficult-to-study elements can be identified. In [2], for example, Ce^{3+} is used as a proxy for Pu^{3+}.

The simplicity of these underlying interactions does not imply that the melts themselves are simple. Although they are modelled as collections of individual spherical ions, the key to understanding their properties is to recognise that they behave as complex mixtures of quasi-molecular coordination complexes, in which the highly charged cations bind a characteristic number of anions to form potentially long-lived complexes. For example, Fig. 1 shows a snapshot of the ionic positions in a simulation of a LiF:ThF_4 mixture [6], with the Th^{4+} ions coloured blue, the F^- ions green, and the Li^+ ions pink. Lines ("bonds") have been drawn between ions, which are separated by less than some characteristic distance, which help to illustrate that each Th^{4+} ion coordinates (on average) eight F^- neighbours. Such coordination complexes persist for quite long times compared, for example, with the structural relaxation time on which the viscosity of the melt is established [15]. On such timescales, then, regarding the melt as being comprised of ThF_8^{4-} ions in a sea of Li^+ and excess F^-

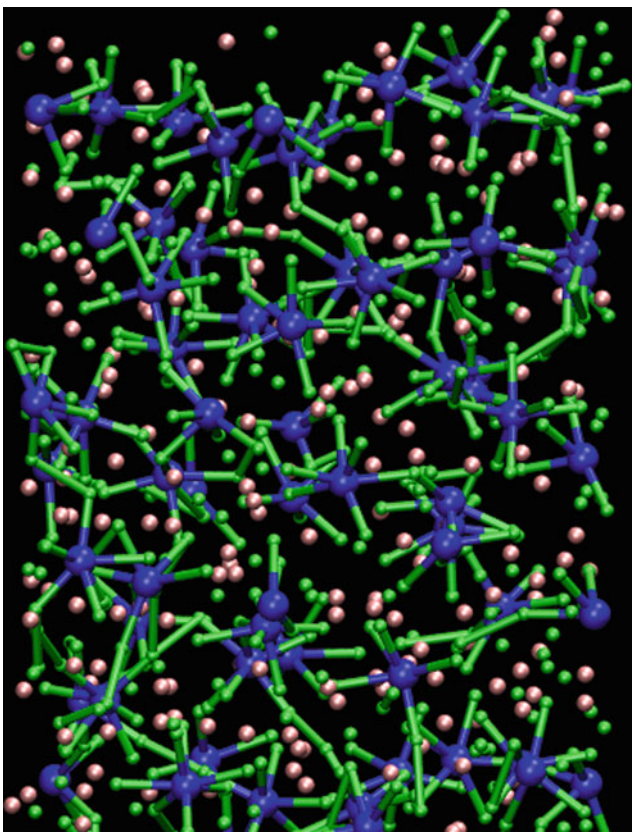

Fig. 1 Simulation of a LiF:ThF_4 mixture at the liquid–vapour boundary

may be the most appropriate description of the fluid. Furthermore, on closer inspection, cases where two Th^{4+} ions are both bonded to a single F^- are seen, corresponding to cross-linking of the coordination complexes to form an oligomer containing two or more Th ions.

The simulation is, in fact, of the liquid vapour interface of the melt [6], with the vapour phase on top (at this temperature, the vapour pressure would be extremely low, so no ions are seen in the vapour region in this snapshot). It can be seen that the liquid is stratified in the vicinity of the interface, with a high concentration of coordination complexes in a layer at the interface. This structural complexity would influence the surface tension.

The assertions made in this section—about the accuracy of the ionic interaction potentials and the atomic-scale structure of the halide melts—have been examined in a considerable body of work, where the structures calculated for the simulated fluids have been compared with structural information obtained by neutron and X-ray diffraction [13, 16], by extended X-ray absorption fine structure (EXAFS) [17], by Raman scattering [18], etc. This fundamental work underpins the validity of the claim that these fluids can be simulated sufficiently accurately for material properties to be predicted. A particularly important realisation has been the role of the polarisation of the anion in constructing accurate interaction models—anion polarisation plays a crucial role in controlling the differences in energy between coordination structures and plays a role in the dynamics of the melts as well as in obtaining good agreement between simulated and experimental structures [12, 19]. The average coordination number of a given cation is not a fixed number [17, 18, 20], as had often been supposed; in a particular melt a range of instantaneous coordination numbers is found and the average coordination number varies with the composition of the melt.

Although the number of cases where careful comparisons of experimental and simulated structures have been made is considerably smaller than all possible lanthanide and actinide melts (and mixtures), the principles of transferability and systematic variation with element have been strongly supported in these studies and lead to the claim that the whole class of materials may be successfully simulated.

Before leaving this point, it is important to realise where limitations to the approach will occur. When several oxidation states of the same element occur in the same material, it has proven possible in some cases to extend the model by treating each ionic oxidation state as a separate species, for example, with a system containing Ce^{4+} and Ce^{3+} as a mixture (this, of course, will not work if electron-transfer events are important). Confining attention to pure halide melts has led to the important simplification that covalent interactions may be neglected. In the presence of other electronegative elements in the melt, in particular, oxygen, this simplification will not hold and genuinely long-lived covalent species may form. Well-known examples are the uranyl ion, UO_2^{2+}, and the oxyfluorides. The simulations could be extended by including such species as molecular ions, alongside the simple atomic Li^+, F^-, etc., ions but, of course, any phenomena that depend on chemical equilibria between such species would not be described. To handle such complexities on a purely objective basis would require a fully ab initio simulation approach—and that would take us into a whole new and computationally prohibitive ball game.

Thermodynamic Properties

A number of significant thermodynamic properties can be calculated straightforwardly in a molecular dynamics simulation.

The density, for example, is easily computed from the average volume of a simulation cell containing a given number of ions in a "constant-pressure" simulation [6, 7]. By performing a series of such calculations at the same pressure but different temperatures, the thermal expansion is obtained. Although it is easy to calculate, predicting the density of a material to (say) 1 % from first principles is quite difficult as it depends quite sensitively on an aspect of the interionic interactions that is difficult to calculate accurately in electronic structure calculations, namely the dispersion interaction [8, 21]. This is however an arena of much current activity and progress [11]. Until recently, many of the comparisons between simulation and experimental values to which we refer had been made by empirically adjusting the strength of the dispersion term in the interaction model to reproduce the experimentally measured density.

The heat capacity is a very important parameter for modelling the characteristics of a reactor. It is easily calculated from the average potential energy of a simulation run at constant pressure [6, 7]. Taking the difference between such enthalpy values obtained from simulations run at neighbouring temperatures gives a finite difference approximation to the heat capacity:

$$C_{\mathrm{p}}^{\mathrm{m}} = \left(\frac{\partial H^{\mathrm{m}}}{\partial T}\right)_{\mathrm{P}}$$

Alternatively, it may be obtained from the fluctuations in the internal energy of the simulated sample. In a simulation of 78 mol% LiF–22 mol% ThF_4, for example, a value of 1.049 J g^{-1} K^{-1} was obtained, compared with an experimental value [3] of 1.0 J g^{-1} K^{-1}.

It is much more difficult to obtain the crucial thermodynamic quantities concerning phase or chemical equilibrium, however. The basic reason for this is that the position of equilibrium is determined by the equality of the chemical

potential of some species on each side of the equilibrium. The chemical potential is the molar Gibbs free energy, which involves the entropy of the material. Entropy cannot be determined by studying any property of the system in a single thermodynamic state, which is what a single simulation run represents. The general problem of characterising equilibria in simulations is discussed comprehensively in [22].

In some limited circumstances, it may be possible to simulate a system at phase equilibrium directly. The liquid–vapour system illustrated in Fig. 1, might be an example. However, the limitation here is that because of the very low density of the vapour phase, the fluctuations in vapour pressure are enormous and make reliably establishing a position of equilibrium very difficult. More reliable ways of dealing with liquid–vapour equilibria (Gibbs Ensemble) are discussed in [22]. In the case of freezing, it might be possible by a process of trial and error, to set up a simulation cell in which the solid and liquid phase of a material coexist at a certain average temperature and pressure, which then determine the coexistence point. This has been used to study the freezing of LiF, for example [21]. It becomes far more complicated to do the same thing for mixtures, however, as, amongst other things, the composition and structure of the crystalline phase that coexists with the liquid will not be known. In the very practical example discussed in [2], for example, one is interested in the limit to the solubility of Pu in the melt due to the crystallisation of Pu-containing solid phases. A direct, predictive approach to such a complex problem by simulation seems impossible.

Although the chemical potential cannot be determined from a single simulation, free energy (chemical potential) differences can be calculated from a series of simulations by using the method of thermodynamic integration [22]. A particularly valuable approach is to calculate the difference in free energy between two related materials by "transmuting" one into the other. The two materials differ only in their interaction potentials, which we could call $U(0)$ and $U(1)$ for reasons which will become clear shortly. We can consider a set of intermediate, virtual materials with an interaction potential $U(\varphi)$, where $U(\varphi) = \varphi U(1) + (1 - \varphi)U(0)$. As φ varies from 0 to 1, the interaction potential changes from that appropriate to material 0 to material 1. It can be shown that:

$$\Delta F = \int_0^1 d\varphi \langle U(1) - U(0) \rangle$$

where ΔF is the difference in free energy between the two materials at pressure P and temperature T. The angle brackets in the equation indicate that the difference in interaction potentials of the two materials is to be calculated in a simulation carried out with the potential $U(\varphi)$ running at pressure P and temperature T. In practice, a discrete series of simulations with φ between 0 and 1 are carried out and the integral is completed using (say) the trapezium rule.

An illustrative application [13] of this method was to calculate the chemical potential differences between a series of lanthanide and actinide elements dissolved in an alkali halide by changing the interaction potentials of, for example, an La^{3+} ion with the other components of the solution into those appropriate to a U^{3+} ion. The chemical potential difference is determined by the difference in activity coefficients of the two ions in the mixture simulated. The actual application was to a chemical equilibrium to examine how the presence of the solvent affects the electrodeposition potential of the different ions, with a view to seeing how tuning the solvent could affect the possibility of selectively electrodepositing the two metals in a pyroprocessing application [23]. However, the applicability of the method is much wider as the activity coefficient is the fundamental unknown in thermodynamic modelling methods of the type described in [2–5]. Combining the output from the simulations with empirical information via a quasichemical model of this type could be a useful way of estimating differences in the solubilities of different elements and other aspects of phase equilibria. A systematic effort to combine the thermodynamic modelling based upon the quasichemical approximation with the information obtainable from simulation would be very profitable.

Transport Properties

The calculation of transport properties is comparatively straightforward. The viscosity, η, for example, is given by:

$$\eta = \frac{1}{k_B T V} \int_0^\infty \langle \sigma_{\alpha\beta}(0) \sigma_{\alpha\beta}(\tau) \rangle d\tau$$

where $\sigma_{\alpha\beta}$ is a component of the stress tensor, itself expressible in terms of the instantaneous positions and velocities of all the ions in the simulation, and the term in brackets is its time autocorrelation function. In practice, the correlation function decays to zero on some timescale (the structural relaxation time of the fluid) so that the running integral reaches a plateau value, which determines the viscosity. k_B is Boltzmann's constant and V is the simulation cell volume. If the relaxation time is long, long simulations may be necessary to achieve a well-converged result for the viscosity.

For a number of materials, comparisons of simulation-predicted viscosities with experimental values have been made and good agreement found. Depending on their structure, molten salts can have markedly different

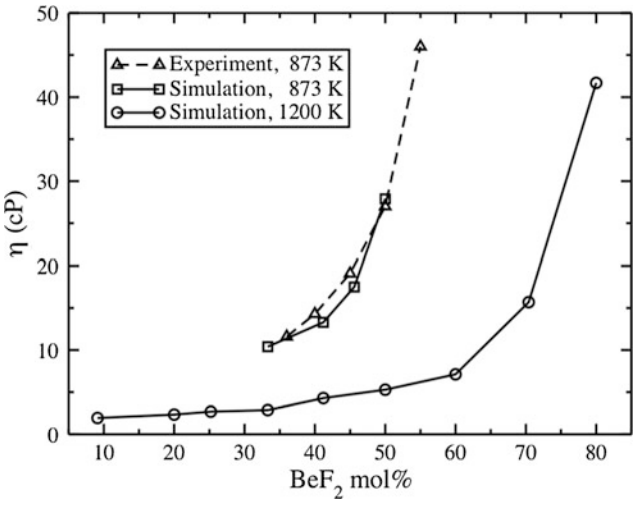

Fig. 2 Viscosity increases sharply in a LiF:BeF_2 mixture as the BeF_2 component increases

fluidities. In the case of mixtures where either no particular structural pattern is observed (molten alkali halides) or in which the coordination complexes are only weakly cross-linked (such as the ZrFx species in molten fluorozirconates [20]), low viscosities are obtained (typically 1 mPa s^{-1}, i.e., of the same order of magnitude as liquid water at room temperature). As soon as a strong network starts to form, the viscosity can become much higher. In the case of LiF:BeF_2 mixtures shown in Fig. 2 [24], this increase is very sharp as the composition of the mixture becomes rich in BeF_2. By extracting the characteristic time from the stress tensor autocorrelation function and comparing it to the Be–F–Be bond forming/breaking relaxation time, we could show that the two quantities were strongly correlated, indicating that this structural event was very likely to be at the origin of the viscosity increase [15]. The LiF:BeF_2 system at the eutectic composition was used as the solvent for thorium fluoride in the Oak Ridge Molten Salt Reactor.

The thermal conductivity may be calculated in a similar way, that is, from the time integral of the correlation function of the energy current [25, 26]. The thermal conductivity is a key quantity for calculating the heat transfer within a reactor or in the fluids used to transfer that heat to a generator. However, the experimental measurement of thermal conductivity is difficult [27] in molten salts, because of the high temperatures involved so that being able to predict it from computer simulations is of paramount importance. In those cases where comparison with reliable information is available (from forced Rayleigh scattering), the agreement with experiment has been good [25] (Table 1).

Unlike the viscosity, it appears that this property is not closely related to any structural feature of the melts.

Other transport coefficients that can be calculated in similar ways include the electrical conductivity [28] and diffusion coefficients [29]. Although these are not directly related to reactor performance, they have played a key role in understanding the dynamical properties of the melts, enabling reliably measureable quantities to be compared with simulation values and then the simulations to be used to examine the atomistic mechanisms that lead to the observed behaviour.

Table 1 Comparison of calculated thermal conductivity with experimental values, for various salts at high temperature [25, 26]

System	LiCl (1200 K)	NaCl (1300 K)	KCl (1200 K)
$l_{calculated}$ (W m^{-1} K^{-1})	0.643	0.509	0.343
l_{exp} (W m^{-1} K^{-1})	0.534	0.478	0.345

Conclusion

The purpose of this article is to call attention to the role that atomistic computer simulation can play in providing values for key quantities that are required to predict the behaviour of molten salts of the type involved in proposed molten salt reactors and heat transfer systems and to provide some links to the primary literature on these methods. The ability to predict transport and some thermodynamic quantities has been demonstrated, and their currently limited role in predicting phase behaviour highlighted as an area for concerted effort.

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Liquid-Fluoride Thorium Reactor Development Strategy

Kirk Sorensen

Abstract

Flibe Energy has worked to advance the technology for liquid-fluoride thorium reactors (LFTRs) since its incorporation in April 2011. Its objectives for modular reactor design and its plans for manufacturing and deployment of these reactors will be described. Flibe Energy has also undertaken a feasibility study of LFTR technology along with its partner Teledyne Brown Engineering. This study will be conducted under the auspices of the Electric Power Research Institute (EPRI) and Southern Company Services (SCS). The outcome of the study will be an estimate of the levelized cost-of-electricity (LCOE) from a 250-MWe modular LFTR built in the 2030 timeframe.

One of the great challenges for electrical utilities in the United States is the accelerated retirement of coal-fired powerplants. These retirements are taking place predominantly in the eastern United States, and particularly along the Ohio River Valley, Virginia, and the Carolinas. Coal plant retirement is being driven in large part by ever-tougher emissions requirements from the US Environmental Protection Agency. Coal and nuclear are two baseload electrical energy sources in the United States. "Renewable" energy options are not going to be able to meet the electrical demand in the regions where coal and nuclear plant retirements are going to be most significant. An intermittent and unreliable energy source such as wind or solar cannot replace a stable, reliable baseload energy source like coal and nuclear.

Several new light water reactors (LWR) are under construction in the United States, most notably the Vogtle 3 and 4 reactors being built in Georgia by Georgia Power, a division of Southern Company. They are AP1000 reactors and many of their components are built in a factory and shipped to the site, but they are still very large reactors and extensive site work is necessary.

The great challenge that looms for the US nuclear industry is that, even with license extensions, the time is coming when large numbers of nuclear reactors will be retired. This is sometimes called "the retirement cliff". The Fukushima incident and the onset of inexpensive shale gas, as well as market distortions surrounding solar/wind incentives, have accelerated the retirement of existing nuclear plants and exacerbated this challenge.

In order to preserve nuclear power's role in US electrical power generation it will be necessary to produce large numbers of new nuclear power plants starting in the late 2020s and continuing for several decades, and the rate at which they must be deployed means that there will need to be a very different approach to siting and construction.

The emphasis on private industrial leadership in new nuclear developments is something that is different in the United States compared with many other countries. For decades after World War 2, development of nuclear technology was the exclusive responsibility of the US Atomic Energy Commission (AEC). Private companies had to be invited to work with the AEC or be excluded entirely from any sort of nuclear enterprise. This approach has been slowly changing over the intervening decades, but it has only been in the last 10 or 20 years that the notion of nuclear "entrepreneurialism" could even be considered. Now, according to the Department of Energy's own outlook for the future of nuclear power, they expect that private industry will lead the way into new technologies. They see their own role as a supporter of research and development, and this is a significant change. It means that private businesses will have to develop business plans that can attract industrial investment

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and show the potential for profit. The DOE will help as they see fit but will not drive any particular technology direction.

Recognizing that it must be private industry that drives new nuclear technology forward in the United States, Flibe Energy was founded in 2011 with the ambition to provide the world with affordable and sustainable energy, water, and fuel. We believe that the way to achieve this goal is to use liquid-fluoride thorium reactor (LFTR) technology, which is an evolution of work done at the Oak Ridge National Laboratory (ORNL) on the Molten Salt Reactor Program (MSRP), which ran from 1957 to 1976.

Flibe Energy is located in Huntsville, Alabama, sometimes called the "Rocket City". Huntsville is a high-tech community and its metro area has the highest per capita concentration of engineers in the US. It's a place that has literally shot for the Moon and achieved it, as this is where Wernher von Braun and his rocket team developed the Saturn V moon rocket. It's also the home of a large military facility, Redstone Arsenal, and NASA's Marshall Space Flight Center facility, where I spent the first 10 years of my career. We consider it a good place to undertake the ambition of solving the world's energy needs.

Huntsville is also geographically blessed for manufacturing and shipping activities. From its position on the Tennessee River, one can conveniently access the Gulf of Mexico, but it is far enough inland that the danger of severe storms is mitigated. An extensive rail network connects the country, and Huntsville International Airport is small in terms of passenger traffic but large in terms of air freight. Heavy manufactured components can be moved anywhere in the world from this location.

Huntsville is also not far from Oak Ridge National Laboratory in Tennessee, which is where some of the earliest discoveries about thorium were made. Thorium carbonate was first irradiated on a large scale to produce uranium-233 in ORNL's Graphite Reactor in 1943, and from these samples it was discovered that uranium-233 had superlative nuclear properties, which permitted the development of a thermal breeder reactor. Later, in the 1950s, Oak Ridge also built the first molten salt reactor, the Aircraft Reactor Experiment, and showed it was stable and self-controlling. In the 1960s, the thorium fuel cycle and molten salt reactor technology was united when the Molten Salt Reactor Experiment was built and operated successfully on uranium-233 as a fuel, which comes only from thorium. Although its molten salt reactor research days ended long ago, proximity to ORNL is still an advantage. Many of the retirees still live in the local community and are willing to share their knowledge of molten salt reactors. There are also a great number of design and operations documents that reside in the Oak Ridge archives. So the proximity between Huntsville and Oak Ridge is advantageous.

To understand how we arrived at our decision to pursue a liquid-fluoride reactor based on the thorium fuel cycle, let us simplify a nuclear power system to its essential components. There is the reactor itself, where thermal energy is generated from over a million trillion fission reactions each second. This thermal energy is carried away by a coolant fluid to a heat exchanger, where that thermal energy is transferred to another fluid that will pass through the power conversion system. The power conversion system has a turbine, which turns a shaft connected to a generator, making electricity. About two thirds of the thermal energy produced is rejected as a waste product to the environment. In a pressurized water reactor (PWR), the primary coolant is pressurized water and the working fluid is also pressurized water raised into steam, with a steam turbine as the power conversion system. But there are other possible ways to build a reactor if different coolants are considered.

The temperature at which thermal energy is delivered from a reactor to its power conversion system is important because it determines how efficiently that thermal energy can be converted into useful work, most often electricity. The higher the temperature at which thermal energy can be transferred, the more of that thermal energy can be converted to work and, thus, less must be rejected as waste heat. So achieving higher and higher operating temperatures have always been a goal of nuclear operations. Achieving higher temperatures has depended greatly on the choice of coolant used in the reactor.

Nuclear reactors have generally been divided into four different families of coolants, and one could consider them in a two-by-two matrix, with operating pressure across the top axis and operating temperature along the side axis. Gas coolants operate at high temperature and high pressure. Fluoride salt coolants operate at high temperature and low pressure. Liquid metal coolants operate at medium temperatures and low pressures, and water operates at relatively low temperatures and high pressures. Ideally, the most desirable kind of coolant would be one that reaches the highest temperatures at the lowest pressures, and this can be achieved by considering the use of liquid fluoride salts as a coolant. Pressurized water, by contrast, must operate at very high pressures to achieve modest temperatures, which limits the efficiency at which it can be used to produce electricity.

There is another important attribute that should be considered very early in reactor design, and that is the volumetric heat capacity of the coolant, in other words, how much thermal energy a unit volume of the coolant can hold. Volumetric heat capacity is the product of the specific heat of the material and its density. In this category water is an exemplary choice, but its performance is exceeded by a fluoride salt composed of lithium and beryllium fluorides. The volumetric heat capacity of liquid sodium is marginal

and gases have very low performance on this metric. Volumetric heat capacity is very important because it is the basic yardstick that sizes the reactor vessel, the piping, and the primary heat exchanger. The higher the volumetric heat capacity, the more compact the reactor can be. Gas-cooled reactors are particularly disadvantaged in terms of the volumetric heat capacity of their coolant because of its very low density.

As a class of materials, fluoride salts embody many advantages. They are the most chemically stable of all materials, which gives them a tremendous liquid temperature range of roughly a thousand degrees Celsius. This is far in excess of the few hundred degrees Celsius of liquid range that can be achieved with water under great pressure. Their ionic bonding structure also makes them entirely impervious to radiation damage from neutrons or gamma rays. This is again contrasted with water, which is continuously broken apart by radiation into hydrogen and oxygen, and must be recombined.

With the goal of reducing construction costs, it is highly desirable to use materials in the reactor to perform multiple functions. The pressurized water reactor is a good example of this, in that it uses water to both cool the fuel rods and to slow down, or moderate, the high-energy neutrons from fission. Liquid fluoride salts can also be more than just a coolant. The mixture of lithium fluoride and beryllium fluoride, sometimes called "flibe", can be used as a solvent to carry nuclear fuels such as uranium, plutonium, and thorium. When fluoride salts carry salts of nuclear fuels in them, they can fulfill the goal of having a material that serves multiple purposes in the reactor, and this has the potential to simplify the reactor and reduce costs.

In a liquid-fluoride reactor, fuel salt containing fissile material passes through metallic piping into the reactor vessel, where internal graphite structures slow down (moderate) neutrons, increasing the probability that these neutrons will cause fission reactions in the fissile material. Fission reactions deposit thermal energy in the fuel salt, which increases its temperature. As the heated fuel salt passes out of the reactor vessel into the piping system, fission reactions are no longer possible because the fuel salt has been separated from the graphite. In an external heat exchanger, the fuel salt heats a coolant salt and returns to the reactor vessel. The coolant salt passes out of the reactor containment region and heats the gaseous working fluid of a gas-turbine power conversion system, analogous to the gas turbines used in today's jet engines. The hot, high-pressure gas expands in a turbine, generating shaft work that turns a generator and produces electricity while also turning a compressor. The turbine gas exhaust, now at low pressure, is cooled either by external air or water. This cooling process can serve as the thermal input for a seawater desalination process if the reactor is located near salt water. The cold, low-pressure gas is then compressed in the compressor using shaft work from the turbine and is ready to be heated again to generate work and power.

A liquid fuel enables a remarkable passive safety feature to be implemented, which can solve perhaps the most vexing problem in reactor safety. For many years, there has been great concern about how to cool the solid fuel of a reactor in the event of a loss-of-coolant accident (LOCA) in order to prevent a meltdown of the fuel and the release of radionuclides. Various approaches have been proposed, including some newer and more innovative ideas involving large thermal sinks. But the use of liquid fluoride fuel enables a simple and remarkable solution, based on the melting temperature of the fuel, which is about 400 °C. The reactor is fitted with a drain line that is kept plugged by a frozen slug of salt. This plug is kept frozen by an active flow of coolant over the outside of the drain line. In the event of a complete loss of power, the salt plug melts, and the fuel salt in the reactor drains through the line into a dedicated tank called a drain tank. This tank is configured to maximize the passive rejection of decay heat to the environment. This enables the reactor to dispense with a multitude of emergency core-cooling systems that are required in solid-fueled reactors, particularly those that operate at high pressure. Because the frozen salt plug requires active cooling, in the event of complete power loss, that cooling will be interrupted and the plug will melt, causing the fuel to drain and the reactor to completely shut down. This remarkable safety attribute of the reactor is a compelling argument for consideration of the liquid fuel approach.

These numerous advantages of liquid-fluoride reactors would only be theoretical if it were not for the successful demonstration of this technology at Oak Ridge National Lab in the late 1960s. There, they designed and operated a molten salt reactor experiment, which used graphite to slow down or moderate the neutrons. Liquid fluoride salt, loaded with dissolved enriched uranium tetrafluoride fuel, flowed through channels milled in the faces of the graphite moderator elements. This structure was loaded in a reactor vessel made from a nickel superalloy called Hastelloy-N, which had been specially developed to be compatible with fluoride salts. As the salt flowed through the graphite lattice in the reactor vessel, it was heated by fission reactions.

The salt then passed into the primary heat exchanger where it was cooled by another salt, called the coolant salt, which carried the thermal energy outside of the reactor cell. The coolant salt was itself directly cooled by air as it flowed through a radiator structure. The reactor vessel, its pump, and primary heat exchanger were quite compact owing to the high volumetric heat capacity of the fluoride salts used, and the operation of the reactor was shown to be highly stable and self-controlling. Those who operated the reactor took the greatest pride in the fact that they had demonstrated that they

could maintain the reactor over 20,000 h of successful operation from 1965 to 1969. The reactor ran on all three different kinds of fissile fuels during its operation, becoming the first and only reactor to achieve this feat.

The engineers, scientists, and technicians were very proud of their accomplishments and wanted to take the next step—a larger reactor that would demonstrate the complete thorium fuel cycle. However, the new Nixon Administration was cutting budgets across the board and insisted that the US Atomic Energy Commission consolidate all of their breeder reactor efforts into a single line of research. The liquid-fluoride reactor, demonstrating the thorium fuel cycle, was cancelled in 1972.

Breeder reactors were the goal of the entire US advanced nuclear enterprise at the time, and they could efficiently use either abundant thorium or uranium-238 as their basic fuels. However, breeder reactors were considerably more ambitious than reactors that just burned the very small fraction of natural uranium that is already fissile. They had to carefully manage their neutron supply and had to incorporate features to allow fuel that had been generated in the periphery of the reactor to be transferred to the central areas of the reactor.

For solid-fueled reactors this was quite a challenge, as it meant that periodically all of the fuel assemblies would have be removed and chemically dissolved. Then, various elements would be separated one from another and new fuel elements would have to be fabricated, which would be a substantial challenge. Molten salt reactors had a tremendous advantage in this respect as their fuel could be processed and refined in the same chemical state in which it was used in the reactor. Complicated steps of fuel removal, disassembly, decladding, dissolution, and later fuel refabrication were all eliminated in this concept.

As Flibe Energy has considered this technology, we plan to take advantage of several technological improvements since the 1970s. The first is that the technology for compact, reliable gas-turbine engines has also advanced considerably since the 1960s. Today's gas turbines, which use air as the working fluid and burn hydrocarbons to generate thermal energy, have excellent power density and are very responsive. However, other types of gas-turbine engines are possible, which would be heated by nuclear energy rather than combustion. They are based on a closed-cycle and have the potential to use fluids other than air, and which have superior properties.

One of the important advantages of a liquid-fluoride reactor is that it can supply thermal energy at temperatures suitable for a gas turbine power conversion system. This can lead not only to improvements in electrical generation efficiency, but also reductions in size and capital costs. The waste heat from a gas turbine is still of sufficiently high temperature to potentially drive a desalination system, which would be of tremendous value and importance in regions where water is just as important as electricity.

There has also been a far greater interest in small modular reactors, with the goal of building reactor components in a factory environment and reducing site preparation time. The researchers at Oak Ridge were proposing small modular reactors based on liquid-fluoride technology as far back as 1968. Creating modular reactors with liquid-fluoride technology is much easier because the reactors do not operate at high pressure and can be shipped and returned unfueled.

Today's approach to generating nuclear energy begins with mining uranium oxide ore out of the ground and chemically converting it to a fluoride salt—first uranium tetrafluoride and then uranium hexafluoride. This chemical conversion, from oxide to fluoride, is undertaken so that the uranium can be enriched. But at the end of the enrichment process, the uranium fluoride salt must be converted back to an oxide form and this is chemically unfavorable. Then, the oxide powder is sintered into pellets, which are loaded into rods that are formed into assemblies. It is these assemblies that are loaded into the reactor, irradiated to produce electricity, and then removed and disposed.

A liquid-fluoride reactor has the potential to dramatically simplify several of the steps in the nuclear fuel chain. Because the reactor uses fuel in the fluoride form, there is no need to convert uranium hexafluoride back to oxide and to form it into pellets, rods, and assemblies. The uranium hexafluoride can be reduced to uranium tetrafluoride as it is loaded into the reactor and used in that form to produce electrical energy. By using the nuclear fuel in fluoride form, the fuel cycle is not only simplified but fuel recycling is far more straightforward.

The long-term viability of nuclear energy will come about when nuclear fuels can be used with far greater efficiency than is done today. Only a small fraction of natural uranium is fissile; most uranium and all thorium is fertile, meaning that it can be converted into fissile fuel inside a nuclear reactor. Both thorium and uranium-238 require two neutrons to release their energies. One neutron converts them into a fissile form and the other neutron actually causes the fission. Thorium absorbs a neutron and becomes uranium-233, which will fission when struck by another neutron. Uranium-238 absorbs a neutron and becomes plutonium-239, which will also fission. However, there is an important difference between these two options. The fission of uranium-233, when one accounts for non-fission absorptions, will produce 2.3 neutrons. This is enough to continue the conversion of more thorium to uranium-233 fuel, even when accounting for various losses. But the fission of plutonium-239 will produce less than two neutrons. It is not sustainable in today's thermal-spectrum reactors.

The only way to sustainably use uranium-238 and plutonium is to design fast-spectrum reactors, which intentionally attempt to keep neutrons at as high a velocity as possible. In these reactors, non-productive neutron

absorption in plutonium is suppressed and plutonium fission will produce more than two neutrons, enabling sustained consumption of uranium. The fundamental disadvantage of fast reactors is that the probability of neutron reactions, typically represented by a cross-sectional area, is much lower when the reactions are caused by fast neutrons than by slowed-down, thermal neutrons. Plutonium-239 will absorb thermal neutrons roughly one out of three times. The cross sections of plutonium-239 for fast neutrons are much, much smaller than for thermal neutrons; so much so that it requires hundreds of plutonium atoms to achieve the same probability of fission as a single plutonium atom to a thermal neutron. The implication of this difference is that fast reactors require much larger inventories of fissile fuel for a given power rating.

When the performance of uranium-233, which comes from thorium, is compared and contrasted with plutonium-239, which comes from uranium, it can be seen that uranium-233 has a much greater probability of fission and less probability of non-productive absorption. This is the reason why only the thorium fuel cycle is sustainable in thermal-spectrum reactors. Indeed, the central advantage of thorium as a nuclear fuel is its unique ability to be sustainably consumed in a thermal-spectrum reactor.

Thus, nature presents three nuclear options for nuclear energy. The first and most obvious is to use the tiny sliver of fissile uranium that occurs naturally, or to use uranium (and plutonium) in fast spectrum reactors with large fissile inventories, or to use thorium (and uranium-233) in thermal-spectrum reactors that have low fissile inventories.

In the thorium fuel cycle, thorium-232 is struck by a neutron, forming thorium-233. Thorium-233 has a short half-life of about 20 min and decays to protactinium-233, which is chemically distinct from thorium. Protactinium-233 decays with a half-life of about a month into uranium-233, which is fissile. If uranium-233 is struck by a neutron it will fission nine times out of ten, releasing 2.5 neutrons and continuing the process of releasing the energy of thorium.

Using the thorium fuel cycle nearly eliminates the production of transuranic waste, which is a major concern in the disposal of nuclear fuel. Because the thorium fuel cycle begins roughly six mass units before the uranium approach, it requires more neutron absorptions before it reaches its first transuranic nuclide, in this case neptunium-237. Because there are two opportunities for fission along this path, in the form of uranium-233 and uranium-235, the theoretical maximum production of transuranics is only 1.5 % and plutonium generation has the potential to be completely eliminated. By contrast, most of uranium fuel is only a single neutron absorption away from plutonium production.

The efficiency at which thorium could potentially be used as an energy source could cause us to rethink some of our opinions about energy resources. Imagine a single cubic meter of material—average continental crust—taken from anywhere in the world. That cubic meter contains, on average, about two cubic centimeters of thorium and half a cubic centimeter of uranium, if each was in its metallic form. If that thorium were converted to energy in a liquid-fluoride reactor, it would be equivalent to the energy in thirty cubic meters of the finest crude oil in the world. Truly, the efficient use of thorium is a transformational technology that can change the energy balance of the world.

Thorium would be used in a liquid-fluoride reactor by generating a thorium tetrafluoride salt similar to the uranium tetrafluoride salt discussed earlier. Thorium tetrafluoride would be dissolved in flibe salt and then introduced into a "blanket" region of the reactor, where it would absorb neutrons and convert to uranium-233. This uranium would be chemically removed from the blanket and introduced into the central fuel salt, where it would be fissioned, releasing energy and generating the neutrons needed to continue fission and produce more fuel. Both the blanket and the fuel could be continuously chemically processed to remove the fission products that built up.

We see a tremendous opportunity for liquid-fluoride reactors to generate energy at high temperatures, low pressures, and low costs. They simplify the fuel cycle and allow the use of any fissile material. They are particularly well-suited to implement the thorium fuel cycle, which can achieve far greater fuel efficiencies that our present use of rare uranium-235.

Alvin Weinberg was the inventor of the pressurized water reactor and the strongest champion of the liquid-fluoride thorium reactor. Dr. Weinberg died in 2006, but in his 1991 autobiography he said: "During my life I have witnessed extraordinary feats of human ingenuity. I believe that this struggling ingenuity will be equal to the task of creating the Second Nuclear Era. My only regret is that I will not be here to witness its success." I hope that we may realize his dreams.

An Industrial View on Thorium: Possibilities, Challenges and Paths Forward

Luc Van Den Durpel

Abstract

Thorium is again the new hype in various debates on sustainable nuclear energy systems but should be demystified, as Th-dedicated systems will take decades to develop and will not be possible without an initial long period of complementarity with the U–Pu cycle and without a government's long-term vision and involvement. There may be a market in the medium term for U–Th and Pu–Th fuels, depending on international requirements for nuclear energy systems and fuels. Areva and Solvay are teaming up to investigate Th fuel options as complement to the U–Pu cycle and to address a holistic thorium valorization approach both in the rare-earth and nuclear energy markets.

Possibilities for Thorium Use in Nuclear Energy Systems

Thorium is increasingly becoming a trendy topic, among others, this is due to its specific technical characteristics and possible synergies among thermal neutron spectrum reactors. Also, in the long term, it offers advantages related to decreased minor actinides production, a higher melting point, a single oxidation state, a better proliferation risk profile, and provides an avenue to "new nuclear".

Most proposals today concern only thorium-dedicated nuclear energy systems. When full account is taken of the fuel cycle development requirements, many of these systems can be tentatively classified as belonging to "Generation 5 and beyond", with exception of India where Th-fuel cycle has been under development since long. Pu and minor actinides management, as well as nuclear sustainability, are also drivers in which the use of thorium can be considered.

However, the socio-political consequences of the present hype entails biased discussions on the potential of thorium and a scientifically and technologically correct debate is needed, starting with the fact that a Th fuel cycle will never exist without, initially and for rather a long time, a complimentary U–Pu cycle. The full claimed benefits of the Th fuel cycle will only be gradually achieved over a period that might last 100 years or more.

There are essentially three major families of scenarios envisaged for the use of Th–^{233}U: complementary use of Th–^{233}U in U–Pu nuclear energy systems; transitioning towards 100 % Th–^{233}U systems coming form generation 3+ systems; and Th-dedicated "Gen 5 and beyond" systems.

Thorium use in nuclear power demands a long-term strategy. Unless a large government-driven program exists, it is doubtful that thorium-optimized nuclear power plants will be available before 2030, given the actual industrial solutions available for investors and the time required to make the UOX/MOX (mixed oxide) systems evolve toward Th–OX-fueled cores. The Th-containing fuel should be complementary with the U–Pu cycle and offer additional flexibility to nuclear power plant operators.

Fuel cycle flexibility is becoming increasingly important, owing to natural uranium availability imbalances, leading to volatile price expectations during the 2030–2050 period, and also owing to the need to reassure investors that fuel availability will not be an issue over the greater lifetime (60+ years) of new nuclear power plants.

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Challenges Ahead

Nuclear power is a hugely capital-intensive industry with a technology lock-in behavior. Also, the switching costs from U–Pu to Th–^{233}U for the nuclear industry are very important. Issues with 100 % use of the Th–^{233}U fuel cycle involve fissile material balance for start-up, recycling, and refabrication (especially with the presence of ^{232}U), as well as not fully settled proliferation risk assessments.

Thorium can, however, improve the U–Pu cycle in several ways, namely the lengthening of the cycle time in light water reactors (LWR) and heavy-water reactors (PHWRs), reducing the use of naturally occurring uranium (nat-U), providing additional multi-recycling options for Pu and reprocessed uranium, and the replacement of burnable poison in specific cases.

Thorium use should be progressive and the "thorefied" fuel cycle should be coupled neutronically, but not physically or chemically, with the U–Pu cycle until a market for the Th fuel cycle fully develops. Multiple paths exist before embarking into a Th cycle, with some already providing complementarity with the U–Pu-cycle in the medium term.

Some of Areva's assessments of Th options in LWR, based on EPR-design evolutionary fuel options (with potential Th use), show that some 20 % of energy can be provided by ^{233}U. A reprocessing delay, and a reduction in the use of nat-U in a recycling scheme can also be expected, as well as lower enrichment needs and minor actinides production. Also, an improved Pu balance in UOX and MOX for the multi-recycling of Pu can be achieved.

Areva has been gathering sizable expertise working with thorium since the 1970s and it holds an inventory of 2300 t of Th (nitrate form) in France. The company's experience extends to the fabrication of Th fuels for pressurized hot water extractors (PHWEs), high-temperature reactors (HTRs), and LWRs and it keeps a ceramic lab available and equipped for Th fuel R&D.

Solvay, worldwide leader in separated rare-earths, holds an inventory of 6200 t of Th (nitrate and hydroxide forms) in France coming from historical monazite ore processing. The company's experience includes industrial thorium purification processes (solvent extraction) and high-grade finished thorium products manufacturing.

Paths Forward: Thorium Can Have a Place in a Growing Nuclear Energy Future

Nuclear energy is a prime contributor to address climate change and energy sustainability objectives worldwide, provided that thorium is introduced progressively and in a complementary manner to the U–Pu cycle, and that such introduction be assessed industrially to ensure its efficiency on a technical and economical basis.

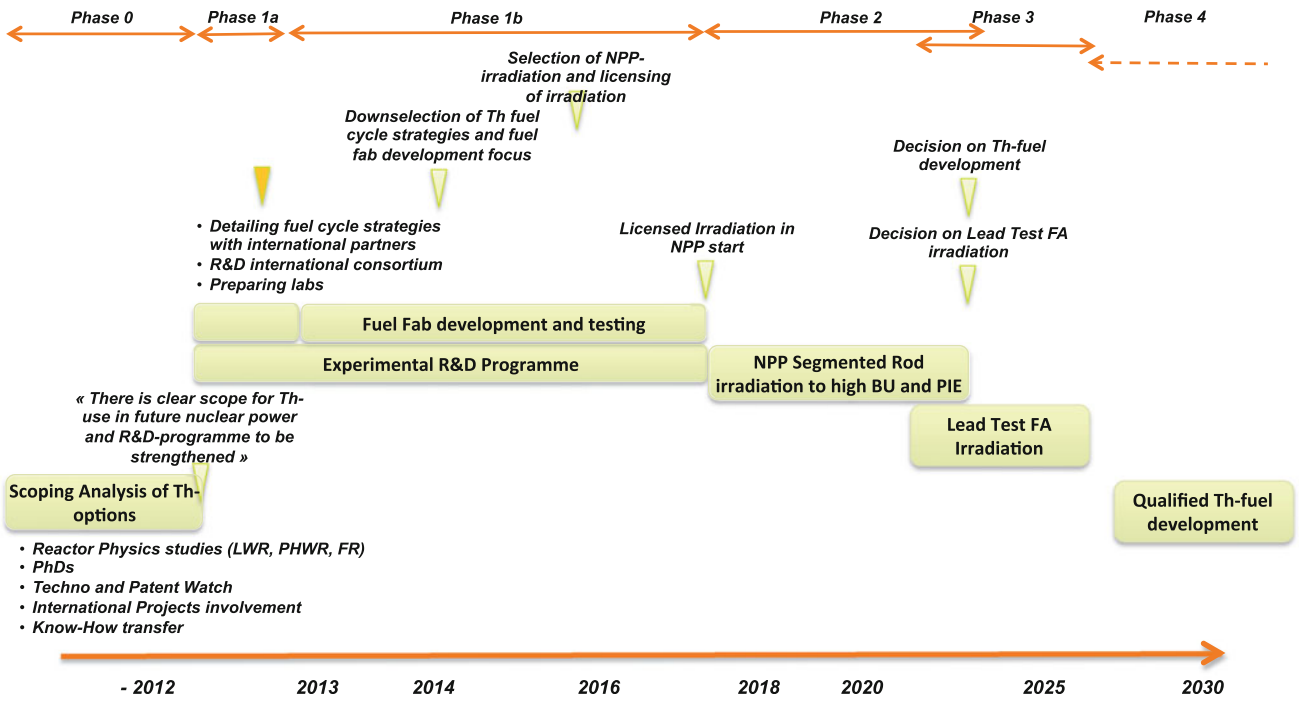

Fig. 1 Timetable of Areva–Solvay's envisaged R&D program on Th valorization in nuclear energy. *Source* Luc Van Den Durpel, ThEC2013—Industrial View on Thorium, 28–31 October, 2013, Areva/Solvay

Given the overall worldwide developments related to thorium, both in the nuclear energy field as in the rare-earth market, Areva and Solvay are announcing a collaboration as they join their know-how to add value to thorium's entire life-cycle. Both companies together master the complete set of thorium valorization routes in the short- to longer-term region with clear synergies that ensure thorium valorization services will also be provided to third parties. This collaborative program encompasses, among others, an industrially robust valorization argumentation focused on thorium valorization in nuclear power in the medium term. An R&D program focused on medium-term thorium valorization in nuclear power is being set up with international R&D partners geared towards the first phase of fuel development with irradiation by 2020 (Fig. 1).

A Global and a Turkish Perspective of Thorium Fuel for Nuclear Energy

Muammer Kaya

Natural Properties of Thorium (Th) (Earth's Forgotten Treasure)

Thorium occurs naturally. It is named for the Norse god of thunder and identified almost two centuries ago. Thorium exists in nature almost entirely as the stable Th-232 isotope and cannot sustain a nuclear chain reaction itself. Thorium is a lustrous silvery-white metal. It is a dense and only slightly radioactive actinide (i.e., less radiotoxic). Thorium is a fertile material; it can accept a neutron and transmute into fissile U-233 and decays to stable Pb-208. Thorium is usually a by-product of the rare-earth element (REEs) containing monazite or bastnasite minerals.

Thorium Basics (New Green Nuke Nuclear Renaissance)

Thorium is plentiful in nature, generally well distributed throughout the earth's crust, and virtually inexhaustible nuclear fuel. Th is one of only a few substances that act as a thermal breeder. It is virtually impossible for terrorists to make nuclear weapons/bombs from Th (i.e., peaceful). Therefore, nuclear power without proliferation has clear political appeal for many governments in the world. Th has a high heat capacity (i.e., smaller reactors). It does not require costly mineral processing methods (i.e., less expensive). Th is an extraordinarily efficient nuclear fuel (i.e., allows longer fuel burnups in reactors). Th produces more neutrons per collision; thus, 20–40 times more energy is generated, less fuel is consumed, and 47 times less radioactive nasties are left behind. Th fuel is completely used up in the reactor; thus, there is relatively little nuclear waste to take care of. Th reactor wastes need to be stored for only a few hundred years, not for a few thousand like that of uranium (U) reactors.

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Th reactors have zero risk of melt down as opposed to conventional U reactors. Th reactors can also use liquid fuel, which has significant advantages in operation, control, and processing over solid fuels used in U reactors. Liquid fuels work at high temperature without pressurization. Owing to the above advantages of Th, it should be at the heart of nations' atomic power effort.

Why Thorium? (New Era of Safe, Clean, and Affordable Energy)

There is a threat of climate change and there is an urgent demand for carbon-free energy/electricity in the world. Th is 3–4 times more abundant than U in nature and is distributed evenly in most countries in the world. Th can be mined with relatively low cost and by using environmentally friendly mining methods from high grade alluvial deposits. Th extraction is relatively straightforward and inexpensive. Th has a better radiation stability and longer fuel cycle. Th has a higher energy density and fuel economy in reactors. Th fuel cycle waste has a lower radiotoxic period owing to the absence of transuranic wastes (i.e., Pu, Np, Ce, and Am). Th Mixed OXide (TMOX), Pu + Th or U + Th, fuel can be burned in any reactor that is licensed to use Mixed OXide (MOX) fuels. There is a huge spent toxic material/Pu stockpile that has been built up from U fuel. Th fuel can be used to safely incinerate these unwanted accumulated old stockpiles. The world's global energy needs for one year can be supplied by burning approximately 6000 tonnes of Th.

Uranium (Today's Problematic Nuclear Fuel)

Uranium is today's preferred conventional nuclear fuel in commercial reactors. U has been adequate to meet today's energy supply needs. The world's U reserve is about 5.5×10^6 tonnes; yearly production of U is about 65×10^3 tonnes. There are 19 major U producers in the

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Table 1 World thorium reserves according to OECD/NEA, 2001 [1]

Country	RAR Th (tonnes)	EAR Th (tonnes)
Brazil	606,000	700,000
Turkey	380,000	500,000
India	319,000	–
United States	137,000	295,000
Norway	132,000	132,000
Greenland	54,000	32,000
Canada	45,000	128,000
Australia	19,000	–
South Africa	18,000	–
Egypt	15,000	309,000
Other countries	505,000	–
World total	2,230,000	2,130,000

Table 2 Properties of Th- and U-containing rare-earth minerals

Phosphate mineral	Formula	Density (g/cm^3)	Magnetic property	Weight (%)		
				REO	THO_2	UO_2
Monazite (Ce)	(Ce, La, Nd, Th)PO_4	4.98–5.43	Paramagnetic	35–71	0–20	0–16
Monazite (La)	(La, Ce, Nd, Th)PO_4	5.17–5.27	Paramagnetic	35–71	0–20	0–16
Monazite (Nd)	(Nd, Ce, La, Th)PO_4	5.17–5.27	Paramagnetic	35–71	0–20	0–16
Silicate Mineral Thorite	Formula (Th, U) SiO_4	Density (g/cm^3) 6.63–7.20	Magnetic Property Paramagnetic	REO <3	THO_2 –	UO_2 10–16
Carbonate mineral Basnasite (Ce)	Formula (Ce, La, Th)$(CO_3)F$	Density (g/cm^3) 4.90–5.20	Magnetic property Paramagnetic	REO 70–74	THO_2 0–0.3	UO_2 0.09
Basnasite (La)	(La, Ce, Th)$(CO_3)F$		Paramagnetic	70–74	0–0.3	0.09
Basnasite (Y)	(Y)$(CO_3)F$	3.90–4.00	Paramagnetic	70–74	0–0.3	0.09

world. The life expectancy of U ore is about 50–60 years. U ores are generally not harmful, but it is dangerously toxic to humans if ingested, inhaled, or even upon prolonged contact. Conventional U reactors require the extremely rare ^{235}U isotope (abundance: 0.7 %), which must be purified/enriched from the natural ^{238}U isotope. U mining, enrichment, and extraction are expensive and complex compared with Th. U fuel leaves a large amount of toxic wastes containing Pu-239, which can be used to make bombs and weapons. However, the used/depleted fuel can be reprocessed by expensive Pu–U extraction (Purex) process to remove fissile material and refabricate new fuel elements. In the near future, R&D on U will fade away in favor of Th. Reasonably assured reserves (RAR) and estimated additional reserves (EAR) of Th as given by the OECD/NEA are shown in Table 1. The total RAR world Th reserve is about 2.23 and the EAR reserve is about 2.13 million tonnes.

Thorium-Containing Rare-Earth Elements (REEs)

Radioactive Th- and U-containing REEs may be phosphate-, silicate-, or carbonate-containing minerals. Their formulas, densities, magnetic properties, and chemical contents are given in Table 2. Monazite contains up to 20 % ThO_2 and 16 % UO_2, whereas bastnasite contains 0.3 % ThO_2.

Monazite (REE-Th)PO_4

Monazite is a phosphate mineral with a rare-earth oxide (REO) content of 70 % (Ce, La, Pr, and Nd). Monazite is found in granites, syanites, pegmatites, beach sands, etc. It includes 3–14 % ThO_2 and a variable amount of UO_2. Th is found in placer deposits, beach sands, and is also a

component of the world's biggest REE deposit, Bayan Obo, in China. The world's yearly monazite production is between 5000–6500 tonnes. Th from monazite deposits can be mined with low cost and with environmentally friendly mining methods because it exists in high concentrations and grades at the surface or in beach sands. The ore/sand can be skimmed off the surface by dredge mining. Monazite mining is more economical owing to presence of Th as a by-product in the REE ore. A U mine can be developed in 10–15 years, whereas a Th mine can be developed in 2–3 years owing to less regulatory scrutiny.

ThO_2 is liberated from monazite by gravitational, magnetic, and electrostatic concentration methods, employing a minimum of dangerous chemicals (i.e., concentrated acids). On the other hand, U is mined by expensive underground mining, or open pit mining, and in situ acid leaching methods. The world's major monazite producers are in Australia (Mt Weld), India (Andhra Pradesh, Tamil Nuda, Odisha), China (Guandong), Malaysia, and Brazil. Figure 1 shows a typical conventional beach sand treatment flowsheet to recover quartz, garnet, magnetite, zircon, ilmenite, rutile, and monazite, which contains both Th and REOs. In this flowsheet, gravity, low and high intensity magnetic separation, and electrostatic separation methods are used.

Bastnasite (REE-Th)FCO_3

Bastnasite is a fluorocarbonate-containing mineral with a REO content of approximately 70 % (Ce, La, Pr, and Nd). Bastnasite is the primary source of light REOs and accounts for 80 % of the overall amount of REO in the world. In the last 50 years, bastnasite has replaced monazite as the chief mineral source of REEs. Bastnasite has been extracted on a large scale only in Mountain Pass, USA, and Bayan Obo, China, which supplies 45 % of the world's demand alone. The production uses expensive underground mining methods. Mineral processing of bastnasite ores depend on fine grinding; gravity, magnetic and flotation separations, calcination, HCl leaching, and solvent extraction (SE) steps.

Reserve Development Studies, Operation and Production Plans of Eskişehir-Sivrihisar-Kızılcaören Light REE + Th Field by Eti Mines

Figure 2 shows the Th and U deposits in Turkey. There are five U and one Th deposits. Figure 3 shows a comparison of some REO deposits according to reserve size. The Eskişehir

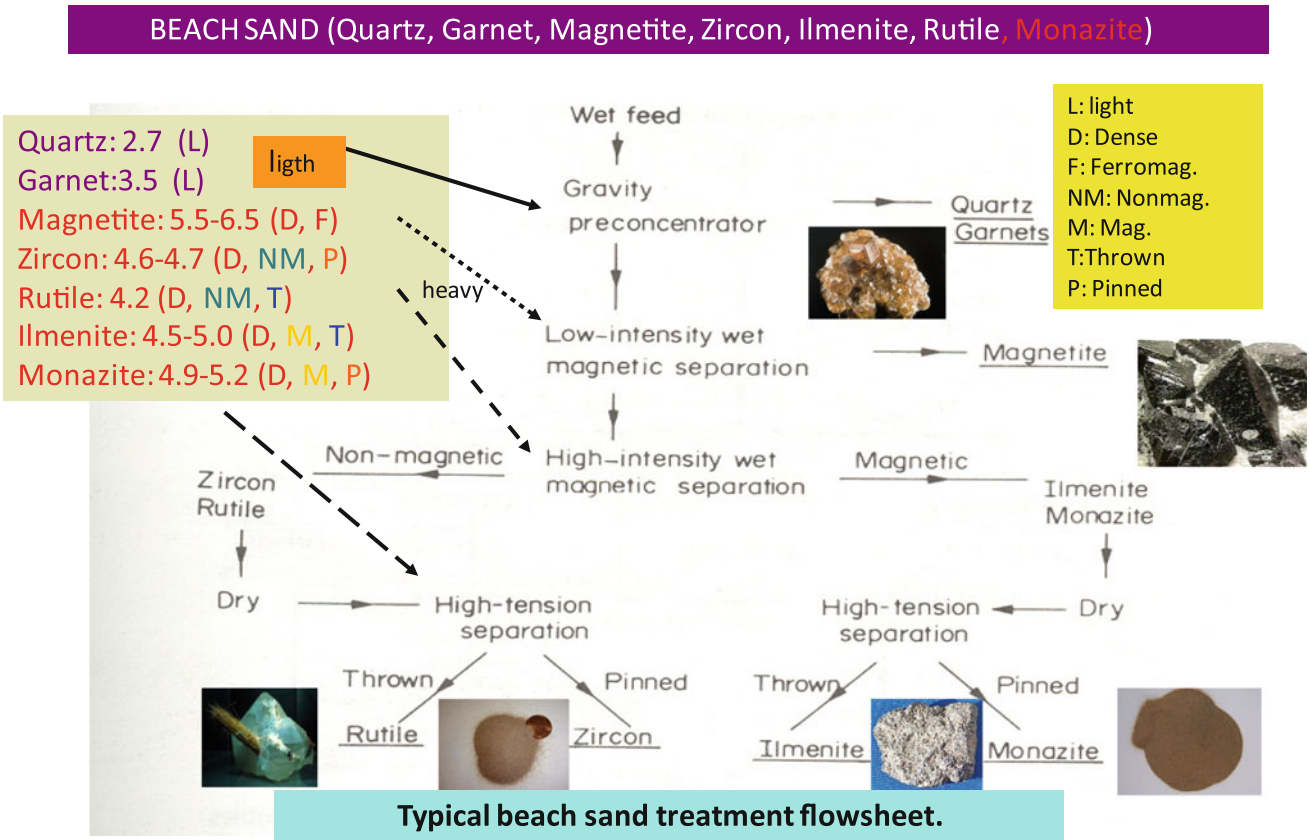

Fig. 1 Typical beach sand treatment flowsheet

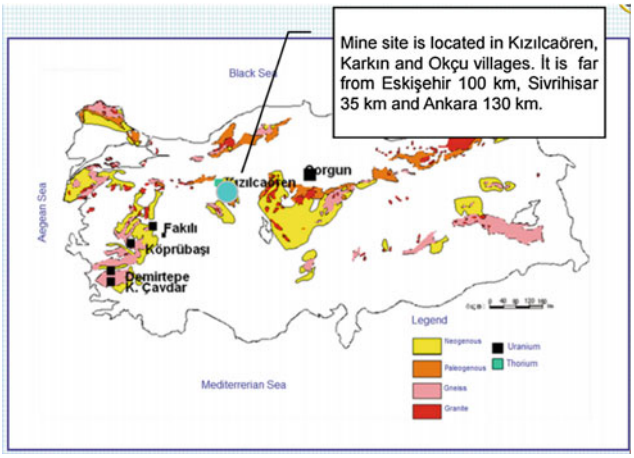

Fig. 2 Thorium and uranium deposits in Turkey

Th deposit is the eleventh largest Th reserve in the world. In terms of the ThO_2 content of REO deposits, the Eskişehir Th deposit is the third largest deposit in the world (Fig. 4).

The Eskişehir Th deposit was discovered by using an air prospecting survey by the General Directorate of Mineral Research and Exploration of Turkey (MTA) in 1959. Initial reserve calculations depended on drilling studies to a depth of 50–200 m at that time. The ore produced is a complex and finely disseminated bastnasite mineral. The ore is found as layers, lenses, and veins in addition to impregnation and brecciate zones. The ore reserve is more than 30 million tonnes. Bastnasite ore contains 37.44 % fluorite, 31.04 % barite, and 3.9 % REE (2.01 % Ce + 1.5 % La + 0.39 % Nd). The average ThO_2 content is 0.21 %.

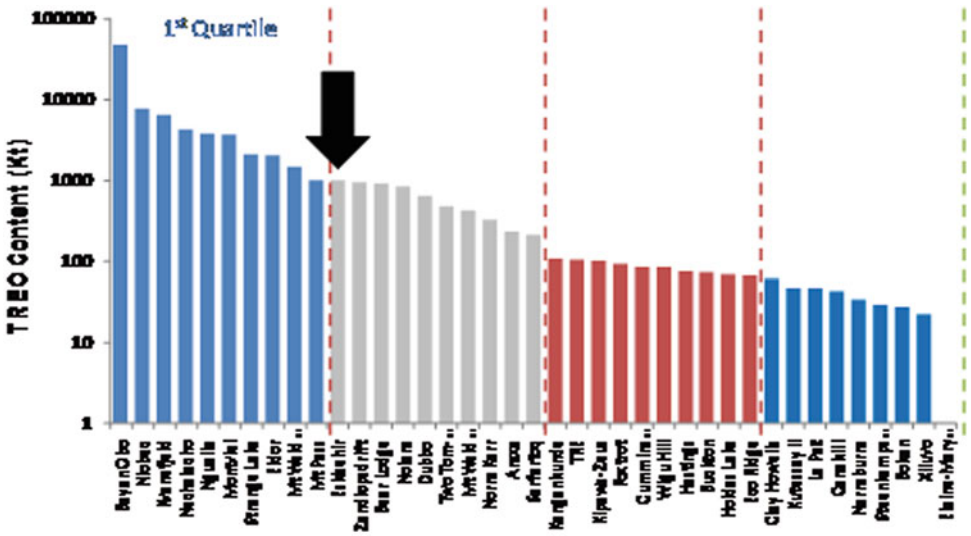

Fig. 3 Comparison of REO deposits according to reserve size

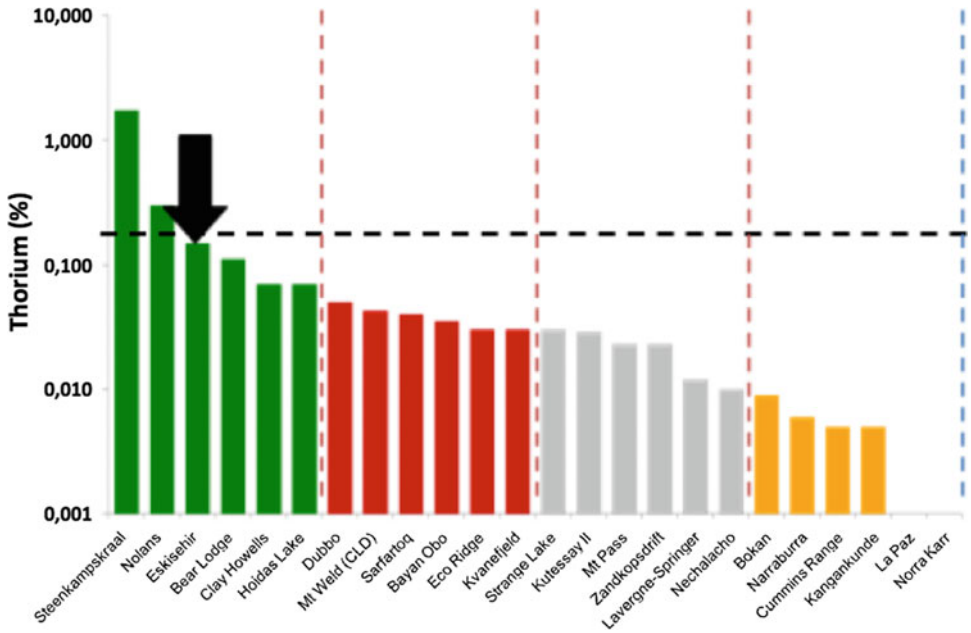

Fig. 4 Comparison of the ThO_2 content of REO deposits

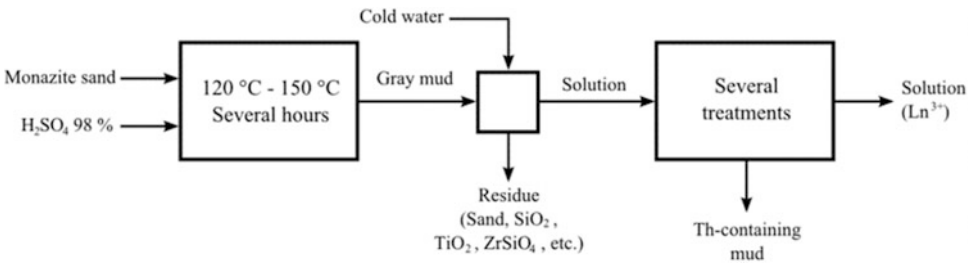

Fig. 5 Monazite extraction with hot concentrated H_2SO_4

The ore also contains psilomelan, ilmenite, rutile, and some mica minerals. The proved and probable ThO_2 reserve is about 380,000 tonnes. In 2011, two 500 m deep drilling bores were opened. Analyses showed further reserves at this depth. In 2012, 10,000 m drilling was performed at 38 locations. Between 2014 and 2017; 116,000 m drilling will be performed at 289 locations. 45,000 samples will be taken for geochemical analysis and 1000 samples will be taken by X-ray diffraction (XRD) analysis.

The operating license of the Th deposit is owned by the Turkish Government Mining Company of Eti Mines for 10 years. Eti Mines prepared a feasibility report for 10,000 tonnes/year REO, 72,000 tonnes/year barite, 70,000 tonnes/year fluorite, and 250 tonnes/year Th productions. Open pit mining methods will be carried out by Eti Mines. Mineral processing will be performed by domestic/foreign private expert companies with service procurement tenders for 10 years. Products will be marketed by Eti Mines. ThO_2 will be stored by Atomic Energy of Turkey until a Th reactor is established in Turkey.

Monazite Cracking/Opening with Hot Concentrated H_2SO_4

Th has been extracted from monazite through a complex multistage process (AMEX). Monazite is dissolved in hot concentrated H_2SO_4 (120–150 °C) for several hours (Fig. 5). Th is extracted as a water-insoluble residue into an organic phase containing an alkyl amine. Then, it is separated using an ion such as nitrate, chloride, hydroxide, or carbonate, returning Th to the aqueous phase. Finally, Th is precipitated and collected. Th extraction is a complex process because of its similar chemical properties with other REOs.

Gravity separation, magnetic concentration, flotation, and acid leaching tests were carried out for Eskişehir Th ore by Ipekoğlu [2]. Optimum leaching conditions were found to be 200 kg/t HCl dosage and 3 h leaching time. The highest Th grade was 99 %. The Thorex process is used for ThO_2 recovery from used U-233 or Th fuels. Hot acid leaching with 13 M HNO_3, 0.05 M HF, 0.1 M $AlNO_3$ at their boiling temperatures can be used. After centrifuging, a Th $(NO_3)_4$ + $UO_2(NO_3)_2$ solution is separated. Solvent extraction columns and purification steps follow. Figure 6 shows the acid leaching, solvent extraction, ion exchange (IE), and calcination flowsheet for ThO_2 production.

Yorukoglu et al. [3] studied the effect of roasting and thiourea ($CS(NH_2)_2$) addition on bastnasite leaching of the Turkish Th ore. Firstly, scrubbing followed by barite and fluorite flotations to obtain a pre-concentrate containing 21.3 % REO were carried out. Leaching with 8 M HNO_3 at 70 °C achieved 100 % REO dissolution, whereas 15 M H_2SO_4 at 25 °C achieved 89.25 %, and 2.5 M HCl at 55 °C achieved 82.2 % REO dissolution. Dilute acid leaching of the bastnasite pre-concentrate was performed at a particle size of 0.053 mm, H_2SO_4 concentration of 1–3 M, a solid/liquid ratio of 1:20 g/mL, and a leach temperature of 25 °C. Leaching with 3 M H_2SO_4 before and after roasting at 750 °C for 1 h achieved 8.5–47.4 % REEs content. Roasting decomposes the fluorocarbonates and oxidizes Ce in the

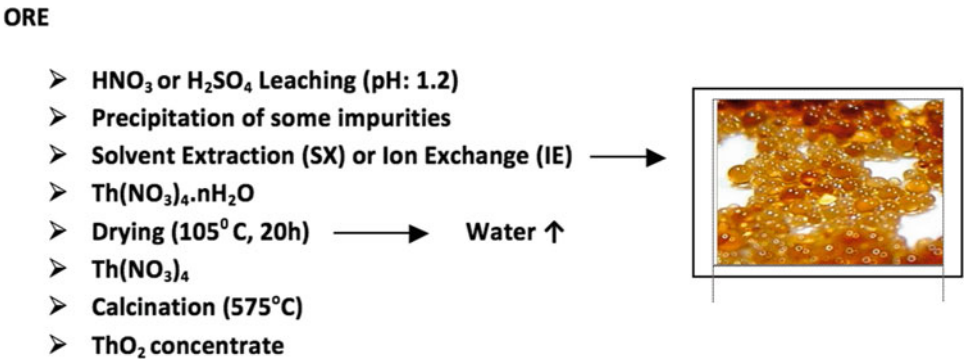

Fig. 6 Thorium concentration flowsheet

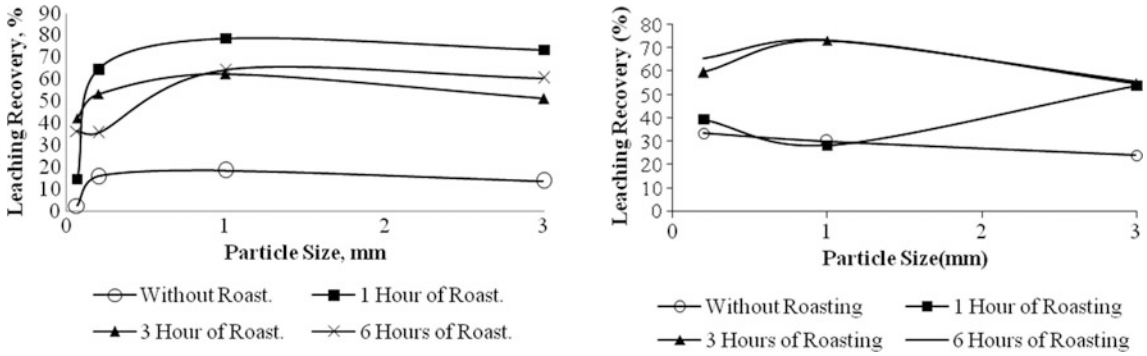

Fig. 7 Effect of particle size and roasting time on leaching recovery with 3 M H_2SO_4 (*left*) and HCl (*right*)

pre-concentrate with the formation of fluorocerite and cerianite as new phases (verified by XRD results). However, addition of 1–2 M thiourea into the leaching medium after roasting improved the dissolution from 18.1 to 89.7 % (at 3 M H_2SO_4 and 1 M thiourea). The filtrate obtained from the leaching experiment of roasted pre-concentrate was precipitated as a hydroxide–oxalate mixture (mish metal) and ignited to oxides at 900 °C for 2 h and a total REE content of 93.6 % and REO content of 92.6 % were obtained [4]. The effects of particle size (0.065 to 3 mm), oxidizing roasting temperature (900 °C), and time (1–6 h) on Th recovery were investigated for a leaching process conducted with 3 M H_2SO_4/HCl for 3 h at 25 °C [5]. Figure 7 shows the effect of roasting time and particle size on leaching recovery with H_2SO_4 and HCl, respectively. The highest Th recovery was 78.74 % when 3 M H_2SO_4 and 1 mm particle size samples were used and 1 h roasting applied. For HCl leaching, the best results (73.38 %) were obtained with 3 M HCl, 1 mm particle size, and 3 h roasting. Overall, 3 M H_2SO_4 achieved better leaching recovery than HCl. H_2SO_4 is also cheaper than HCl. Oxidizing roasting at 900 °C for 1 h, before atmospheric leaching is necessary for the Eskişehir Th ore. The best particle size for leaching is 1 mm. 79 % REEs + Th dissolution can be achieved.

Conclusion

Energy is a major concern for developing Turkish society. Turkey today exports more than 70 % of its energy as fossil fuels. In the last 50 years, Turks have become skeptical, anxious, and indecisive about using nuclear energy for electricity production. In the last 10 years, thorium has been brought to Turkish government's attention and national discussions have begun as to its use as a new, safe, clean, affordable, CO_2-free, alternative, strategic, and domestic nuclear energy fuel. In Turkey, funds to support education and R&D on Th are sought by some scientists and a Thorium Research Center (ThoReC) for Th mining, enrichment, extraction, fuel fabrication, prototype reactor testing, and nuclear fuel cycle testing should be established.

Turkey recently contracted the development of four VVER-1200 type Russian nuclear power reactors using U as a fuel in Akkuyu (Rosatom) and another four units will be established in Sinop in the next decade by Japan (Mitsubishi-Westinghouse). The Turkish government will be able to use Th along with U as an alternative fuel in the newly constructed reactors. If Turkey can process its important domestic low-grade Th-containing bastnasite resources in Eskişehir, it can supply its own Th fuel for more than 100 years and become self-sufficient in its energy source.

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Opportunities and Challenges for Thorium in Commercial Molten Salt Reactors

J.D. Turner and A.J. Donaldson

Abstract

The commercial potential for molten salt reactors (MSRs) fuelled with thorium is discussed. Specific technical, regulatory, and organisational challenges are highlighted, which could prevent, or limit, commercial realization. These include materials, safety, reprocessing, and proliferation concerns. Potential issues are also discussed regarding the MSRs' likely competitors, and how this may affect research and development. It is concluded that thorium and MSRs are complementary technologies, and that the introduction of the thorium fuel cycle will likely be tied to MSR development.

Introduction

Thorium and molten salt reactors (MSRs) are often considered complementary technologies, as MSRs appear to be exceptionally well-suited for operation with a thorium fuel, and have been successfully demonstrated as such historically. Thorium fuel is most attractive as a fuel option when high burnups can be attained, and in conventional reactors this is often limited by the performance of cladding materials at high radiation doses. MSRs suffer no such limitation, and are, therefore, ideally suited for operation with thorium fuels.

Within the present work, several potential MSR deployments have been considered, based largely upon the availability of information within the literature. MSR projects vary widely internationally in both technical specifications and commercial status, with a range of designs developed both from small commercial entities and national laboratories.

MSRs utilising a thorium fuel may offer several advantages over current state-of-the-art nuclear technology, although the extent of many of these advantages remains unproven in a commercial setting.

Furthermore, technical concerns exist around MSR operation, namely in the areas of materials performance, safety, fuel reprocessing, and proliferation. Although it is certainly possible to address the vast majority of these concerns given appropriate time and funding, such resources are not assured and will require effort drawn largely from national research programmes.

Although a thorium-based MSR system does potentially offer several advantages over existing technology, these must be considered on a commercial basis, as opposed to a purely scientific or aesthetic selection. For example, commercially, improved safety over currently licensed plants is only a real advantage if it represents significantly lowered capital or operational costs, despite its clear benefit in the event of an accident. Commercial factors must, therefore, be taken into account without bias towards or against the system, in order to highlight technical areas that may be improved to attract potential private funding.

What Is 'Commercial'?

To discuss the prospects for a 'commercial' MSR system based on thorium fuels, it is necessary to define what is meant by 'commercial' in this instance.

It should not be taken to imply a particularly sized reactor, as both large-scale and small modular reactor

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(SMR) variants offer advantages over their counterpart in the correct setting.

In general, consideration has been given as to what issues remain that prevent or hinder bringing thorium-based MSRs to the commercial market, where they could be offered as a product by a reactor vendor and, therefore, be licensed by an operator.

The discussion of 'commercial' limitations should also not be taken to imply that such issues must be solved by private entities. Indeed, the scope of the problem is considered to be sufficiently large that only national research interests possess sufficient resources to offer an appropriate solution.

The Advantages of Thorium

MSRs have been demonstrated both with a thorium–uranium fuel cycle and with enriched uranium fuel. As they are clearly able to operate with either fuel cycle, a consideration of the advantages of the thorium fuel cycle within MSRs is useful.

In a light water reactor (LWR), the use of thorium enables a decrease in the initial load of fissile material required at reactor start-up because the benefit from extra fissions following conversion of fertile material is greater with thorium. This benefit can be further improved as burnups are increased. The value of thorium in a thermal spectrum increases with the ability of cladding material to achieve high burnup, and, hence, an MSR design is ideally suited to the use of thorium fuels, as cladding restrictions are no longer relevant.

Economically, thorium fuel may be added to an MSR as a steady fissile load, which can be considered a non-recurring investment. The thorium contribution to energy output may be relatively high (compared with alternatives) even if true breeding is not achieved.

Furthermore, the use of the thorium cycle is known to produce lower quantities of very long-lived waste compared with the uranium–plutonium fuel cycle, a fact that may be highly advantageous from both a political and economic viewpoint, but would require substantial investment in specialized fuel cycle facilities.

The Advantages of Molten Salt Reactors

The advantages of MSRs over conventional modern reactors are typically given by the concept's proponents as [1–3]:

1. Fuel can be added as required;
2. Troublesome isotopes can be removed during operation;
3. Fissile inventory may be removed from the core by using passive systems, which may form part or all of the reactor shutdown mechanism;
4. Molten salts have a high boiling point, which enhances thermodynamic and safety considerations;
5. The core lifetime is dictated by moderator performance, rather than a fuel burnup limit;
6. Fuel salt density decreases with increasing temperature;
7. A broad range of fuel options is available, with no fuel fabrication required;
8. MSRs operate at a high temperature compared to LWRs;
9. The thorium fuel cycle offers enhanced proliferation resistance.

These points are typically taken 'as read' by many researchers within the field, and are mentioned (in various forms) as the key benefits of MSR systems over LWR counterparts. If these advantages can be both demonstrated and proven, they would represent a significant improvement over current technology. However, this should be considered potentially problematical, as many of these benefits are either the focus of technical concerns or require some further evidence of their extent, as discussed in the following sections.

Projects Considered

It has already been noted that the authors consider that the overwhelming majority of the required research and development that remains to be done on the MSR concept will require national research programmes. This is due largely to the scale and nature of the work required.

As such, the projects considered for the present analysis were the following nationally funded MSR research programmes:

- France—The MSFR, a fast-spectrum molten salt, with an altered salt composition from that historically demonstrated [4, 5];
- Russia—MOSART, an actinide burning MSR, probably with limited commercial interest [6];
- China—Limited information available, but apparent recent interest. Assumed to be probably a thermal spectrum reactor. Unknown timescale for deployment;
- Japan—The Thorium-NES programme, a high-technology concept requiring the development of accelerator-driven breeding systems to fuel graphite-moderated MSRs;
- India—Early plans closely based on Oak Ridge National Laboratory's Molten Salt Breeder Reactor (ORNL's MSBR), a long-term plan intended to utilize India's large thorium reserves [7].

Technical Concerns

From these concepts some general and more specific technical concerns have been highlighted:

Firstly, the technology basis for the majority of MSR projects appears to be founded on the successful operation of historical demonstration plants at ORNL. Although this is certainly an admirable achievement, the operation of such historical demonstrators cannot reasonably be considered sufficient to ensure current regulatory or technical feasibility, nor commercial success. The requirements for nuclear designs have changed significantly since the operation of the demonstration MSRs at ORNL, and significant reworking of their technical basis would likely be required, which is not always considered in MSR development programmes.

Furthermore, the majority of the literature compares MSRs to existing LWR technology. Advantages over LWRs may not be sufficient to ensure commercial attractiveness for investors and vendors, given the alternative Generation IV concepts that are likely to be at a similar (or more advanced) state of technological readiness.

Comparison with Alternative Generation IV Projects

From the previous list of MSR advantages, several can be said to apply to all or most of the Generation IV systems.

All liquid metals have high boiling points, and significantly more experience exists with liquid-metal-cooled fast reactors than with MSRs, for example. All Generation IV concepts are, by definition, relatively flexible with regards to the fuel cycle required, and are able to accept a range of fuel by design. All are also able to operate at higher temperatures than current LWRs, and all can be said to be proliferation resistant.

This is not to claim that a thorium-fuelled MSR would offer no advantage over an alternative Generation IV system, but rather to suggest that greater focus must be placed on the unique operating features of the MSR compared with Generation IV alternatives, rather than compared with LWR technology.

Materials

As for other Generation IV reactors, materials improvements are a research requirement.

Hastelloy-N is a nickel-based alloy that was successfully demonstrated at ORNL and may be considered for service today with some alloying additions, which appear to be well understood [8].

One area of concern for MSRs is that these modifications reduce the maximum operating temperature to around 650 °C. Given the relatively high melting point of fuel salts, and if an approximate temperature rise through the core of 100 °C is assumed, with a reasonable 50 °C temperature margin maintained below the maximum at the core outlet, then the inlet temperature is likely to be very close to the salt's freezing temperature, and possibly below, depending on the salt employed. It is, therefore, likely that alternative alloys would be required for commercial operation. Success is not guaranteed in efforts to develop these improved materials, although some developments appear to show promise.

Safety

Drain Tank/Freeze Plug System

The main safety feature within most MSR concepts is the passive removal of fissile inventory from the core. Technical concerns for this mechanism centre on the need for cooling the fissile material following draining from the core, the effectiveness of such systems at removing fissile inventory in a satisfactory manner and the licensing impact their use would have.

A plug of frozen salt is the usual mechanism described to act as a passive drain for the fuel salt within the primary circuit. Although this is an attractive option on first inspection, studies have suggested that it may take several minutes to operate [9, 10]. Given the previously highlighted concerns regarding the temperature margins available for normal operation, a safety feature acting on such a timescale may not be quick enough to drain material from the core before the structural alloy begins to melt.

A drain tanks operation will also be hampered by the system thermodynamics, as the hottest salt will naturally be found at the top of the core, which increases these difficulties. A freeze plug located at the bottom of the core will also take longer to melt during a high-temperature transient, as it will be in contact with salt at the lowest temperatures within the primary circuit.

Alternative mechanisms currently available, such as the inclusion of a simple mechanical valve or burst disc, may be more easily employed, but lack the elegance of the freeze plug system. The identification of a suitable passive mechanism, preferably acting rapidly on temperature, rather than pressure, is a key requirement for MSR deployment on a commercial basis.

Once drained, the fuel salt will still possess significant decay heat, and will require cooling. Although this can be performed passively by using well-understood technology, similar to that used to passively cool LWR cores under severe accident conditions, this limits the benefit that the core drain system provides both functionally and economically. It is not clear as to the benefit of the core drain system over a currently used control rod shutdown, given that the need to cool the fuel passively remains the same, regardless of its location.

Temperature Coefficient of Reactivity of Fuel Salt

The increase in fuel salt density with temperature is typically described as another key safety feature of an MSR, as it is typically taken to imply that such a system would have an inherently negative change in reactivity with temperature. Such negative feedback is an important step towards reactor control and licensing. However, positive feedback coefficients are predicted for some MSR systems, owing to the unique combination of expanding fuel salt and high temperature moderator [11]. Indeed, some national research programmes have moved away from a thermal spectrum MSR for this reason.

Furthermore, and potentially more concerning from a licensing standpoint, is the historically discussed suggestion that a system with a strong negative feedback coefficient and a drain-tank system does not require traditional control rods. Although a case may eventually be made for such a claim, it is unlikely to be economically significant enough to warrant the research required to demonstrate its validity, especially given that a regulator unfamiliar with MSR practices would likely require control rods be included in the first instance regardless.

Reprocessing

On-line reprocessing, such that fission products are removed from the fuel salt in a continuous process, is an extremely attractive proposition for various reasons. The effect of such reprocessing on fuel breeding rates means that it is in fact required for some concepts to function as intended.

Where reprocessing is required, this represents a significant technology risk. Although reprocessing concepts have been outlined, this has only been at a preliminary level [11]. The demonstration of these techniques on an industrial scale appears likely to be very expensive and initially highly complex, and is, therefore, likely to be prohibitively expensive from a commercial standpoint.

Although promising avenues for reprocessing do exist, suitable demonstration of their effectiveness is required to provide confidence in the proposal.

Proliferation

Proliferation resistance has recently been highlighted as a potential concern with the thorium–uranium fuel cycle [12]. Although there is little to suggest that thorium-fuelled reactors are unavoidably more susceptible to proliferation than those operating on the uranium–plutonium fuel cycle, the often quoted ‘inherent proliferation resistance’ of thorium may be much more limited than previously thought.

Commercial Considerations

The fuel burnup limit typically determines the required time between refuelling outages in LWRs. Although refuelling could be removed as the main cause of system outages in an MSR, which would improve the system economically, the extended outages that result from needing to replace a substantial volume of in-core material may outweigh this, given that thermal spectrum MSR designs typically require replaceable graphite-moderating structures. An increase in graphite waste volume is also likely to be of concern, as no suitable disposal route has yet been identified for irradiated graphite.

A further, and often overlooked potential benefit of fuel salt expansion in the core, is that of automatic and intrinsic load following, which may be controlled from a turbine plant, for example, and has the potential to exceed LWR abilities. Such a system would require a deeper understanding of the feedback mechanism and confidence that future behaviour could be predicted with very little uncertainty.

Although MSRs offer high temperature operation when compared with LWRs, the timescales involved in their deployment is likely to ensure that direct competition largely consists of alternative Generation IV concepts. In this regard, a MSR is relatively limited in temperature output compared with, for example the Very High Temperature Reactor (VHTR). As such, although high temperature heat output may be preferable for many tasks, it is not unique to MSRs.

Fundamentally, for MSRs to be attractive now, they must out-compete existing technology sufficiently that they are worth significant initial investment. A range of possible areas of competition exist, but both the technical feasibility and the scope of the MSR offering must be defined more completely before a fully informed decision could be made.

Conclusion

The successful introduction of the thorium fuel cycle is likely to be tied to the introduction of MSRs, owing to the complementary nature of the two technologies.

Further work is required to quantify what real-world commercial benefits an MSR concept would provide.

It is also necessary to confirm the real-world feasibility of the advantages commonly ascribed to MSRs.

Concerns must be addressed regarding materials, safety, reprocessing, and proliferation before widespread commercial interest is expected to emerge.

Fundamentally, the MSR must provide some benefit to industry above contemporary technology for investment to be attractive.

For MSRs to progress, national programmes must develop an MSR concept to the point at which commercial investment becomes viable.

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Current Czech R&D in Thorium Molten Salt Reactor (MSR) Technology and Future Directions

Jan Uhlíř and Vlastimil Juříček

Abstract

Molten salt reactor (MSR) systems and thorium–uranium fuel cycle technology have been under theoretical and experimental development in the Czech Republic since 2000 and 2005, respectively. The investigations have been realized by a consortium of Czech institutions and companies supported by the Ministry of Industry and Trade. The program has covered theoretical and experimental activities in MSR physics, fuel salt and fuel cycle chemistry, on-line reprocessing technology, molten salt thermo-hydraulics, structural material development, and testing of apparatuses for molten fluoride salt media. As the existing development was relatively independent, current intentions in this field are moving towards the collaborative R&D of fluoride-salt-cooled, high-temperature reactors (FHR) and MSR technology together with US institutions.

Introduction

The technology of nuclear reactor systems with liquid molten salt fuel has been investigated in the Czech Republic since 1999. The original effort came from the national partitioning and transmutation concept based on the subcritical accelerator-driven system (ADS) for incineration of trans-uranium elements with liquid fluoride fuel and fluoride pyro-chemical partitioning fuel cycle technology. After 2005, the original R&D intentions were gradually converted to classical molten salt reactor (MSR) technology and to the thorium–uranium fuel cycle. Arguments for this decision were presented by a group of prominent Czech nuclear scientists. These arguments were based on the fact that the Czech Republic should support the development of a technology that can minimize the environmental impact of nuclear power, save natural resources, and has some potential to be deployed in future in non-superpower countries, unlike fast reactors, which need a high percentage of fissile material (highly enriched uranium or a high content of plutonium), and will probably represent the future of nuclear power for superpowers.

Theoretical and experimental development of MSRs and liquid thorium fuel technology has been realized by a national consortium of institutions and companies originally led by the Nuclear Research Institute, Řež. Based on the favorable–evaluation of the R&D project proposal, the Ministry of Industry and Trade of the Czech Republic decided to support these activities financially.

Main Results of Existing R&D Projects

After the first R&D activities in 2000–2003, which were devoted mainly to subcritical molten salt systems for incineration of trans-uranium elements, since 2004, the main R&D effort has been focused on critical MSR systems and, since 2005, also thorium–uranium fuel cycle technology has been under intensive study.

The Ministry of Industry and Trade supported two important R&D projects de-voted to MSR systems and the thorium–uranium fuel cycle. The first one, which was opened in 2004, was entitled “nuclear system SPHINX with molten fluoride salt based liquid nuclear fuel”, the second

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one, opened in 2006, was "fluoride reprocessing of GEN IV reactor fuels". The investigations come out of the knowledge and experience obtained by the US Oak Ridge National Laboratory (ORNL) during the Molten Salt Reactor Experiment (MSRE) project in the 1960s [1] and partially also from the exchange of scientific information with the French EDF team working on the AMSTER project at the beginning of this century [2].

The SPHINX project was devoted to exploring a broad spectrum of MSR technology, covering theoretical and experimental activities in MSR physics, fuel salt and fuel cycle chemistry, molten salt thermo-hydraulics, structural material development, and testing of apparatuses for molten fluoride salt media. The project was carried out by a consortium of institutions and companies led by the Nuclear Research Institute, Řež, in cooperation with ŠKODA JS (ŠKODA–Nuclear Machinery), the Nuclear Physics Institute of the Academy of Sciences of the Czech Republic, the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague, and Energovyzkum Ltd. Brno, and later also with the Research Centre, Řež, and COMTES FHT. The main aims of the project were to contribute to the knowledge of MSR reactor physics, core design and safety, structural material development, MSR fuel cycle technology, and to experimentally verify selected important areas of MSR technology and to contribute to the solution of existing bottlenecks. The research work, realized under the SPHINX project, was divided into following work packages (WPs):

- WP1: MSR core and primary circuit;
- WP2: MSR fuel cycle technology;
- WP3: Experimental MSR core and its control system;
- WP4: Secondary circuit and its components;
- WP5: Structural materials for MSR technology;
- WP6: System study of MSR-SPHINX;
- WP7: Experimental program SR-0.

The second project "fluoride reprocessing" was devoted to the experimental development of two fluoride partitioning technologies, specifically to the fluoride volatility method and to electrochemical separation processes from fluoride molten salt media. As the fluoride volatility method can also be used for oxide fuels from fast reactors, the investigation of electrochemical separation processes has been exclusively devoted to the thorium–uranium fuel cycle of molten salt reactor systems. The work within this project covered the experimental verification of fresh liquid molten salt fuel processing—a technology for ThF_4 and UF_4 preparation from ThO_2 and UO_2, respectively, and final processing of MSR fuel salts LiF–BeF_2–ThF_4 and LiF–BeF_2–UF_4. Other objectives of the MSR fuel cycle investigation were the system studies focused on the material balance calculations and conceptual flowsheet design of MSR on-line reprocessing. Both one-fluid (single-fluid) and two-fluid (double-fluid) systems of MSR core design were investigated and for both design systems the conceptual flowsheets were designed. Finally, the project covered studies devoted to the non-proliferation and physical protection aspects of the Th–U MSR fuel cycle technology.

The emphasis on the theoretical and experimental development of Th–U MSR fuel cycle technology was based on the fact that, although MSR design and operation were already verified by ORNL in the 1960s, the on-line reprocessing was never fully realized and still represents a major stumbling block for the whole MSR technology, which it is necessary to solve before MSR deployment in the future is possible. The "fluoride reprocessing" project was solved by the Nuclear Research Institute, Řež.

The main achievements of both projects are as follows:

- Challenging reactor physics experiments with inserted molten salt zones were realized in the LR-0, LVR-15, and VR-1 reactors at the Nuclear Research Institute, Řež, the Research Centre, Řež, and the Czech Technical University.
- Computer codes for calculation of the neutronic characteristics of MSR systems and for calculation of the composition evolution during the burnout of liquid fuel were developed.
- In the fluorine chemistry laboratory of the Nuclear Research Institute, Řež, the handling of beryllium-containing molten salts was mastered, fresh thorium and uranium molten salt fuel processing for MSR systems was verified in semi-pilot conditions, and the basic studies of electrochemical separation of actinides (Th, U) from fission products were realized. These electrochemical studies were devoted to MSR on-line reprocessing development.
- MSR fuel cycle mass balance calculations and conceptual flowsheets of on-line reprocessing were designed.
- A special nickel alloy called MONICR, which is resistant to molten fluoride salt media, was developed by the companies, ŠKODA JS and COMTES FHT. Irradiation and corrosion tests of the MONICR alloy and metallographic studies were performed and experimental production of sheets, tubes, and rods was realized.
- Basic design and theoretical and experimental development of impellers and valves for molten fluoride salts and "salt/salt" and "salt/air" heat exchangers were realized by Energovýzkum Ltd.
- The Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague opened new facultative topics "liquid nuclear fuels" and "MSR system technology" for undergraduate and PhD students.

The LR-0 reactor, now operated by the Research Centre, Řež, has proved to be extremely suitable equipment for the measurement of molten salt neutronics. The measurements were realized in inserted molten salt zones in the central part of the reactor core. The standard VVER fuel served as the neutron driver (Figs. 1 and 2). Basic critical parameters (the critical height of the moderator, the moderator level coefficient) were determined for each arrangement according to approved methodology of initial critical experiments for the LR-0 reactor. These, along with a description of the amount and degree of enrichment of the fuel and the material chosen for the filling, represent the fundamental data needed to determine the effects of the filling on the physical properties of the core and serve as the input for calculations and benchmark comparisons. The initial critical experiments were followed by measurements of neutron flux and reaction speed to determine the characteristics of the neutron and photon field inside the arrangement. The increase in reactivity was determined by differences in the critical moderator level of the arrangements with and without the salt [3, 4].

Molten salt reactor concept is classified as a non-classical nuclear reactor type, which exhibits some very specific features as a result of the use of liquid fuel circulating in the MSR primary circuit. The other specific features of the reactor type

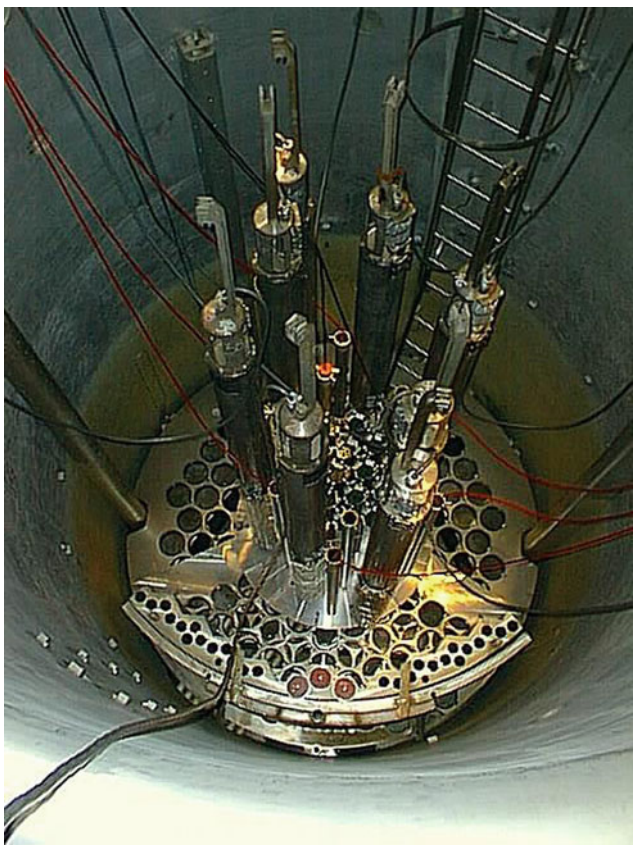

Fig. 1 View into the LR-0 reactor core with inserted salt zone

Fig. 2 Manipulation with the salt zone before insertion into reactor core

are based on the fact that the fuel circuit has to be directly connected with the on-line fuel reprocessing unit, a requirement that is necessary to keep the reactor in operation for a long time. MSRs can be effectively operated as thorium breeders within the ^{232}Th–^{233}U fuel cycle. The on-line reprocessing principle (fuel salt clean-up) allows not only continuous removal of fission products, but also a very effective extraction of freshly constituted fissile material (^{233}U). The fuel salt clean-up technology should be linked with the fresh MSR fuel processing to continuously refill the new fuel (thorium) into the reactor system. The simplified fuel cycle scheme of a thorium-fueled MSR is depicted in Fig. 3.

The theoretical as well as experimental investigation realized at the Nuclear Research Institute, Řež, has been focused mainly on electrochemical separation processes in fluoride media suitable for utilization within the MSR on-line reprocessing technology. The main objective of the experimental activities in the area of R&D on electrochemical separation technology has been to investigate the separation possibilities of the selected actinides (uranium, thorium) and fission products (lanthanides) in selected fluoride melt carriers. The cyclic voltammetry method was used to study the basic electrochemical properties.

The first step was the choice of fluoride melt suitable for electrochemical separations. The chosen melt should meet some basic characteristics: low melting point, high solubility of studied compounds, high electrochemical stability, and appropriate physical properties (electrical conductivity, viscosity, etc.). Unfortunately, no melt fulfilling all the requirements was found. Therefore, three candidate melts were selected for further electrochemical separation studies: an eutectic mixture of LiF–NaF–KF (acronym FLiNaK,

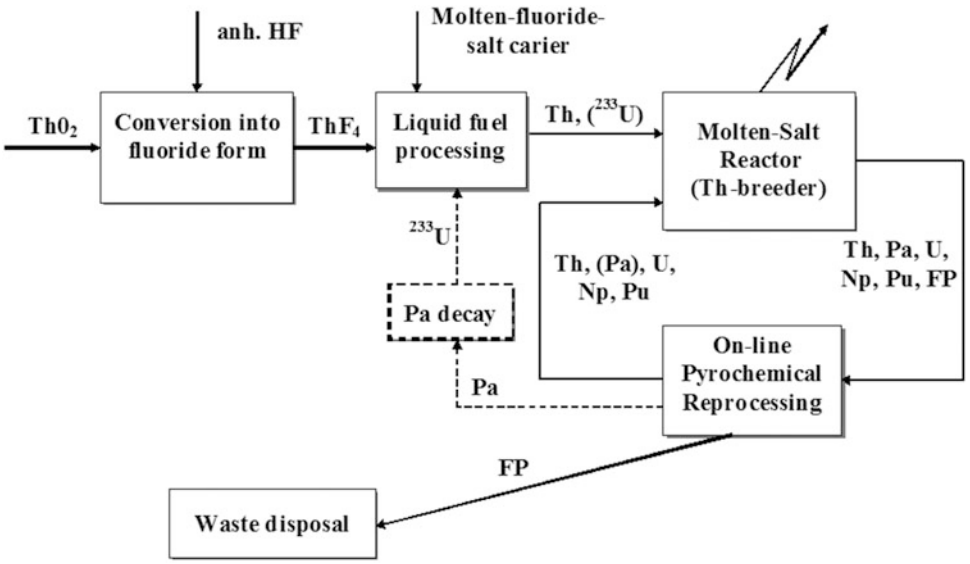

Fig. 3 Fuel cycle scheme for a MSR Th breeder. *FP* fission products; *anh* anhydrous

melting point 454 °C, limited electrochemical stability), a mixture of LiF–BeF_2 (acronym FLiBe, melting point. of chosen composition 456 °C, limited electrochemical stability), and an eutectic mixture of LiF–CaF_2 (melting point 766 °C, good electrochemical stability, but difficult to handle due to the high melting point). A special reference electrode based on the Ni/Ni^{2+} redox couple was developed to provide reproducible electrochemical measurements in the fluoride melts [5, 6].

Results obtained from the measurements can be interpreted in following way:

- In the FLiBe melt, there is a good possibility for electrochemical separation of uranium. Despite the fact that the electrochemical studies of protactinium have not been realized yet, based on the thermo-dynamical properties of PaF_4, there is a presumption that protactinium could be separated from this melt as well.

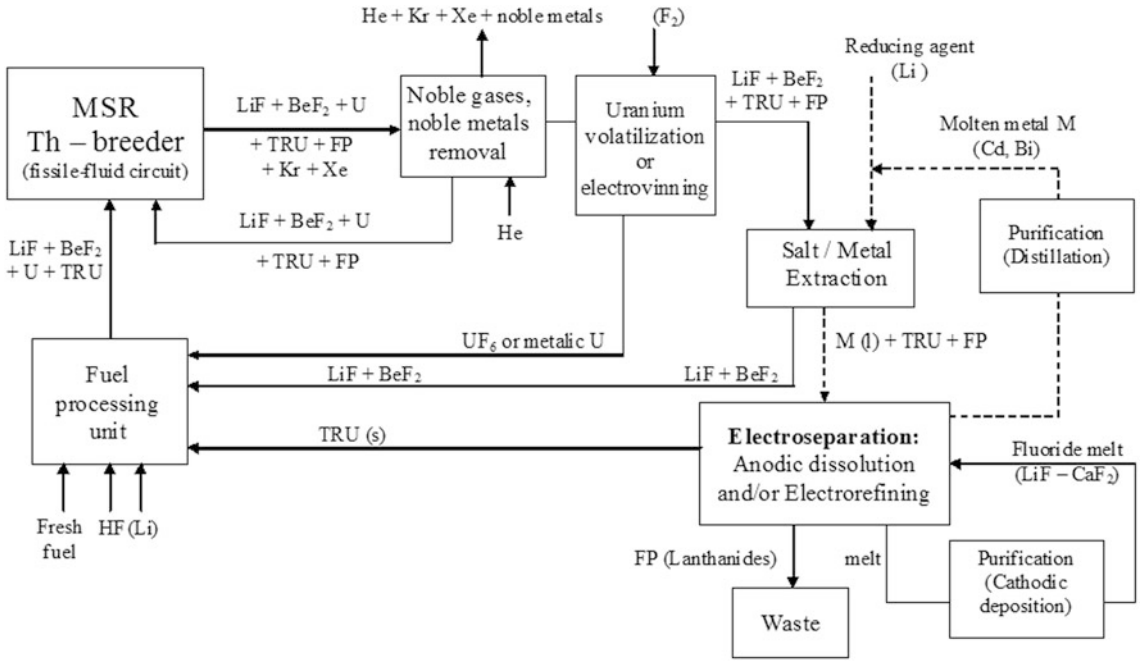

Fig. 4 Conceptual flowsheet for MSR Th breeder fissile-fluid circuit on-line reprocessing technology. *TRU* transuranium elements; *FP* fission products

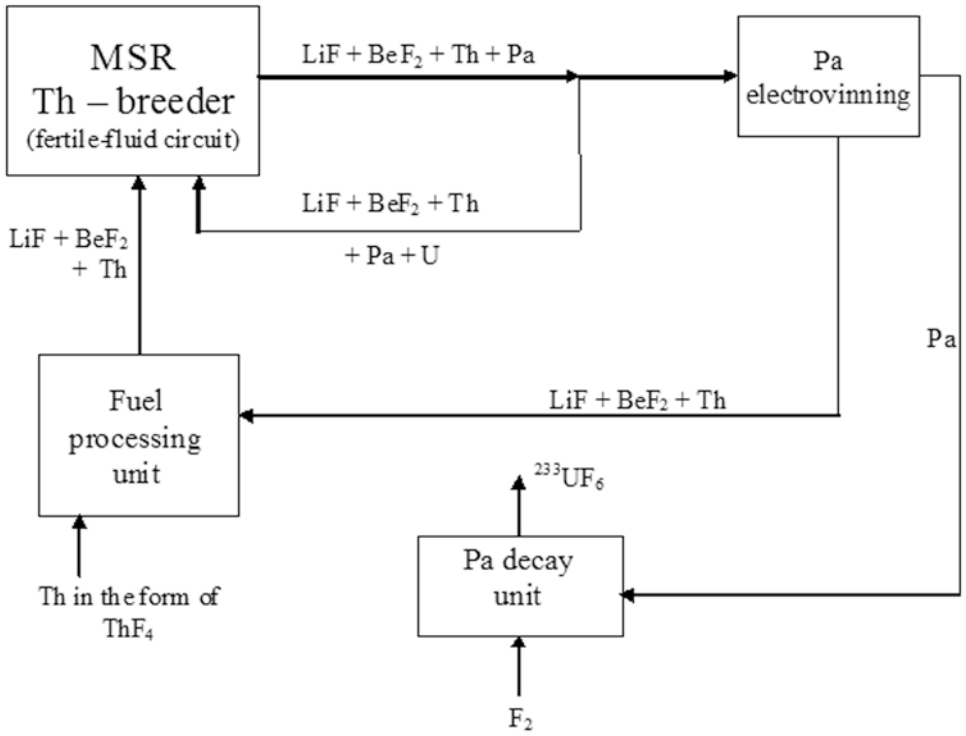

Fig. 5 Conceptual flowsheet for MSR Th breeder fertile-fluid circuit on-line reprocessing technology. *TRU* transuranium elements; *FP* fission products

- In FLiNaK and/or in LiF–CaF_2 melts, both uranium and thorium and most of the fission products (lanthanides) can be electrochemically separated.

Whereas the original MSR studies published by the Generation Four International Forum mainly considered the single-fluid MSR core design with the mixture of fissile and fertile material in the primary circuit, some older ORNL studies and the more recent development of MSR technology have been focused mainly on the double-fluid core design with separate channels for fissile and fertile material. Although the double-fluid core design is technically more complicated, it brings significant advantages: the ^{233}U breeding factor can be maximized [4] and the reprocessing technology is much more feasible than in the single-fluid core system. The requirements for the reprocessing of fissile-fluid and fertile-fluid circuits are different. As the speed of fissile-fluid reprocessing is determined mainly by the requirement to keep the concentration of fission products at the acceptable level, allowing the running of the reactor, the extraction of ^{233}Pa from the fertile-fluid circuit has to be as fast as possible because the speed of protactinium separation has direct impact on the breeding factor of the system [7]. The reason for the prompt separation of protactinium is to eliminate the nuclear reaction of ^{233}Pa(n, γ) → ^{234}Pa, which leads to production of undesirable ^{234}U. Based on the new electrochemical experimental data and the older experience from ORNL, the conceptual on-line reprocessing flowsheets for the fissile-fluid and fertile-fluid MSR circuits have been designed. These flowsheets are shown in Figs. 4 and 5, respectively.

Conclusions and Future Directions

Both projects described above were successfully finalized in 2009 (SPHINX) and 2012 (fluoride reprocessing) [8, 9]. The results and scientific knowledge achieved during these projects also enabled individual Czech research teams to participate in several international projects devoted to MSR technology (namely, the MOST, ALISIA, and EVOL projects of the 5th, 6th, and 7th Framework Programs of EC-EURATOM, some CRP projects of IAEA, and studies by OECD-NEA) and enabled the active participation of Czech representatives in the work of the Provisional System Steering Committee of MSR Systems of the Generation Four International Forum as a members of the EURATOM team.

The future plans for the further development of MSR technology in the Czech Republic are connected to cooperation with the United States. In 2012, the US Department of Energy and the Ministry of Industry and Trade of the Czech Republic concluded a Memorandum of Understanding focused on the support of collaborative R&D of fluoride-salt-cooled, high-temperature reactors (FHR) and molten salt reactor technology. Based on this Memorandum, in 2013, the US Department of Energy shipped a container with 75 kg FLiBe coolant (with isotopically pure ^{7}Li) to the Czech Republic for common neutronic experiments intended to be realized in LR-0 reactor in Řež. A new collaborative R&D program of FHR and MSR technology development is now under final preparation.

Although the Czech Republic is a relatively small country, which is not able to finalize the development of MSR systems alone, its existing and planned activities in MSR technology create a precondition to be an important partner for final collaborative development of MSR systems.

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ThEC13 Welcome Talk

Rolf Heuer

Abstract

CERN is very much concerned about energy and energy efficiency.

Thank you very much, Egil. This is something of a late welcome to this conference, I realize. I know you have been here since yesterday morning, but I have just got back from Spain where, as you might have heard, François Englert, Peter Higgs, and CERN were presented with the Prince of Asturias award for science and technology. This is the first time that such a high level prize has been given to theory and experiment together, an accurate reflection of how modern physics works, and perhaps a model for other award givers.

It's a pleasure to welcome you here to this wonderful landmark, the Globe of Science and Innovation, which, if I am not mistaken, is here due to the initiative of Monsieur Couchepin, whom I also want to warmly welcome to this place, to your conference home so to speak. It is a fantastic landmark and we use it very, very often. On the ground floor, there is a permanent exhibition on particle physics, and up here we host seminars and colloquia, as well as events such as this conference.

Turning to the conference, I hear that it has already started very successfully. First of all, there are people from many countries here: it is the first time that so many experts from all over the world have come together to discuss new directions towards thorium-based energy. In particular, I hear that this morning there was a special announcement [1]. This really is a conference that not only takes stock of what is going on, but also looks forward to what can be done, on a timescale that may be long. But we particle physicists are used to long timescales and we have learned to adapt accordingly.

Why did CERN agree to help organize this conference? First of all, we are very much concerned about energy and energy efficiency, subjects that are important for us. A laboratory like CERN can also help a lot in exploring the basic physics involved in these questions. And here I would like to mention, coming back to that long timescale, that in the mid-1990s Carlo Rubbia started his pioneering work on the transmutation of nuclear waste. That's already about two decades ago. Things are slowly developing, but I think that a conference like ThEC13 will very much help to accelerate it, because when people get together with a shared objective and general consensus on how to go forward, that makes an impression on others, and things do move ahead. Accelerator-driven systems make a good example: a field in which I hope we can help, working together with other countries and other regions, India and China, for example. There is interest on our side, and I am happy we have representatives of those countries here.

I am also happy to see in the program, that at the end of the conference, for those of you who are still alert after all your discussions, there is the opportunity to visit our facilities. I hope many of you will take the opportunity, because going down to the experiments is always spectacular, and at the moment the experiments are open, so you can see a great deal. It is very humbling to the see the places where 3000 scientists work together in order to study the smallest particles.

CERN is a unique place, with a remarkable atmosphere, which serves as a model of how people can work together when they share a common goal. This is important, not only for CERN, but also for the thorium energy community. Forget about individual ways of doing things. Unite behind a common goal, and then you can really achieve something. And here, I hope that the CERN model will inspire you, by seeing how we are doing projects, and what can be achieved

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when individual differences are put aside. After all, next year CERN turns 60 and we remain attractive. I wish you a good continuation of this workshop, and many fruitful discussions. And when the conference is over, I hope you will tell me where the future lies.

Thank you very much.

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Thorium Nuclear Power and Non-proliferation

Hans Blix

This conference on the use of thorium in nuclear power generation is most welcome. The aim is not to downgrade uranium, which has served the world well and long dominated our thinking. It has been the feed of the nuclear work horses for the last 50 years. Enormous experience has been accumulated, and it has helped us to refine the technology and also to gradually develop a nuclear safety culture.

Even though designers and operators are by no means at the end of the uranium road, it is desirable today that they use their skill and imagination to explore and test other avenues as well.

The propeller plane that served us long and still serves well gave way to the jet plane that now dominates. Diesel engines have migrated from their traditional home in trucks to a growing number of cars and cars with electric engines are now entering the market. Nuclear power should also not be stuck in one box. I have no doubt that the current uranium-fueled reactors will get brothers and sisters.

Although we live in an era when fear of radiation is sufficiently widespread to lead, after the accident in Fukushima, to snap decisions snuffing out nuclear power in Germany, there is still room for long-term thinking and financing of innovation in the nuclear sector. The international fusion project in Cadarache (ITER) is still being pursued despite its long-term perspective and high costs. Breeder reactors continue to be built in a few countries—such as Russia and India—and practical experience is being gathered that can help us to extract dramatically more energy from uranium fuel.

In my view, the lively research and development work that is being pursued in a number of countries and that aims at Generation IV reactors, shows that there remains much vitality, curiosity, and expectation in the nuclear sphere. The work on thorium is part of this dynamism and it is attracting increasing attention around the world. Rightly so, as the thorium line has some particularly attractive features. The participants in this conference are very familiar with them. I only briefly note that:

- global thorium resources are assessed to be three to four times greater than those of uranium. We do not, of course, accept the populist cry that only renewable sources of energy are viable. By such logic we should not rely on iron, copper, tin, etc. Nevertheless, it is a merit that a valuable resource is plentiful and can be relied on for a very long time;
- thorium fuel gives rise to waste that is smaller in volume, less toxic, and much less long-lived than the wastes resulting from uranium fuel.

Seen from the viewpoints of the environment and global security, two other features should be highly appealing. The first is that the use of thorium fuel can offer us a way to burn up large stocks of plutonium that now sit idle but require expensive care and protection. The other is that thorium fuel does not give rise to material that can be used for bombs. How important are these features?

The risk of proliferation of "weapons of mass destruction"—WMD—is declared by some to be the greatest threat to security in today's world. The question deserves serious and sober consideration. Let me first explain what is generally meant by the term WMD and begin with some readings in the lighter vein.

Shortly after the invasion of Iraq in 2003, US groups of experts arrived to eradicate the WMD that 700 UN inspections had failed to locate. When the weapons continued to be elusive, it became common to talk about them as "weapons of mass disappearance".

Personally, I encountered another reading of WMD. After my return to Sweden one day in 2003, I received a mail from a lady who asked me if I had any objection if she gave her CAT my name—Blix. I replied that my wife and I loved cats. We would be honored, but we wanted to know whether the cat

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accepted the name. The answer came that the cat seemed very pleased and it worked beautifully as a WMD—a "weapon of mice destruction". Actually, the cat came close to the essential common feature of WMDs—that of causing terror.

Unlike mice, men are not terrorized by cats but we are urged by governments and the media and led by our own feelings to focus on three categories of terror weapons that we term WMDs: Nuclear, Chemical, and Biological.

There is general support for the eradication of these three types of weapons, but the world community has so far succeeded only in concluding conventions under which vast number of states—including the great military powers—have legally committed themselves not to use, nor to possess chemical and biological weapons. For nuclear weapons, there is the non-proliferation treaty (NPT). It aims to prevent the further spread of these weapons and to achieve nuclear disarmament—but does not prohibit the states that still have nuclear weapons to use them.

When we discuss these weapons that are expressly branded as WMD, we should not forget that there are many other weapons that are particularly odious or likely to be indiscriminate, for example, incendiary weapons, cluster bombs, and antipersonnel mines. We should also be mindful that the real mass killers of people today are small caliber weapons. Despite the efforts made in most states to largely disarm the civilian population, many people are killed by small arms. In areas of conflict or civil strife the numbers of deaths from these weapons become horrendous. It is only recently that a treaty—the Arms Trade Treaty—has been concluded under UN auspices with the modest aim of at least preventing uncontrolled international trade in some conventional weapons.

Among the three categories of weapons that we do term WMD, biological and chemical, being of less strategic importance, have received less attention than nuclear. Although concerns have been voiced that some deranged scientist could use a high-tech lab to develop a devastating biological weapon, we have only once experienced such a character in operation. In 2002, anthrax in powder form was sent by letter and killed a number of persons in the US.

Saddam Hussein did have a biological weapons program, including anthrax—but never used it.

Chemical weapons—notably chlorine and mustard gas—by contrast, were used extensively with horrible effects in the First World War. Although they were prohibited for use in war already in the Geneva Protocol of 1925, we must shamefully note that there was not much international reaction when Saddam Hussein used sarin in the war against Iran in the 1980s and against his own citizens at Hallabja.

The reaction to the recent use of sarin in Syria was different. After nearly triggering a US punitive military strike, it led to a US–Russian agreement that prompted the Syrian government to accede to the Chemical Weapons Convention and now to cooperate in verified elimination of Syria's chemical weapons.

The Syrian drama has much to tell us. First, that although the number of people killed by the chemical weapons in Syria was very small compared with the number killed by other weapons, the global reaction to this use was the stronger by far. Second, while the Geneva Protocol of 1925 and the Chemical Weapons Convention of 1993 have regard to international armed conflicts, governments around whole world took the view that the use of gas was prohibited under any circumstance—including in a civil war. What we could see 25 years after the use of gas in the Iran–Iraq war was that chemical weapons have become subject to a taboo.

Since the fateful time when nuclear weapons were dropped on Hiroshima and Nagasaki, their numbers and explosive power have grown exponentially. During the peak of the Cold War there were more than 50,000 nuclear weapons in the world—most of them in the US and the Soviet Union. We learnt at that time that a nuclear war could have resulted in a nuclear winter causing the end of human civilization. Einstein famously said that he did not know how the third world war would be fought, but the fourth, he predicted, would be fought by stones and sticks.

Gradually, I believe, a taboo has developed also against the use of nuclear weapons. But is it unbreakable? Nuclear weapon states assure the world that the purpose of the weapons is to deter attacks and the International Court of Justice has declared the use of nuclear weapons illegal in all but the very limited case of securing a state's survival. Even so, it has not proved possible to persuade the nuclear weapon states to commit themselves by treaty to non-first use. They condemn the Syrian use of chemical weapons as illegal, but they do not accept to make a first use of nuclear weapons illegal.

Unlike the Biological Weapons Convention (BWC) and the Chemical Weapons Convention (CWC), the non-proliferation treaty—NPT—that entered into force in 1970, did not prohibit the use of the weapon. The treaty may be said, rather, to have aimed at preventing use of nuclear weapons by eradicating them. It reflected the ambition that:

- all the then non-nuclear weapon states (NNWS) should become parties, should stay away from nuclear weapons, and should accept safeguard verification of all their nuclear programs, and that;
- all the then nuclear weapon capable states—the P5—should also become parties and through disarmament do away with their nuclear weapons and facilitate NNWS parties' access to their peaceful nuclear technology.

How Did it Work Out?

- Almost all states that were without nuclear weapons in 1970 did adhere.
- Three states—Israel, India, and Pakistan—did not, and they have all developed nuclear weapons.
- Two of the states that adhered—Iraq and Libya—tried to breach the treaty and move secretly to nuclear weapons, but were stopped.
- One state—North Korea—withdrew from the treaty and has developed nuclear weapons.
- One state—Iran—is suspected by many to be moving toward a weapon, but it denies any such intention and there are now talks that may or may not lead to agreement.

So, all in all over the near 45 years of the treaty there has been an addition of only four new nuclear weapon states in the world. And, we may note that during the same period, four states have walked back from a nuclear weapon status: Ukraine, Byelorussia, Kazakhstan, and South Africa.

This is far better than President Kennedy's fears in the 1970s that there could be 15 or 20 nuclear weapon states by 1975.

Even with the relative success that we can note, any further spread of nuclear weapons would raise new risks—risks that some even hold are the gravest to world security. We are warned that the NPT could unravel if Iran and North Korea are not brought into line. Ideas are explored about how to raise obstacles to any withdrawal from the NPT and how to prevent or at least discourage states from developing their own enrichment capacity—as Iran has done—and as Japan, Brazil, Argentina, and South Africa did long before Iran.

I fully agree with the ambition to avoid the spread of nuclear weapons to further countries or—indeed—to terrorist groups. Retention of a nuclear weapon capacity in North Korea and development of one in Iran could lead to dramatic risks in the regions. Having said that about these two cases, I think some of the general concerns we hear are exaggerated and perhaps consciously or unconsciously advanced to draw attention away from the dismaying reality that the nuclear weapon states are much more interested in keeping other states away from nuclear weapons than in doing away with their own nuclear weapons. It should not have to be that way.

With the end of the Cold War, the severe military tensions in the world drastically subsided. All the landmasses of the southern hemisphere are within zonal agreements that exclude nuclear weapons. Many states in the northern hemisphere feel adequately protected by the nuclear umbrellas held over them or by membership in alliances or both.

Countries like my own, Sweden, or Switzerland or Austria that fall into neither category show not the slightest tendency to move to nuclear weapons. We should be aware that it is not the NPT per se that keeps states away from nuclear weapons. For various reasons—and I have mentioned some of them—the vast majority of states in the world do not feel a need to have the weapons. Some may reject the weapons as abhorrent and as an expensive burden that might be more dangerous to have than not to have. Adherence to the NPT registers this rejection and adds what we might term a legal threshold that gives a measure of stability.

Rather than fixing our eyes almost exclusively at the large number of states that do not have nuclear weapons as potential dangers, perhaps we should focus on the nuclear weapon states that still possess nearly 20,000 nuclear weapons and that, even 25 years after the end of the Cold War, are not reducing their arsenals—except for reasons of costly redundancy.

When we also see that the military expenditures of the world remain at some 1600 billion dollars a year—as high as during the Cold War—we may wonder if the security establishments have not noticed that the Cold War is over.

It took a famous US quartet of elder statesmen to put the reduction of the existing stock of nuclear weapons on the world's agenda. In an article in the Wall Street Journal in 2007, George Shultz, Henry Kissinger, Bill Perry, and Sam Nunn urged that the US and Russia, with the largest nuclear arsenals, should initiate nuclear disarmament. The Cold War was over long ago and if the behavior of these countries continued to suggest that they considered nuclear weapons indispensable, it would be difficult, they argued, to avoid that other states emulate that view. They were given much support at all levels in the US and in the world and if their advice had been followed, the US, Russia, China, France, and the UK would now be on a somewhat higher moral ground when dealing with North Korea and Iran.

Regrettably, the drive for nuclear disarmament that President Obama initiated in 2009 and that had a positive echo from President Medvedev, ran into a wall, when the chief result of the drive, the START was submitted for the consent of the US Senate and obtained this consent only with the greatest difficulty. Since then, there has been progress regarding the security of nuclear material and equipment, which is welcome to prevent any attempts by terror groups or others to make 'dirty' bombs that do not produce nuclear explosions but spread radioactivity and terror.

For the rest there is stagnation. The Geneva Conference on Disarmament is going into its second decade of coma, the comprehensive nuclear test ban treaty that was adopted in 1996 is not ratified by the US, China, and others and not even tactical nuclear weapons have been removed by NATO

and Russia from the European sphere, although they appear to have no military significance.

Are there no rays of hope? Yes. The framework agreement that Foreign Ministers Kerry and Lavrov recently attained in Geneva was not only highly constructive in itself but could also inject some hope that the US and Russia will seek to partner further to end the civil war in Syria. Let us hope it was also a mini reset to get them onto a road that will start significant nuclear weapons reductions. Perhaps the ministers of finance around the world should unite and tell their governments that there are better ways of using 1600 billion dollars a year than buying and maintaining hardware that is obsolete in 20 years. For instance, to counter budget deficits and to defend the planet.

I conclude: the civilian nuclear community must do what it can to help reduce the risk that more nuclear weapons will be made from uranium or plutonium. Although it is enrichment plants and plutonium-producing installations rather than power reactors that are the key concerns, this community can and should use its considerable brain power to design reactors that can be easily safeguarded and find a fuel and fuel supply organizations that do not lend themselves to proliferation. I think in these regards the thorium community may have important contributions to make.

The Road to Enablement for a Liquid-Fuel Reactor Fuelled by Thorium

John Durham

Abstract

There is growing recognition that thorium-fuelled Molten Salt Reactors (MSRs) will be one of the key clean energy technologies of the 21st Century. However, thus far few people have addressed the question of how to expedite MSRs' development and deployment. We must avoid a situation where MSRs are "always ten years away". This presentation will suggest several solutions to this quandary. The first part will address the economic and political hurdles that stand in MSRs' way: in the context of historically low levels of fission R&D, how can we leverage private investment in MSRs, and engender public support for MSR R&D?

The second part of the presentation will address the issue of global scientific cooperation: what tools do MSR researchers need to accelerate their research, and how can we, as MSR supporters, lead the development of these tools? For example, should we aim for a global database of MSR safety codes and models? Should scientists call on the OECD Nuclear Energy Agency and IAEA to fund a facility dedicated to MSRs research?

The third part of the presentation will argue that MSR designers must pay close attention to regulatory requirements from the outset of their research. If a design is too complex, then the chance of regulatory approval, and therefore investment, is slim. In order to satisfy both regulators and potential investors, future MSRs must not look to directly imitate the Oak Ridge design, but rather to simplify the reactor system. For example, is it possible to "design out" the more troublesome components of MSRs such as pumps and valves? Could we design an MSR that uses only previously certified materials?

In light of the points above, the final part will posit a draft road-map of MSR development, leading from simple once-through burner to fully fledged fast-spectrum reactor. We will conclude by arguing that the road to MSR enablement requires MSR designers to "Economise, Collaborate, and Simplify".

No contribution submitted by the author.

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Part IV

Invited Speakers: Thorium-Fuel Cycle and Transmutation

Overview of the Thor Energy Thorium Fuel Development Program

Saleem S. Drera, Julian F. Kelly, Øystein Asphjell, and Klara Insulander Björk

Introduction

THOR Energy, a Norwegian nuclear fuel technology company, has developed and is currently testing two new thorium-based fuels for use in existing nuclear power plants—the ThADD and the ThMOX fuels. In the effort to license these fuels, the company has initiated an internationally supported fuel campaign, which includes a 5 year irradiation trial at the Halden test reactor in Norway. The first fuel rods containing ThADD (thorium additive) and ThMOX (thorium mixed oxide fuel) fuel pellets were loaded into the reactor in April 2013. Detailed fuel irradiation data will continually be collected over the coming years and utilized in the many aspects required for fuel qualification.

THOR is seeking to have regulatory approval for commercial use of the ThADD fuel by the end of 2017 and to offer the fuel for commercial production in 2018. It intends to license the fuel designs to one or more major fuel manufacturers. Fuel designs are compliant with current industry standards and techniques such that manufacturers will not need to invest in further manufacturing capability.

Company Background

Dr. Alf Bjørseth established THOR Energy AS as a subsidiary of his company, Scatec AS in 2006. THOR Energy is a fuel technology company and THOR's aim from the outset has been the development and commercialization of thorium-based fuels.

A comprehensive feasibility study was initiated in 2006 in partnership with the Swedish utility Vattenfall AB. From 2008 onwards, research and development led to the start of irradiation trials at the IFE Research Reactor in Halden, Norway. The first test fuel rods, containing different thorium fuel types, were loaded in April 2013 and the trial program is scheduled to run until mid-2018. Over the course of the 5-year test period THOR Energy will study the fuel's operational performance and gather data to secure the necessary regulatory approval and licensing for use in commercial reactors.

Program Financing

Approximately half of the program costs will be financed via grants from the Norwegian Research Council and contributions from consortium members providing a significant leverage on equity capital from investors.

THOR Energy's major shareholder is Scatec AS, a privately owned Norwegian technology company, which specializes in developing new technologies related to the energy sector. Scatec currently owns 57 % of Thor Corporation (a holding company where Itochu Corporation is amongst the shareholders), which in turn owns 77 % of THOR Energy. THOR's other shareholders are: STL (14 %), Thorium Foundation (5 %), and Statoil Ventures (3 %). STL (Steenkampskraal Thorium Limited) is a South African nuclear technology company that has close links with THOR.

THOR has established an international consortium in order to finance and conduct the fuel irradiation trials in the OECD Halden Reactor. Consortium members have signed a 3-year contract and actively support the development of the fuel technology. The Consortium has received support from the Norwegian and the UK governments as well as from the Consortium members, which include: KAERI (Korean Atomic Energy Research Institute), Fortum OY (the Finnish nuclear utility), Westinghouse Nuclear Fuels Ltd.

Øystein Asphjell presented this paper at the Thorium Energy Conference 2013 (ThEC13)

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(US/SWE), National Nuclear Laboratories (UK), EU Institute of Transuranics (DE), and the Institute of Energy Technology (N).

The Seven-Thirty Program

The primary objective of the thorium fuel manufacture and irradiation program is to generate information that enables the licensing of thorium-based fuels that can be used in commercially operating light water reactors (LWR). To achieve this, data needs to be collected on how the candidate thorium-based fuels change as they operate for long periods under typical operation conditions. Fuel irradiation behavior information is drawn from direct on-line measurements, including fuel temperature, rod pressure, and cladding elongation, as well as post-irradiation examination, which yields data on chemical and mechanical interactions within the fuel rod. As the pellets are isolated within the fuel rod, the only effect of the heavy water moderator in the Halden Boiling Water Reactor (HBWR) is its effects on the fast flux, which can be taken into account by physics codes. For these reasons, the Halden Research Reactor is ideal for studying on-line pellet behavior and has provided a rich source of data, which has been long used to support fuel licensing efforts.

The data gathered during this program will serve the safety licensing case for subsequent lead test thorium-based fuel rods (LTR) and/or lead test assemblies (LTA) in a commercial reactor. Furthermore, the understanding of thorium oxide fuel performance gained from this test campaign will enable the development of a fuel performance code for predicting thorium fuel behavior during commercial operation—a vital part of the fuel developer's intellectual property. As an important benefit of thorium-based fuel is the potential for high burnup, an important objective of the irradiation campaign is to measure high burnup data.

Only data representative of the final commercial product can be used for fuel licensing. To be able to guarantee the representativeness of the collected data, the research program includes work on fuel manufacture procedures to ensure that the relevant thorium fuel ceramics are tested, prototypic of future commercial fuels.

ThADD (Thorium Additive) Fuel

The ThADD fuel offers utility operators a compelling advantage versus standard UOX (uranium oxide) fuel. By improving reactor core power flattening, the DNB (departure from nucleate boiling) fuel margins are improved, leading to the ability to increase power output by 2–3 %. Additionally, ThADD fuel reduces the amount of burnable poisons, such as gadolinium, used in reactor fuel by 33 %.

In terms of its manufacture, ThADD fuel does not require plant improvements or involve any major technical issues over standard UOX. It can be fabricated in conventional oxide pellet form by any of the fuel vendors and in any existing fuel manufacturing plant.

ThMOX (Thorium Mixed Oxide) Fuel

Beyond ThADD fuel, the other fuel research and development program for THOR is the thorium-based MOX fuel (ThMOX). As well as testing ThADD fuel pellets in the Halden reactor, THOR is also testing ThMOX pellets. Thor Energy's ThMOX fuel is a fundamental first step towards a thorium-based plutonium disposition option and possibly the first step towards a sustainable, long-term nuclear fuel cycle, which could be based on the use of thorium as a fertile component.

As a mixed oxide (MOX) fuel variant it is familiar to the nuclear industry, but ThMOX fuel offers certain key advantages compared with conventional UMOX fuel, which is being used in a several countries at present. Specifically, ThMOX fuel has greatly improved 'in-core' and 'spent-fuel' safety characteristics due to its inability to oxidize, leachability resistance and its lower thermal conductivity. Aside from safety characteristics, its key attribute is its ability to incinerate 'waste' plutonium. Unlike UMOX fuel, which only incinerates 20–30 % plutonium and produces a substantial volume of long-lived toxic waste, ThMOX fuel incinerates 60–70 % of the 'waste' plutonium and produces no new plutonium as it operates. Additionally, a higher amount of plutonium can be disposed of in a ThMOX fuel assembly as opposed to a UMOX fuel assembly while maintaining current fuel safety standards.

The potential for ThMOX fuel is substantial given that all current-generation light water reactors are capable of operating on (approx.) 30 % ThMOX fuel loads without any major design changes or modifications to the existing plant. In addition, all new light water reactors will be able to operate full cores (i.e., 100 %) of ThMOX fuel in the future.

Using ThMOX fuel in existing light water reactors does not require any technological or hardware design changes for reactors already licensed to burn standard MOX fuel. Furthermore, ThMOX fuel pellets can be manufactured in the same fuel plants currently producing MOX fuel due to the production similarities.

The Halden Verification Program

The instrumentation used for on-line measurements in the IFA-730 rig is listed below:

- Fuel thermocouples: The centerline temperature of the fuel pellets is measured by means of a thermocouple inserted into a narrow hole carefully drilled through a few pellets at the top or bottom of the fuel stack as shown in Fig. 1.
- Pressure transducers: The internal gas pressure of a fuel rod is measured by means of a pressure bellows within the fuel rod that communicates with a sensitive transformer mounted on the rod exterior as shown in Fig. 2.
- Cladding extensometers: Elongation of the cladding is measured by means of a magnetic rod mounted on the cladding, extending through a magnetic coil picking up its longitudinal movements. In addition, the neutron flux is monitored by means of neutron detectors at several locations in the rig, the moderator temperature is monitored by thermocouples at the coolant inlet and outlet, and the flow is measured by an inlet coolant flowmeter as shown in Fig. 3.

The rig that is currently loaded in the Halden Research Reactor comprises six rods of 30 cm active length, arranged and instrumented as shown in Fig. 4.

Rods 1 and 3 contain the $Th_{0.92}Pu_{0.08}O_2$ pellets manufactured at ITU. The $Th_{0.92}Pu_{0.08}O_2$ pellets have a small diameter of only 5.9 mm. This choice of diameter was made during the OMICO (Oxide Fuels—Microstructure and Composition Variations) project for which the pellets were originally designed. Reducing the diameter and increasing the fissile plutonium loading allows for an acceleration of the irradiation, which was prioritized in that context.

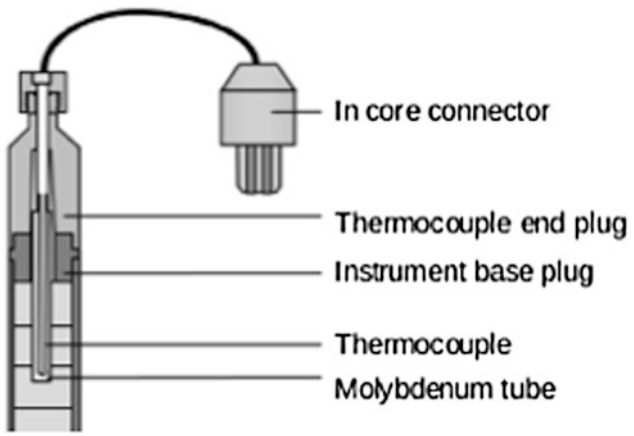

Fig. 1 Schematic drawing of a fuel rod instrumented with a thermocouple

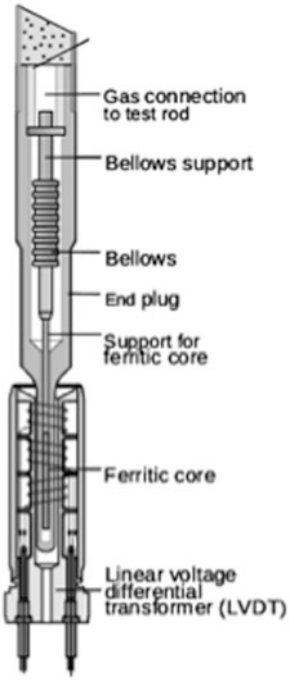

Fig. 2 Schematic drawing of a fuel rod instrument with a pressure bellows

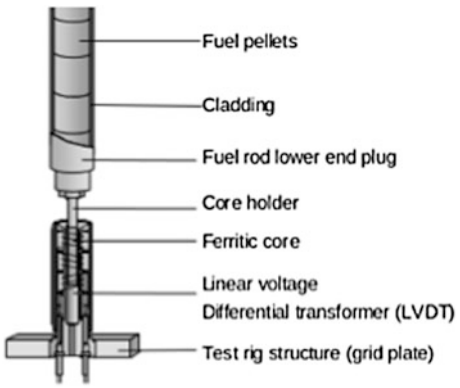

Fig. 3 Schematic drawing of a fuel rod instrumented with a cladding extensometer

The thermocouple extends into the $Th_{0.92}Pu_{0.08}O_2$ pellets and measures the operating centerline temperature of these pellets. In addition to the thermocouples, rod 1 is instrumented with a cladding extensometer and rod 3 with a pressure transducer.

Rod 5 is a reference rod for rods 1 and 3, containing only $Th_{0.42}U_{0.58}O_2$ pellets of the same diameter. This rod is also instrumented with a fuel thermocouple and a pressure transducer. This experiment setup does not allow for an accurate assessment of the fission gas release behavior of the $Th_{0.92}Pu_{0.08}O_2$ material, but a difference in rod pressure between rod 3 and its reference rod 5 would indicate that there may be a difference between the $Th_{0.42}U_{0.58}O_2$ and $Th_{0.92}Pu_{0.08}O_2$ material in this respect. Rods 2 and 4 contain

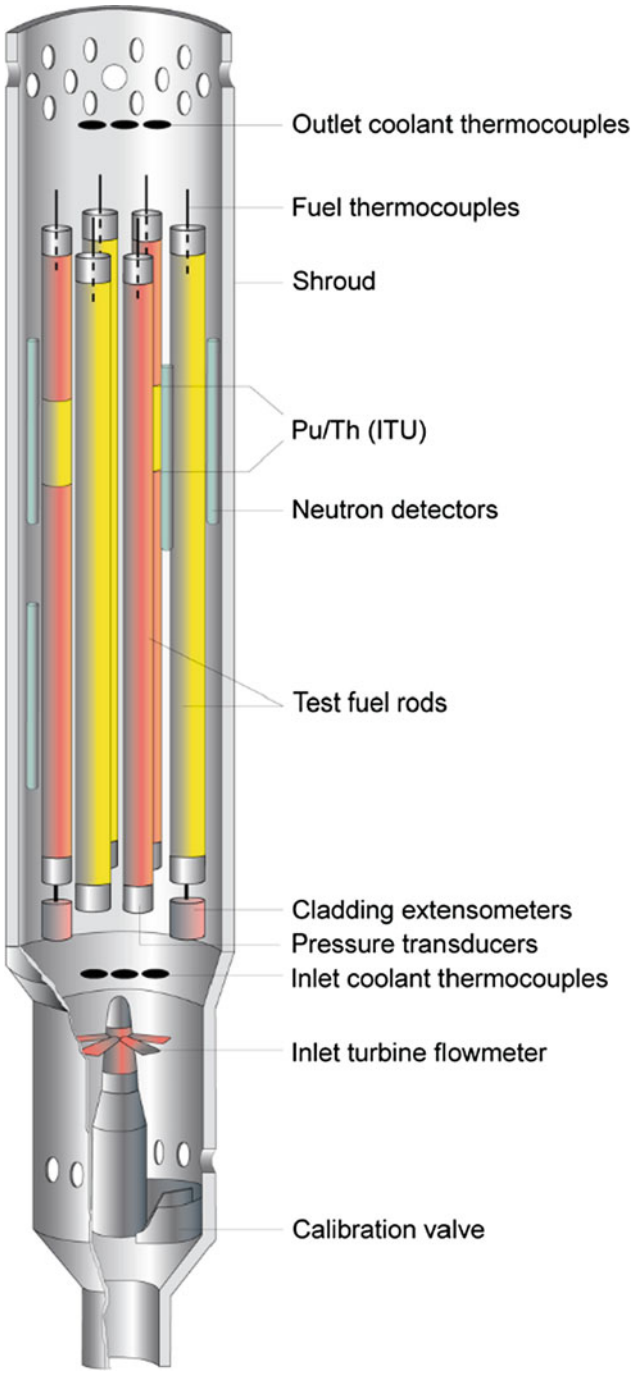

Fig. 4 IFA-730 irradiation rig

$Th_{0.07}U_{0.93}O_2$ pellets with a diameter of 8.48 mm. This is a standard boiling water reactor (BWR) diameter and was chosen to yield data representative for commercial reactors. Choosing the same diameter as the other rods would have facilitated comparison of the fuel types, but the representative aspect was prioritized as applications of the different fuel types are quite different, so that a choice between them will not primarily be based on differences in fuel performance. Rods 2 and 4 are both instrumented with fuel thermocouples, rod 2 with a pressure transducer and rod 4 with a cladding extensometer. Rod 6 is a reference rod for rods 2 and 4 with 8.4 % enriched uranium (UO_2) pellets of the same diameter, and is instrumented with a fuel thermocouple and a pressure transducer.

All pellets are enclosed in standard Zircalloy-2 cladding tubes.

In the following phase of the experiment, three rods will be discharged from the current rig and replaced by two rods containing the freshly manufactured $Th_{0.86}Pu_{0.14}O_2$ pellets from the IFE Kjeller laboratory, and a uranium oxide reference rod for this material. These three rods will have the same diameter of 8.48 mm and be instrumented similarly to the current rods, that is, with fuel thermocouples on all rods, as well as a cladding extensometer on one of the $Th_{0.86}Pu_{0.14}O_2$ rods and pressure transducers on the remaining two.

Outlook

In the third phase of the Seven-Thirty program, a new test rig will be introduced for testing (Th, Pu)O_2 pellets manufactured at a new alpha laboratory, operated by the UK National Nuclear Laboratory (NNL). This rig will comprise a 12-rod double cluster consisting of two stacked six-rod clusters. The upper cluster will be located at the axial flux peak in the Halden Research Reactor, a location chosen to accumulate burnup as fast as possible. The lower cluster will experience a tapered flux profile, which is useful for acquiring samples of varying burnup for post-irradiation experiments (PIE). Irradiation of these two rigs will continue until late 2017 at which point the oldest material will have accumulated a burnup of approximately 50 MWd/kgHM.

Although on-line measurements are invaluable for understanding in-pile fuel behavior, post-irradiation examination is crucial for understanding the physical mechanisms behind the manifested macroscopic behavior. The characterization of microscopic phenomena such as grain growth, formation of high burnup structure, and diffusion of oxygen and fission products within the fuel ceramic is important for building reliable fuel behavior models. To assess these processes, a PIE program is proposed comprising rod puncture and gas spectrometry, non-destructive neutron radiography and gamma scanning of the fuel rod, optical microscopy and scanning electron microscopy (SEM), punch tests for micro-hardness measurements, and measurement of elemental composition by mass spectrometry.

The objective of the Seven-Thirty program is to provide the requisite data that can enable the licensing of thorium-based fuels for use in commercially operating light water reactors. After completion of this program, the target is to insert lead test rods (LTRs) and, thereafter, lead test assemblies (LTAs) into a commercial reactor. The fuel pellets

for these rods will be manufactured in accordance with the process developed within the Seven-Thirty program. For licensing purposes, a well benchmarked fuel performance code must also be in place. The development of such a code is in progress, and the collected data will be used for benchmarking. Full-core simulations such as the ones described and partly completed in Sci. Technol. Nucl. Installations, 2013, Article ID: 867561, must also confirm the feasibility with respect to neutronic behavior. With all these pieces in place, thorium-based fuels will be ready for large-scale deployment in currently operating light water reactors.

Acknowledgments The Seven Thirty thorium fuel irradiation consortium is grateful for support from the Norwegian Research Council through its program for user-driven research-based innovation (BIA). The authors would like to thank the many IFE employees who have been crucial in enabling this experiment, in particular Margaret McGrath. We also thank Joseph Somers at JRC-ITU for valued technical insight.

Utilization Potential of Thorium in Fusion–Fission (Hybrid) Reactors and Accelerator-Driven Systems

Sümer Şahin

Abstract

World thorium reserves are approximately three times more abundant than natural uranium reserves. Furthermore, nuclear power plants are producing nuclear waste materials in substantial quantities in the form of minor actinides (MA). Large quantities of reactor-grade (RG) plutonium have been accumulated in the course of nuclear electricity generation over the past 50 years in civilian reactors as nuclear waste. Two emerging nuclear energy system for the utilization of thorium and reactor-waste actinides have been investigated. (D, T) fusion reactions produce highly energetic neutrons at 14.1 MeV, which can easily fission thorium. Calculations on a laser-fusion-driven fusion–fission (hybrid) reactor has led to fissile burnups exceeding 400,000 MWd/MT. In addition to fission energy production in situ, such a reactor would produce $\sim$160 kg of ^{233}U per year. Highly energetic protons near the GeV range can destroy heavy nuclei so that each proton can initiate creation of multiple highly energetic neutrons, called evaporation neutrons or spallation neutrons. These secondary neutrons will themselves lead to the production of new neutrons in a cascade so that the proton energy will be multiplied through fission processes and, additionally, new fissile fuel will be bred from relatively passive nuclei, such as ^{232}Th and ^{238}U. Calculations have shown that the spallation neutron spectrum in infinite medium by incident 1 GeV protons can range from thermal up to 1 GeV. We note that the high energy tail plays an important role in neutron multiplication. The spallation neutron spectrum peaks at approximately 1 MeV. The maximum number of fission events per proton in ^{232}Th, ^{238}U, and natural uranium will be 2.754, 11.446, and 17.888, respectively. The corresponding combined ^{233}U and ^{239}Pu production will be 48.357, 69.013, and 78.045 atoms per incident proton.

Introduction

World thorium reserves are approximately three times more abundant than natural uranium reserves. Turkey, for example, is rich in thorium resources. Hence, thorium remains a potential energy source for future energy strategies in Turkey. At the same time, nuclear power plants are producing nuclear waste materials in substantial quantities in the form of minor actinides (MA). MA can be regarded as the most hazardous radioactive waste products because of their long-term, high-level radioactivity. A significant fraction of reactor-waste MA consists of diverse plutonium isotopes, which represent a source of serious public and political concern with respect to misuse of this plutonium and also accidental release of highly radiotoxic material into the environment.

The (D, T) fusion reaction produces highly energetic neutrons:

$$\mathrm{D} + \mathrm{T} \rightarrow \alpha(3.5\ \mathrm{MeV}) + \mathrm{n}(14.1\ \mathrm{MeV}) \qquad (1)$$

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Thus, fusion neutron energy is significantly higher than the threshold fission energy of ^{232}Th and ^{238}U. Hence, passive isotopes become fissile fuel through irradiation with 14 MeV fusion neutrons. This would increase the potential exploitation of nuclear fuel resources several hundred-fold. Two emerging nuclear energy systems have shown great potential for the efficient utilization of thorium and reactor-waste actinides, and which are the subject of investigation in this work:

1. Thorium and reactor-waste actinides, either in the form of mixed fuel or separately, in fusion–fission (hybrid) reactors.
2. Thorium and reactor-waste actinides, either in the form of mixed fuel or separately, in accelerator-driven systems (ADS).

Minor Actinide Burning in Fusion–Fission (Hybrid) Reactors

Energy multiplication in a fusion–fission (hybrid) reactor could lead to early market penetration of fusion energy for commercial utilization. Progress at the National Ignition Facility (NIF) at the Lawrence Livermore National Laboratory (LLNL) brings fusion into focus as a viable energy source in the foreseeable future. A hybrid reactor design concept, the so-called Laser Inertial Confinement Fusion Fission Energy (LIFE) engine, has emerged out of these considerations.

The neutron transport calculations have been performed by solving the Boltzmann transport equation with the SN transport code XSDRNPM in the S8-P3 approximation by using the 238-neutron groups' data library. The resonance self-shielding weighted cross sections have been processed for unresolved resonances and for resolved resonances. Hence, calculations are conducted with high precision.

In the investigated (LIFE) engine design, the first wall is made of oxide dispersion strengthened (ODS) steel (2 cm) and followed by a Li17Pb83 zone (2 cm), which acts as a neutron multiplier, tritium breeding, and front coolant zone. It is separated by an ODS layer (2 cm) from the FLIBE molten salt zone (50 cm), containing MA as fissionable fuel. A third ODS layer (2 cm) separates the molten salt zone on the right side from the graphite reflector (30 cm). For burnup and time calculations, a driver with constant fusion power of 500 MW_{th}, by a plant factor of 100 %, has been assumed. This gives a continuous neutron source strength of 1.774×10^{20} (14 MeV-n/s). The main coolant consists of FLIBE, a mixture of lithium fluoride (LiF) and beryllium fluoride (BeF_2). MA carbide fuel is placed in the center of TRISO particles, as described in a separate work in these proceedings [1]. The particles are homogeneously dispersed and suspended as microparticles in the FLIBE coolant. The choice of TRISO-type fuel in FLIBE coolant gives three very significant advantages:

1. Very high fuel burnup capability of the TRISO fuel had been successfully demonstrated with Peach Bottom experiments: TRISO fuel pellets charged with plutonium were irradiated in the Peach Bottom Reactor over 30 years ago and reached burnup values in the range of ~740,000 MWd/MT without damage to the coated fuel particles (Fig. 1; adapted from the General Atomics reports [2, 3]).

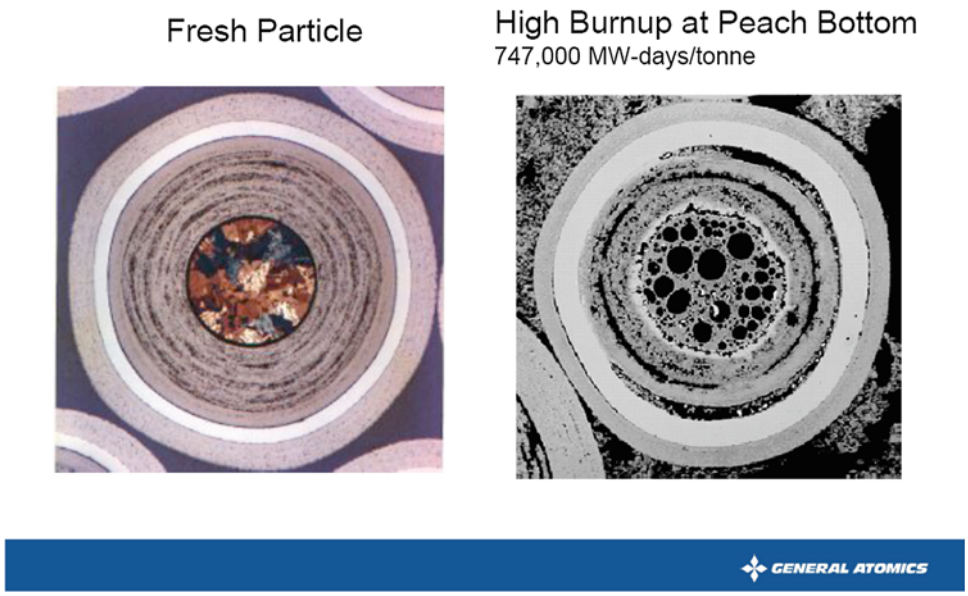

Fig. 1 Very high burnup in a ceramic-coated (TRISO) fuel, as experimentally demonstrated at Peach Bottom-1 Modular Helium Reactor (MHR). Adapted from [2, 3]

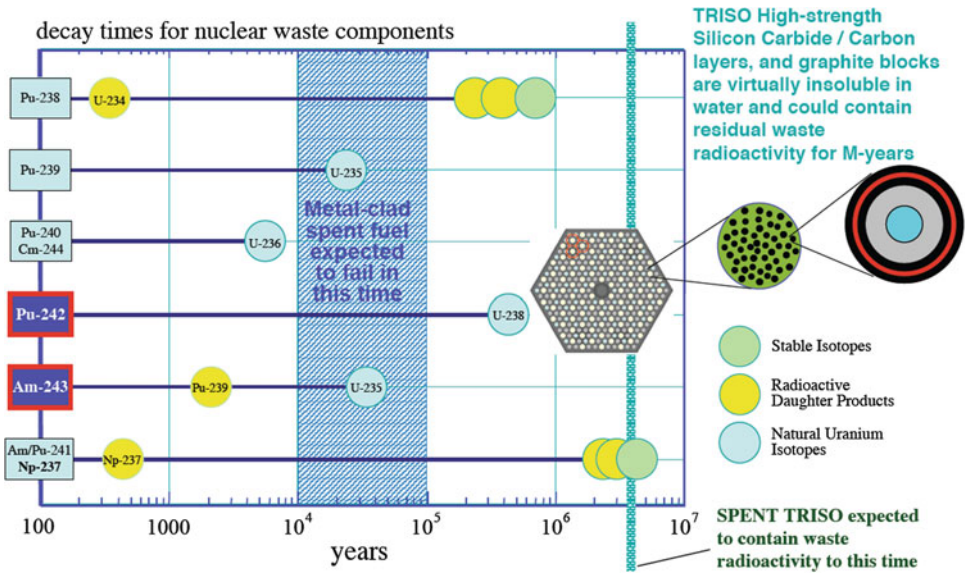

Fig. 2 TRISO coating provides structure stability and contains fission products

2. Safe accommodation of highly radioactive fission products and fissionable elements over millions of years (~4 million years) would be possible. TRISO fuel can contain the totality of actinide isotopes produced over long periods without being damaged nor destroyed (Fig. 2; adapted from [2]). Actinides will be transformed in the capsule to either stable isotopes or uranium. Metal clad contains fission products for $\sim 10^4$–10^5 years.
3. The main coolant, FLIBE, remains free of all sorts of fission products provided that the TRISO particles do not fail. As no mechanical force will be applied to the TRISO particles, it can be assumed that TRISO failure is not expected.

The volume fraction of TRISO particles in the FLIBE coolant has been varied as 0, 2, 3, 4, and 5 %. Highly energetic 14 MeV neutrons confer additional neutron production through multiple fast fission processes on all actinide isotopes. This leads to enhanced breeding characteristics and energy multiplication in the blanket. The corresponding tritium breeding ratio (TBR) values have been calculated as TBR = 1.134, 1.286, 1.387, 1.52, and 1.67, respectively. Blanket energy multiplication values become at startup M = 1.36, 3.3, 4.6, 6.15, and 8.1, respectively. Tritium breeding and energy multiplication will be gradually reduced by the continued recycling of the same fuel charge over 10 years. This can be circumvented by seeding fresh fissile fuel periodically.

Under irradiation with 14 MeV fusion neutrons, all actinides become highly fissile materials. For the selected TRISO volume fractions in the FLIBE coolant, fissile burnup exceeds 400,000 MWd/MT. Drastic reduction of final nuclear waste per unit energy production is expected. TRISO particles can withstand such high burnups without being damaged (Fig. 1).

Major damage mechanisms at the first ODS wall, which directly faces the fusion chamber, have been calculated as dpa = 50 (displacements per atom) and He = 176 appm per year. Under these conditions, the first ODS wall will need to be changed approximately every 3 years. Hydrogen production is calculated as 648 appm per year [4]. But, the latter will not reside permanently in metallic lattice as helium atoms. Detailed analysis has been given in [5].

Thorium in Fusion–Fission (Hybrid) Reactors

The use of thorium in connection with fusion–fission (hybrid) reactors has been considered since the 1980s [6–11]. In a recent work, a detailed analysis has been outlined for a laser-fusion reactor for thorium production [12]. Calculations have been conducted with the MCNP6.1/MCNP5/MCNPX code [13] for continuous energy. The main results can be summarized as follow.

The reactor consists of a spherical fusion chamber of 5 m diameter, which is surrounded by a multilayer spherical blanket. The first wall is made of S-304 steel (2 cm) and is followed by the natural lithium coolant zone that is 50 cm thick. A second S-304 steel zone (2 cm) separates the coolant–fission zone on the right-hand side from the graphite reflector (30 cm). ThO_2 is placed in the kernel of TRISO particles, which are homogeneously dispersed and suspended as microparticles in the natural lithium coolant with volume fractions of V_{tr} = 1, 2, 3, 4, 5, and 10 vol%, as described above. Calculations with ΔR_{Li} = 50 by variable V_{tr}, yield TBR = 1.229, 1.222, 1.214, 1.206, 1.1997, and 1.1622, respectively. Parasitic neutron absorption in thorium

Table 1 Integral data for an infinite medium

Fuel	^{235}U(n, f)/p	^{238}U(n, f)/p or ^{232}Th(n, f)/p	Total (n, f)/p	E_f/p[a] (GeV) or M	k_∞	^{238}U(n, γ)/p ^{239}Pu/p or ^{232}Th(n, γ) ^{233}U/p
Nat-U	4.7068	13.181	17.888	3.241	0.764	78.0450
^{238}U	–	11.4455	11.4455	2.075	0.675	69.0129
^{232}Th	–	2.75408	2.75408	0.473	0.321	48.3568

[a]E_f = 190 MeV/fission

leads to a decrease in the TBR values. For $V_{tr} < 5$ vol% of TRISO in the coolant, the increase in the neutron absorption by thorium will be compensated to a great degree by neutron multiplications via ^{232}Th(n, f) and ^{232}Th(n, 2n) reactions, so that the sacrifice of TBR remains acceptable. However, for $V_{tr} > 5$ vol% of TRISO, neutron absorption by thorium increases rapidly. Conversely, blanket energy multiplication, M, increases with more thorium, that is, M = 1.2206, 1.2322, 1.2426, 1.2536, 1.2636, and 1.3112, respectively, owing to the contribution of the fission energy. Annual fissile fuel production in the blanket are calculated as 17.23, 33.09, 48.66, 64.21, 79.77, and 159.71 kg/year of ^{233}U, respectively. The reactor remains deeply subcritical with fissile blanket criticality values of k_{eff} = 0.181, 0.188, 0.195, 0.202, 0.209, and 0.273, respectively. This gives a high degree of reactor safety.

Thorium in Accelerator-Driven Systems

One of the potential exploitation options of thorium are accelerator-driven systems (ADS). A high-intensity linear accelerator with proton beam energy of 1 GeV, directed on heavy nuclei can cause evaporation of a great number of highly energetic spallation neutrons. These can initiate a cascade of secondary multiple fission neutron multiplication, all of which would result with energy multiplication and fissile fuel breeding. In that respect, a great number of papers have published with the expectation of high grade energy multiplication in a subcritical blanket criticality close to unity. A recent example has been presented at the Thorium Energy Conference (ThEC13) at CERN, where a subcritical value as high as k_{eff} = 0.997 and a gain factor of G = 700 was cited [14]. The most advanced experimental ADS project, MYRRHA [15], considers k_{eff} = 0.97 as the highest permissible blanket criticality limit with respect to reactor safety [16]. It is true that a proton-driven subcritical ADS with $k_{eff} < 1$ would increase the neutron level, and consequently, the fission energy level proportional to the inverse of the times $1/(1 - k_{eff})$. However, one would need either enriched uranium or plutonium to have a subcritical blanket criticality close to unity! Availability of such high-quality nuclear fuel would make such complicated machines like fusion hybrids or an ADS hybrid obsolete, because the same fuel can be used much more simply in conventional nuclear reactors with well-known technologies.

In preceding works, different models in the MCNPX code for ADS calculations have been compared [17–19]. In this work, we investigate the possible upper infinite medium criticality limits, that is, the maximum k_∞ values, for potential breeder materials, namely, metallic thorium, natural uranium, and ^{238}U. Calculations have again been conducted with the MCNP6.1/MCNP5/MCNPX code [13] for continuous energy with the CEM2 k model. Table 1 shows the volume and energy integrated values for fission rate, fission energy release (E_f), k_∞, and infinite fission energy blanket energy multiplication, M, for these breeder materials. One can easily see that the blanket criticality would remain deeply subcritical and energy multiplication via fission remains very modest, even for hypothetical infinite blanket dimensions. On the other hand, fissile material breeding properties are high and needs to be exploited.

Conclusion and Recommendations

A series of calculations has been carried out to assess minor actinides transmutation from LWR spent fuel, as well as the thorium breeding capability of laser-fusion hybrid reactors. The main conclusions can be summarized as follows.

Utilization of a TRISO-type fuel suspended in molten salt coolant or metallic coolant brings new advantages:

- Coolant is not damaged by radiation and, therefore, could be recycled over a long period provided that no particle failure occurs during reactor operation;
- Separation of fission products and highly radioactive actinides from the coolant allows simplified handling of molten salt coolant;
- Very high burnup (>400,000 MWd/MT) becomes achievable with the same nuclear waste charge;
- Thorium in liquid lithium coolant produces high-quality nuclear fuel and multiplies fusion energy;
- Drastic reduction of final nuclear waste per unit energy production will be possible;
- Permanent immobilization of residual radioactivity in TRISO spent fuel after irradiation due to containment of

fission products in the TRISO particles over millions of years;

- Infinite medium criticality calculations for potential breeder materials (metallic thorium, natural uranium, and ^{238}U) have revealed deep subcriticality and modest fission energy multiplication in the blanket; whereas, high-quality nuclear fuel breeding properties are very promising. For blanket criticality close to unity and higher fission energy multiplication values, either enriched uranium or plutonium will be needed. However, high-quality nuclear fuel can be used in a simpler way in conventional nuclear reactors with well-known technology.

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A View on the Thorium Fuel Cycle

Daniel Mathers and Kevin Hesketh

Abstract

The thorium fuel cycle is analyzed in relation to the advantages demonstrated and expected, and of the known disadvantages. The need for R&D is described in order to get definite answers on the economics of the fuel cycle, the viability of Thorex reprocessing, and the proliferation issues. The time to build up a fleet of Th reactors is simulated for typical scenarios of installed capacity.

Background

The thorium fuel cycle is based on the transformation of naturally occurring Th-232, which is fertile, into fissile U-233 by neutron capture [see (1)]. Thorium is therefore not a nuclear fuel and the neutron capture needed to generate U-233 is an expensive initial investment. U-233 has a very favourable fission to capture ratio, superior to U-235, Pu-239 or Pu-241. In a thermal neutron spectrum, the fission to capture ratio is favourable enough to allow a breeding cycle, which is not possible with any other fissile nuclide. The Th-232/U-233 fuel cycle has been considered attractive, because it offered the prospect of a breeder or near breeder in a thermal reactor, with fewer technical obstacles than were posed by fast reactors. Early efforts to establish the thorium fuel cycle included a demonstration in the thermal breeder programme in the Shippingport PWR. This was designed to demonstrate a breeding ratio close to 1.0 and in this respect it was successful. But it is unlikely that this fuel cycle would be economic under current conditions. U-233 can also be fissioned in a fast neutron spectrum, in the same way as other fissile nuclides. However, it is disadvantaged compared with Pu-239 with a smaller number of neutrons per fission.

$$\text{Th-232}\ (\mathrm{n},\gamma) \rightarrow \text{Th-233}\ (\beta^-) \rightarrow \text{Pa-233}\ (\beta^-) \rightarrow \text{U-233} \tag{1}$$

The thorium fuel cycle can be deployed in a once-through fuel cycle or with recycle. The once-through fuel cycle is simpler technologically, but only offers very limited benefits in terms of uranium utilisation. Full recycle with Th-232/U-233 offers an unlimited resource, but also poses many technological challenges, especially reprocessing and fuel manufacture.

Pros and Cons

The Th-232/U-233 has often been claimed to have advantages over the U-Pu fuel cycle, some of which are justified and some are not:

- *Sustainability*: Th-232 is without doubt more abundant than uranium and if a market developed it would represent an enormous energy resource. The fact that no enrichment is required is also helpful, as it reduces the mining requirement by a factor of about 10 compared with current LWRs. If the Th-232/U-233 cycle could be

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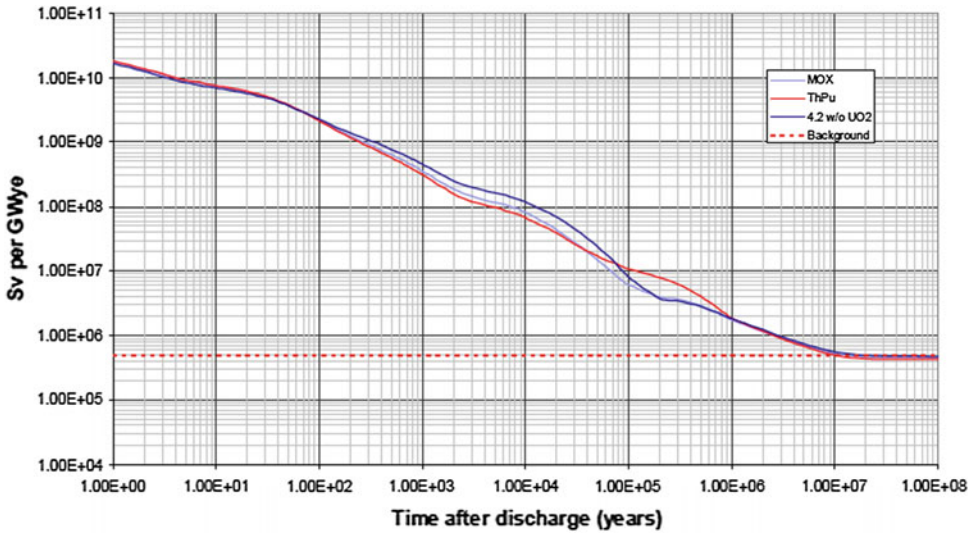

Fig. 1 Radiotoxicity over time for a range of fuel types

taken to equilibrium with recycle, it would represent an unlimited resource. Once-through thorium fuel cycles only give a marginal improvement in sustainability.

- *Neutrons per fission*: favourable in a thermal spectrum, but unfavourable in a fast spectrum as noted above.
- *Waste inventories*: The fission products produced from U-233 fissions are essentially the same as those from the U-Pu fuel cycle. The much reduced transuranic inventory is not necessarily advantageous in a repository, where transuranic transport to the environment is not the controlling factor for dose rates to the limiting groups. Considerable R&D would be needed to establish the waste forms that will arise from a full recycle scheme with Th-232/U-233.
- *Radiotoxicity*: Because the thorium fuel cycle starts at an atomic mass of 232, it takes many more neutron captures to generate transuranics such as Np, Pu, Am and Cm. Once fission products have decayed after about 500 years, the radiotoxicity of the U-Pu fuel cycle is dominated by transuranic elements and the Th-232/U-233 fuel cycle has a much lower radiotoxicity. However, in the very long term Th-232/U-233 systems always have a higher radiotoxicity than their U-Pu equivalents because of the build-up of daughter nuclides from the U-233 decay chain. The radiotoxicity of the thorium fuel cycle therefore depends on the timescale in question and is not always favourable. Figure 1 illustrates how radiotoxicity varies for selected U and U-Pu (MOX) fuels and Th-Pu fuel.
- *Fuel properties*: ThO has some advantages over UO_2 as a fuel matrix, with higher thermal and chemical stability. ThO additives to UO_2 fuels are being developed currently, with irradiation testing already under way. The benefits are likely to be worthwhile, but nevertheless incremental improvements on current UO_2 fuel technology.
- *Reprocessing*: The reprocessing of thorium fuel is less straightforward than with the uranium–plutonium fuel cycle. The recovery and purification of U-233 from neutron irradiated Th reactor fuels through Th extraction (the Thorex process) has been demonstrated on a small scale, but will require R&D to develop it to commercial readiness.
- *Inherent proliferation resistance*: It is often stated that U-233 is inherently proliferation resistant, but this is questionable. U-233 has very favourable properties for nuclear weapons, either as part of state sponsored proliferation or for an improvised nuclear device (IND). The presence of U-232 at hundred ppm levels is claimed to protect against its use in an IND, but the fact is that the protective radiation field from the U-232 is insufficient to cause rapid incapacitation and therefore only a partial barrier to an IND. Overall, the industry assesses the thorium fuel cycle as posing a comparable proliferation threat to that posed by the U-Pu fuel cycle.
- *Economics*: It is sometime stated that the thorium fuel can be more economic than the U-Pu fuel cycle. But this does not account for the fact that a thorium fuel cycle would require the development and deployment of a new infrastructure in competition with the existing U-Pu infrastructure. At present, there is insufficient understanding at the detail needed to judge whether the thorium fuel cycle would be more economic in practice.
- *Plutonium disposition*: ThO fuel theoretically offers a more stable matrix for plutonium disposition, with the advantage of avoiding the production of new Pu-239 from U-238 captures.

- *Void coefficient mitigation*: Thorium fuels drive the void coefficient toward more negative values in thermal systems. In light water reactors (LWRs), a positive void coefficient is usually considered unacceptable and limits the total plutonium load in mixed oxide (MOX) fuels to <12 w/o. This is a potential restriction with poor fissile quality plutonium. Instead, a Th–Pu fuel could allow significantly higher total plutonium loads (up to ~18 w/o), giving more flexibility for plutonium re-use in LWRs. Therefore, the Th fuel cycle could provide a possible way to manage plutonium stocks with poorer fissile quality and to allow time for thorium–plutonium MOX qualification, supplementing a U/Pu recycle strategy.

The thorium fuel cycle with full recycle will require a long term R&D programme that commercial companies cannot be expected to commit to funding and therefore will require government or supra-national investment. Globally there are thorium R&D programmes in Canada, Europe, India, Norway, China and USA. In Europe, there have been thorium projects under 5th Framework and there were historic R&D projects on thorium, including deployment in HTR and PWR. Within Gen IV, the thorium fuel cycle forms a small part of the MSR programme and also is an option for the Super Critical Water Reactor (SCWR) being led by Canada. India currently is in a leading position, with irradiation test programmes approaching commercial scale. Norway is carrying out experimental scale irradiation testing. Research requirements include:

- Irradiation testing of thorium fuel rodlets
- Thorium fuel properties measurements
- Fuel performance code development for thorium fuels and validation
- Qualification of ThO fuels for deployment in commercial reactors
- Remote fuel fabrication and design of commercial fuel fabrication plants to the point where meaningful construction and operational costs can be estimated
- Development of thorium reprocessing methods, including an understanding of waste form characterisation.

Prospects

Deployment times for utilisation of the thorium fuel cycle at commercial scale are necessarily very long. Licensing of thorium fuels in current reactor types will require fuel qualification, with lead times estimated at 10–15 years, largely determined by the time required for in-core irradiation testing. The development of new reactor designs specifically for thorium fuels will take longer. Initial cores for thorium breeder systems will need to use U-235 or plutonium as the

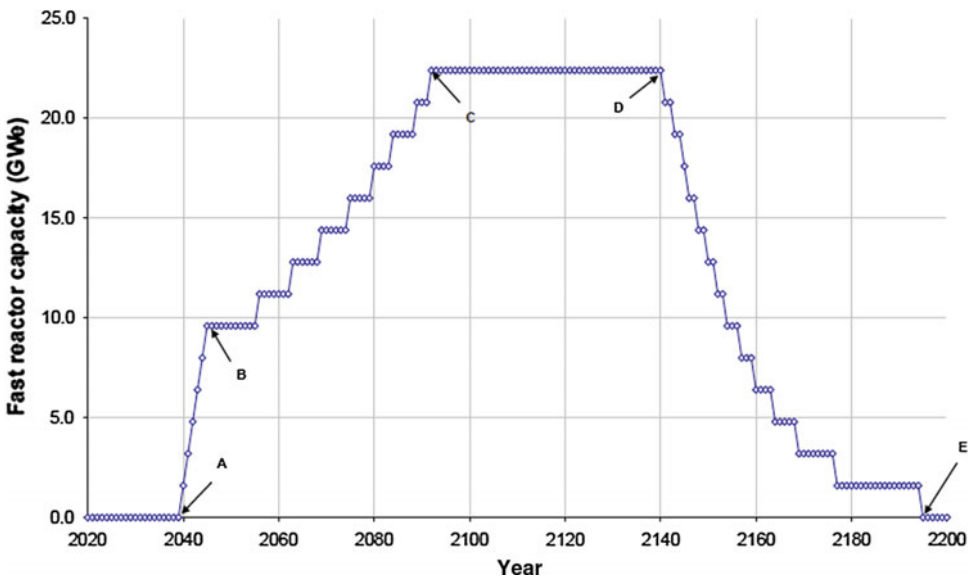

Fig. 2 Hypothetical capacity of a self-sustained breeding system

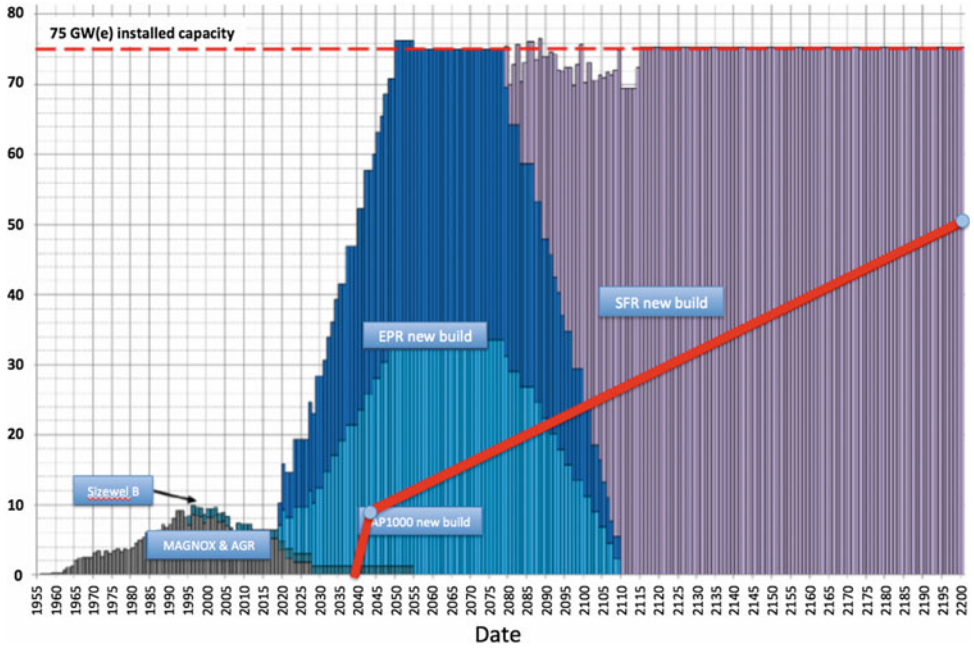

Fig. 3 Illustrative transition to a self-sustaining fast reactor fleet

fissile driver until such time as U-233 breeding reaches equilibrium. Doubling times for practical reactor designs tend to be very long and the prospect of a fully self-sustained thorium cycle is only realistic on a timescale approaching the end of the century (see Fig. 2, which illustrates the typical timescales needed for self-sustained growth in capacity of a realistic breeder reactor system and Fig. 3, which illustrates the timescales for a scenario which transitions to a self-sustained breeder reactor fleet). The full benefits of thorium recycle in terms of reducing uranium demand and lower radiotoxicity will not be achievable until the self-sustained equilibrium is established. A major impediment to the deployment of thorium fuel cycles will be the need to develop an entire new fuel cycle infrastructure to complete against the established uranium-plutonium fuel cycle. However, an even more important factor is that currently utilities do not see any clear economic incentives to develop the thorium fuel cycle. While some utilities may be amenable to hosting small scale irradiation tests of thorium fuels in their plants, there is currently little prospect of any of them investing in new reactor and fuel cycle plant designs. Getting the utilities on side will require clear drivers for them to back the thorium fuel cycle and a priority for R&D should be to identify such drivers and demonstrate that they are feasible.

Recycling Challenges of Thorium-Based Fuels

P.K. Wattal

Abstract

Deployment of thorium would involve mixed oxide fuels of Th–^{239}Pu initially, which will progressively breed ^{233}U. The recovered ^{233}U would be recycled as Th–^{233}U MOX fuel. The challenges in the recycling of Th–^{233}U MOX fuels are on account of ^{232}U which decays to hard gamma emitting daughter products ^{212}Bi (gamma: 0.7–1.6 MeV) and ^{208}Tl (gamma: 2.6 MeV), calling for heavy shielding and remote handling during transportation, fuel fabrication and reprocessing. The high content of ^{232}U in the uranium isotopic mixture impacts the separate requirements of ventilation systems during reprocessing and fuel fabrication on account of noble gas Radon (^{220}Rn) from the ^{233}U decay chain. Reprocessing of thorium-based fuels would pose quite a few challenges that are unique to Th–^{239}Pu and Th–^{233}U MOX fuels. Thoria dissolution would be in the presence of sodium fluoride or HF as catalyst on account of its inertness. Reprocessing would call for disassembly of the fuel pins in order to segregate Th–Pu pins and Th-U pins initially till enough of ^{233}U is available finally to avoid ^{235}U or ^{239}Pu in the MOX fuels. High Level Liquid Wastes (HLLW) generated during reprocessing of thoria based fuels would have high concentrations of alumina as mentioned earlier. This would call for high melting and pouring temperatures for the vitreous formulations to be deployed for this HLLW. Partitioning of HLLW, if deployed, would additionally call for removal of ^{231}Pa ($t_{1/2}$ 32,500 years) because of its being highly mobile. This is in addition to those of minor actinides (MA) which would in any case be also required to be portioned even in uranium-based spent fuel. In India, based on years of R&D work on various aspects of Th-^{233}U-based fuel along with our expertise on the PUREX process over four decades, a process sheet for three component reprocessing of thoria based irradiated fuels has been studied and laboratory scale experiments carried out to validate the process steps. Processing of irradiated thoria bundle from research reactor has been carried out to establish the process. A plant scale processing of irradiated thoria bundles from Pressurized Heavy Water Reactor (PHWR) is under construction.

Introduction

Thorium has a natural abundance of three to four times that of uranium and is uniformly distributed in the Earth's crust. Utilization of the abundant resources of Th has had a great impact on the economical sustainability of nuclear energy. Thorium, being a fertile material, requires fissile material such as ^{235}U, ^{239}Pu, or ^{233}U for energy extraction. As an example, deployment of thorium would involve using a mixed oxide (MOX) fuel, for example, Th–^{239}Pu initially, which would progressively breed ^{233}U. The recovered ^{233}U can be recycled as a Th–^{233}U MOX fuel for energy utilization.

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Table 1 Minor actinide generation (g/tonne) for the uranium versus thorium fuel cycles for a burnup of 60 GWd/tonne

MA	$U^{235} + U^{238}$	$U^{233} + Th^{232}$
Np	900	3
Am	470	0.002
Cm	220	0.0006

Attractive features of the thorium cycle are not only on account of resource availability, but also the relative ease of waste management. This is on account of:

1. better stability in geological repositories, if disposed of directly;
2. ^{233}U proliferation resistant owing to the presence of high-energy ^{232}U;
3. reduced generation of long-lived minor actinides (MA) compared with the uranium cycle.

Table 1 gives a comparison of MA generation for a burnup of 60 GWd/tonne for the uranium and thorium cycles.

A prerequisite for the introduction of thorium, a fertile element in the fuel cycle, is to generate the much needed fissile inventory in it. The design of three stage Indian nuclear energy programme is the outcome of this realization. Parallel with the generation of fissile inventory, development of Th based fuel reprocessing and recycling technology is also vital to meet the challenges offered by this cycle. In this direction a systematic work is being carried out at Bhabha Atomic Research Centre, (BARC), Trombay. This programme is directed towards dissolution of thoria irradiated in research and power reactors, followed by separation of Th, U and fission products (FPs) deploying solvent extraction.

Processing of Irradiated Thoria Pellets

Since the late sixties, thorium has been irradiated in the research reactor CIRUS, located at Trombay, India, and the first pilot-plant-scale separation of ^{233}U from irradiated thoria was achieved in 1970 by using a process based on the acid THOREX process. The first pilot-plant processed thoria was irradiated in the reflector region of the research reactor CIRUS with thermalized low flux. Chemical decladding with NaOH was adopted in view of aluminium being the cladding material. The exposed thoria was dissolved in a thermosyphon dissolver containing HNO_3, sodium fluoride, and aluminium nitrate. The dissolution could be completed in about 12 h. The fission product activities as well as the ^{232}U content were very low. All the operations were carried out in a series of glove boxes arranged in a row. ^{233}U was separated from thorium and fission products by extraction in 5 % tributyl phosphate (TBP) in dodecane, followed by scrubbing with 1–2 M HNO_3, and stripping with demineralized water.

After successful completion of the above campaign, another pilot facility at Kalpakkam, India, was set up to test and standardize equipment and remote-handling operations. These campaigns provided enough impetus to develop a more robust acid "THOREX" process. The ^{233}U recovered from these campaigns was deployed for fuelling an experimental reactor, "KAMINI". This reactor is perhaps the only reactor in the world with ^{233}U as fuel and it delivers 30 kW of thermal energy at full power.

Reprocessing of Irradiated Thoria Bundles from Research Reactors

In 2002, an engineering facility for ^{233}U separation, known as the Uranium Thorium Separation Facility (UTSF), was designed and commissioned at the Bhabha Atomic Research Centre (BARC), Trombay, for the processing and recovery of ^{233}U from CIRUS-irradiated thoria rods on a regular basis. A modified process flow sheet using 3 % TBP in *n*-dodecane was deployed for preferential extraction of ^{233}U with lesser co-extraction of thorium in the organic phase. Specially designed Combined Airlift Mixer Settler Units (CALMSUs) were chosen as contactors for the solvent extraction. A photograph of the facility, showing the in-cell piping, is given in Fig. 1.

The process flow sheet deployed for the preferential separation of ^{233}U in UTSF involved using a single-cycle, modified THOREX flow sheet. The thoria fuel irradiated in the research reactor was processed successfully. Near complete recovery of thorium from the THOREX raffinate stream was also achieved based on a flow sheet developed in-house. Over 99.5 % recovery of thorium was achieved. The thorium lean raffinate (TLR) was vitrified, thereby closing the back-end of the thorium-based fuel cycle. A typical process flow sheet is shown in Fig. 2.

Reprocessing of Irradiated Thoria Bundles from Power Reactors

A Power Reactor Thoria Reprocessing Facility (PRTRF) has been set up at BARC, Trombay. This facility has a mandate to reprocess thoria bundles irradiated at 220 MW_e

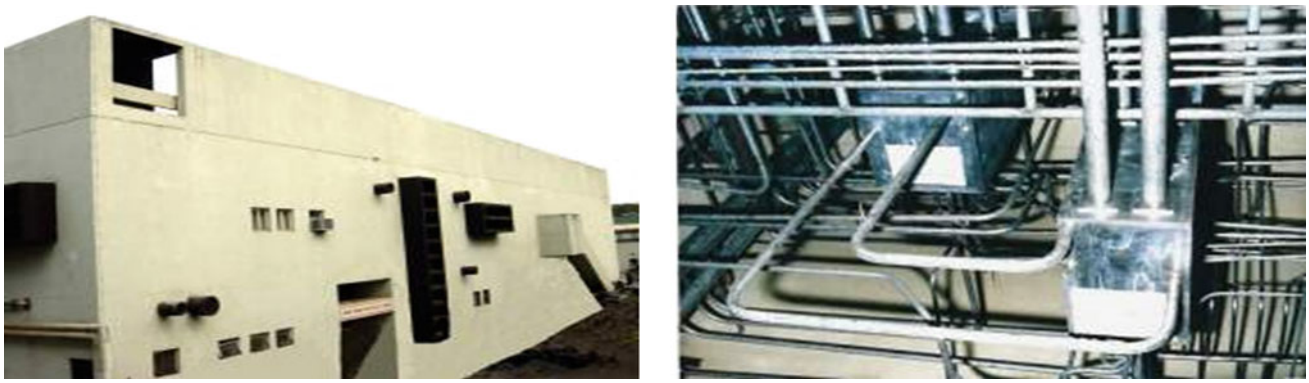

Fig. 1 Uranium–thorium separation facility with in-cell piping

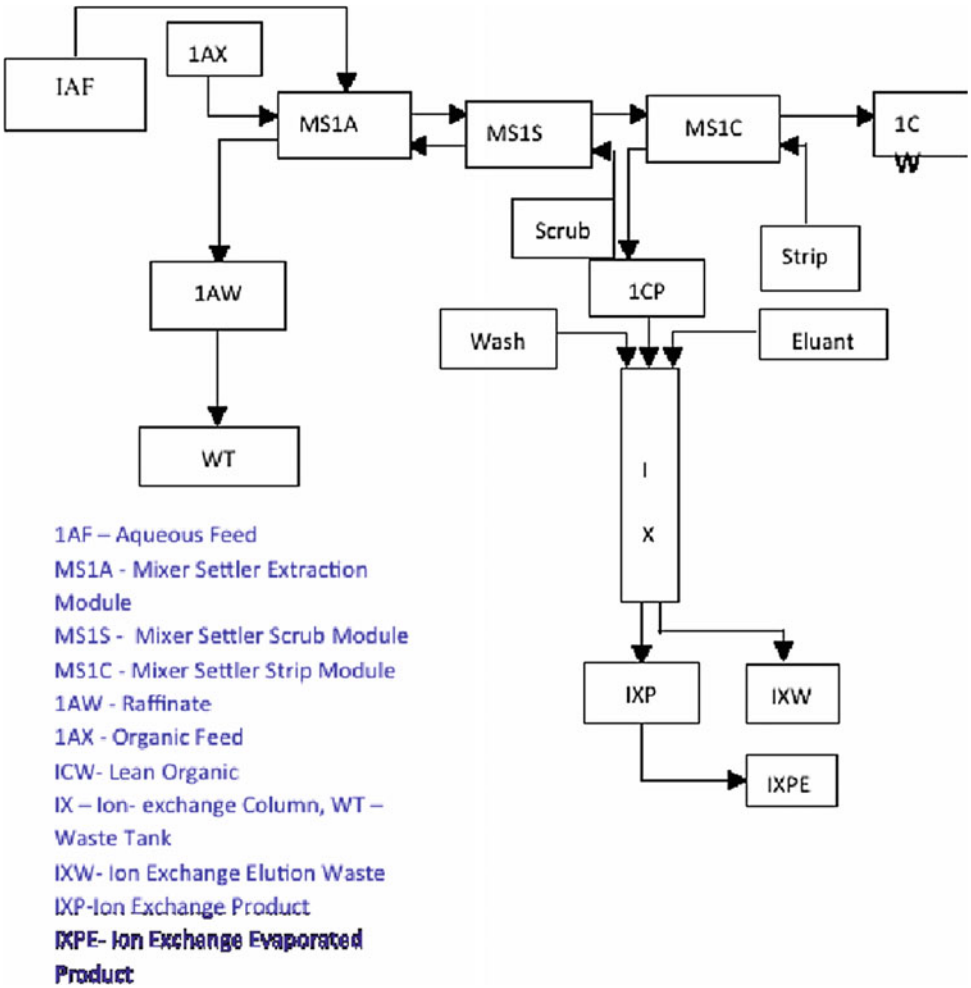

Fig. 2 Process flow sheet for preferential separation of ^{233}U

pressurized heavy water reactors (PHWRs) for extraction of ^{233}U. A few hundred of such irradiated bundles are available in different PHWRs and are awaiting reprocessing in this engineered facility. These bundles have been put in the reactor for initial flux flattening. An in-cell view of this engineered facility is shown in Fig. 3.

The PRTRF facility is comprised of shielded cells that have chopping and dissolution, followed by separation using the solvant extraction process. This facility is ready for hot commissioning to embark on the third stage of the Indian nuclear power program. Chemical dissolution of zircaloy-clad spent fuel bundles in nitric acid has been

Fig. 3 Control room and in-cell extraction facility of PRTRF

Fig. 4 Sequence of indexing, laser chopping, a cut fuel bundle, and fuel pin dropping

deployed. Development of a "Laser Assisted Fuel Bundle Dismantling and Single Pin Chopping System" has been adopted in this facility. A good experience in laser dismantling of multipin bundles on an industrial scale is required for achieving clean-cut quality to reduce losses in the hull. Laser dismantling of multipin bundles also provides a platform for the segregation of pins with different fuel materials for different streams of reprocessing. Figure 4 shows different operations of laser-assisted fuel bundle dismantling and chopping.

The challenges in the recycling of Th–^{233}U MOX fuels are a result of ^{232}U, which decays to hard gamma-emitting daughter products, ^{212}Bi (gamma: 0.7–1.6 MeV) and ^{208}Tl (gamma: 2.6 MeV), requiring heavy shielding and remote handling during transportation, fuel fabrication, and reprocessing. The high content of ^{232}U in ^{233}U impacts the requirements of the ventilation systems during reprocessing and fuel fabrication on account of noble gas radon-220 from the ^{233}U decay chain. ^{220}Rn gas can pass through high-efficiency particulate air (HEPA) filters and then decay to solid ^{208}Tl. The considerations of hard gamma emissions would also necessitate the co-location of fuel fabrication with reprocessing facilities to minimize the time interval between the two operations and to encapsulate the fuel pins before the hard gamma activity builds up.

Reprocessing of thorium-based fuels would pose quite a few challenges that are unique to Th–^{239}Pu and Th–^{233}U MOX fuels. Although dissolution of uranium-based fuels is performed in hot concentrated nitric acid, thoria dissolution would require the presence of sodium fluoride or HF as a catalyst on account of its inertness. To mitigate the concerns of fluoride corrosion in the equipment (for example, dissolvers), aluminium nitrate can be added as a fluoride complexing agent. The presence of aluminium, along with traces of free fluoride, would eventually have an impact during vitrification of the raffinate generated.

Reprocessing would call for disassembly of the fuel pins in order to segregate Th–Pu pins and Th–U pins initially until enough ^{233}U is available to avoid ^{235}U or ^{239}Pu in the MOX fuels. Reprocessing Th–U pins will require two-stream reprocessing, whereas as Th–Pu pins will call for three-stream reprocessing to enable separation of Th, U, and Pu. Thus, this would call for separate sets of reprocessing cycles. Additionally, as a measure against the deployment of externally generated U^{IV} during the separation of U and Pu, chemical reductants, such as HAN, or in situ electrochemical

reduction shall have to be employed, as isotopic dilution of ^{233}U would not desirable with natural uranous salts.

Management of High Level Liquid Wastes from the Reprocessing of Thoria Based Fuels

High level liquid wastes (HLLW) generated during the reprocessing of thoria-based fuels will have high concentrations of alumina, as mentioned earlier. This would call for high melting and pouring temperatures for the vitreous formulations to be used for this HLLW. High temperature availability in the vitrification melters may, therefore, be desirable, which would lead to requirements for different types of vitrification melters, such as cold crucibles. Partitioning of HLLW, if deployed, would additionally call for removal of ^{231}Pa ($t_{1/2}$ = 32,500 year) because of its high mobility. This is in addition to the removal of minor actinides (MA), which would also need to be portioned in uranium-based spent fuel anyway.

The problems associated with ^{232}U being present (500 ppm) in ^{233}U can be resolved by providing adequate residence time (delay) in the process off-gas system before discharging through stacks (after filtration). After separating ^{233}U from thorium, the ^{233}U should be processed immediately in the laboratory to avoid the dose build-up due to the presence of ^{232}U. The separated thorium should be stored in a storage facility for 20 years to allow decay of ^{228}Th before reuse in the reactor. This will facilitate the handling of recovered thorium in unshielded containment systems.

Reprocessing of Thorium Fuel: Pyrochemical and Aqueous Routes

Sylvie Delpech and Davide Rodrigues

Abstract

Nuclear fuel processing is a prerequisite for nuclear energy to be a clean and sustainable energy. Studies on the treatment of thorium-based fuel follow two options, pyrometallurgical (or high temperature using molten salts) and hydrometallurgical depending on the nature (liquid or solid) of the fuel. For a liquid fuel, as in the case of a molten salt reactor (MSR), pyrometallurgical processing is an obvious route. For fuel based on thorium oxide, the classical treatment is the hydrometallurgical route, but the high-temperature processes could also be considered. The treatment method for the liquid fuel of MSRs is divided into two parts: (1) in situ injection of helium gas into the fuel extracts the gaseous fission products and some of the noble metals; (2) reprocessing is performed by "batch", which aims to recover the fissile material and to separate the minor actinides from fission products. Minor actinides are returned to the reactor to close the fuel cycle and the fission products are managed for storage. The scheme reprocessing involves several chemical steps based on the redox and acid–base properties of the different elements in the liquid fuel salt. These processes are currently being studied experimentally in the frame of national and European research programs. Regarding the treatment of thorium solid fuel, two methods are generally envisaged. The Thorex process is a process similar to the liquid–liquid extraction Purex process. It has been successfully implemented at the pilot scale, but requires further study, in particular, for the dissolution step of thorium, which is inefficient. The other method adopted by India is the Interim-23 process, which extracts uranium but leaves the thorium in the waste. Finally, pyrometallurgical routes for fuel processing of thorium oxide have not been extensively studied. The aim of this paper is to present the reprocessing of thorium as both solid and liquid fuel.

Introduction

Nuclear fuel processing is a prerequisite for nuclear energy to be a clean and sustainable energy. Nuclear reprocessing technology has been developed to chemically separate and recover fissionable plutonium from irradiated nuclear fuel. Thus, the aim of reprocessing is to free the unspent nuclear fuel from the radioactive fission products and to separate the fertile and fissile materials, ^{238}U and ^{239}Pu or ^{232}Th and ^{233}U, depending on the fuel cycle considered. In a closed fuel cycle (advocated for Generation IV reactor systems), fertile and fissile materials are put back into the fuel cycle while fission products are treated and disposed in deep storage.

The current power plants operate with uranium solid fuel. The aqueous process Purex [1] is today the principle process employed worldwide for the reprocessing of spent nuclear fuel. However, the quest for pyrochemical alternatives to aqueous reprocessing [2] has been under way for many years in various countries and in the USA since the late 1950s [3]. Various techniques have been studied [4]: alloy melting,

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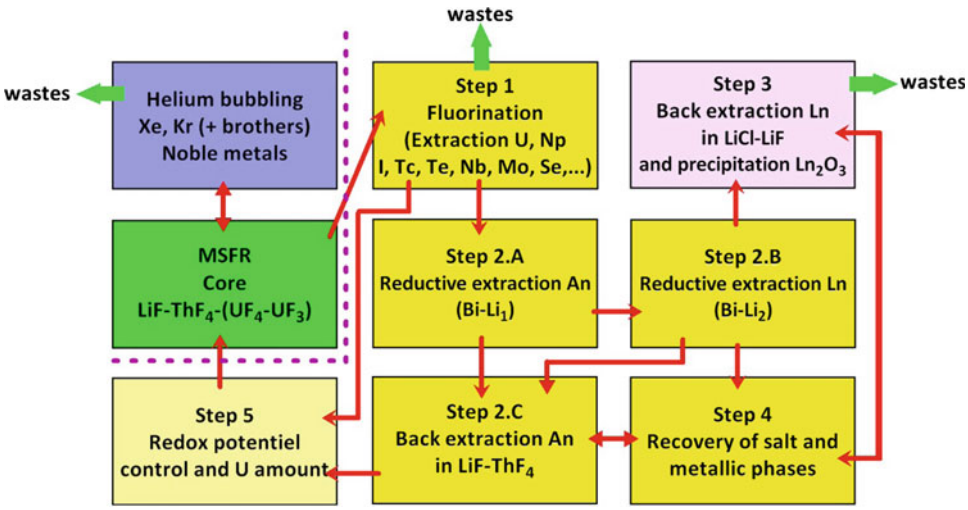

Fig. 1 Reprocessing scheme for the MSFR concept

fission product volatilization and adsorption, fluoride and chloride volatility methods, redox solvent extraction between liquid salt and metal phases, precipitation and fractional crystallization, electro-winning and electrorefining of actinide metals and oxides.

Concerning the thorium fuel cycle, the Thorex process is similar to that of the Purex process for aqueous reprocessing, but is based on liquid–liquid extraction. When the fuel is liquid, as it is the case for the molten salt reactor (MSR) concept, pyro-reprocessing is a clear choice. A global reprocessing scheme has been established these last years in the frame of the EURATOM EVOL program. Several elementary steps involved in the scheme have been demonstrated from a thermodynamical point of view.

This paper initially presents the pyro-reprocessing scheme for molten salt fast reactors (MSFR), which is the concept of MSR developed by CNRS [5] in the frame of the EVOL project, including descriptions of each step and the behavior of the several elements during the treatment. The second part is dedicated to the aqueous reprocessing of solid thorium oxide fuel currently under study.

Pyro-Reprocessing for the MSFR Concept

The MSFR concept is an innovative type of MSR under development by CNRS since 2004. Contrary to the numerous previous MSR concepts [6], MSFR is operated in a fast neutron spectrum owing to the removal of graphite in the core and of BeF_2 in the molten salt composition. The salt is constituted of LiF–ThF_4–UF_4 with 77.5 mol% of LiF. The working temperatures range between 700 and 900 °C.

The objectives of reprocessing are:

- The production of ^{233}U (necessary to ensure nuclear energy sustainability, part of the MSFR as a breeder concept);
- The extraction of fission products (FPs) for deep storage. Fission products have to be removed mainly because of their low solubility, which can induce the formation of solid particles in the reactor. The neutron captures of lanthanides, which were a drawback in previous concepts (operated in a thermal neutron spectrum), is not an issue in the fast neutron spectrum.
- The introduction of actinides (An) back into the reactor core (to close the fuel cycle, MSFR being a burner concept). The separation FPs/actinides strongly decreases the toxicity of the waste.

The reprocessing scheme proposed for MSFR is given in Fig. 1. It was established by considering the chemistry of the elements in the fuel salt. Each step is based on the redox and acid–base properties of the elements.

Figure 1 shows that the reprocessing is divided into two parts: the on-line reprocessing, which consists of the injection of helium gas into the core, and the off-line reprocessing, which is realized by batch and constitutes several steps. The volume of salt removed from the core to be treated each day is about 40 L.

Unlike the concept molten salt breeder reactor (MSBR) developed by the Oak Ridge National Laboratory (ORNL) in the 1960s [7, 8], which operated in the thermal neutron spectrum, the removal of ^{233}Pa is not an issue in the MSFR concept because it operates in a fast neutron spectrum. Indeed, owing to the neutron captures occurring with ^{233}Pa and FPs in the MSBR, the optimized rate of reprocessing was about 4000 L/day. Therefore, the reprocessing rate is strongly decreased as a result of the fast neutron spectrum, from 4000 L/day in the MSBR concept to 40 L/day in the MSFR. Moreover, no dedicated step for ^{233}Pa removal is required in the scheme.

Table 1 List of the elements produced in the MSFR core during operation, the chemical state of the element when it is produced, the process used to remove the element, and some comments

	Extraction process	Comments	State of element in fuel salt
		The state of the elements depends on the redox conditions of the fuel salt. The redox conditions are given by the ratio U (IV)/U(III) which varies from 100 to 10. Therefore, the redox potential of the salt is ranging between −3.0 and −3.3 V	
Es	ER1	No data	
Cf	ER1	No data	
Bk	ER1	No data	
Cm	ER1		CmF_3
Am	ER1		AmF_3
Pu	ER1		PuF_3
Np	ER1		NpF_3
U	FLUO		UF_4/UF_3
Pa	STOP + FLUO		PaF_4
Th	CORE		ThF_4
Ac	ER1		AcF_3
Ra	ER1		RaF_2
Bi	FLUO	Bi produced at metallic and liquid state. Can form BiF_3 gas under fluoration. Can also form BiNi alloys with structural materials	Bi
Pb	FLUO	Pb produced at metallic and liquid state	Pb
Er	ER2		ErF_3
Ho	ER2		HoF_3
Dy	ER2		DyF_3
Tb	ER2		TbF_3
Gd	ER2		GdF_3
Eu	ER2		EuF_3
Sm	ER2	SmF_2 can be also stable (depends on redox conditions)	SmF_3
Pm	ER2	No data	PmF_3 ?
Nd	ER2		NdF_3
Pr	ER2		PrF_3
Ce	ER2	Can also be oxidized to CeF_4 gas by fluorination	CeF_3
La	ER2		LaF_3
Ba	CORE		BaF_2
Cs	CORE		CsF
I	FLUO		I
Sb	FLUO	Sb produced at metallic and liquid state. Can form SbF_5 gas by fluorination. Formation of NiSb alloys possible	Sb
Sn	FLUO	Sn produced at metallic and liquid state. Can form SnF_4(g) gas by fluorination at 800 °C. Formation of Ni_3Sn, Ni_3Sn_2 and Ni_3Sn_4 alloys possible	Sn
In		E = −3.36 V. ER seems possible but activity coefficients of In in Bi required to conclude	InF_3
Cd	He	E = −2.87 V. Cd is produced at metallic and liquid or gaseaous state (Tvap = 765 °C). So extraction by He bubbling	Cd
Zr	He	E = −4.2 V. ZrF_4 is gaseous at 912 °C. Can be extracted with He and F_2(g)	ZrF_4
Y	ER2	E = −5.13 V	YF_3

(continued)

Table 1 (continued)

	Extraction process	Comments	State of element in fuel salt
Sr		E = −5.45 V	SrF_2
Rb		E = −4.77 V	RbF
Br	FLUO		Br(−)
Ge	He	Ge produced at metallic state. Can produce GeF_2 gas by fluorination. Ni_2Ge alloys possible	Ge
Ga	FLUO	Ga produced at metallic and liquid state. All Ga fluoride are gaseous. No data about Ni-Ga alloys	Ga
Cu	He	CuF_2 after oxydation by F_2(g). CuF_2 logK = −25 at 800 °C for CuF_2(g). ER possible	Cu
Ni	He	NiF_2 by fluorination. Not gaseous. ER possible	Ni
Co	He	E = −2.83 V. CoF_3 by fluorination. Gas at 900 °C. ER possible	Co
Fe	He	E = −3.07 V. Extraction possible by He bubbling depending on redox conditions. ER seems possible depending on activity coefficients of Fe in Bi and solubilities in Bi	Fe/FeF_2
Mn		Formation of MnF_3 under fluorination but MnF_3 is not a gas. Reduction of MnF_2 to Mn in Bi pool possible by ER1 and ER2. Efficiency calculation required activity coefficients of Mn in Bi	Mn and MnF_2
Cr		Idem Mn	
V			
Ti			
Sc	ER1?	E = −4.86 V. ER possible. Requirement of activity coefficients in liquid Bi	ScF_3
Ca	CORE		CaF_2
K	CORE		KF
Cl	FLUO		Cl(−)
S	FLUO	Li_2S for E < −3.43 V. All the fluoride sulfurs are gaseous. Formation of SF_6(g) by fluorination	S and S(–II)
F	CORE		F(−)
O	FLUO	O_2(g) by fluorination	O(–II)
B	He	BF_3(g) in the redox conditions of the salt	BF_3(g)
Be	CORE		BeF_2
Li	CORE		LiF

[a]RE1 and RE2 are respectively the first and the second reductive extractions, FLUO the fluorination step, He the removal by helium bubbling. When CORE is indicated that means that the element is not removed from the salt

Step-by-Step Reprocessing

On-Line Reprocessing

The action of helium injection into the core (helium is a gas with a very low solubility in the molten salt) is (i) to help remove gaseous fission products (particularly Xe and Kr, which are neutronic poisons) and (ii) to allow the extraction of the produced noble metals in their metallic state by a physical process similar to flotation. The elements being removed in this step are given in the Table 1.

Off-Line Reprocessing

The spent fuel salt treated in this part of the reprocessing is the salt removed by batch from the core; thus, 40 L/day.

Step 1: Fluorination

This step consists of the injection of fluorine gas into the spent fuel salt. In this way, all the elements are oxidized to their higher oxidation state by the chemical reaction:

$$MF_x + F_2(g) \rightarrow MF_{(x+2)} \quad (1)$$

In some case, the resultant $MF_{(2+x)}$ is in the gaseous state. All the metal fluorides in their gaseous state are naturally removed from the liquid salt. That is case for uranium, neptunium, and numerous fission products. The facility to perform the fluorination step was described by ORNL (Fig. 2). To avoid corrosion of the structural material, the wall of the reactor is cooled by a NaK coolant. That leads to the formation inside the reactor of a frozen wall of salt, which protects the materials against corrosion.

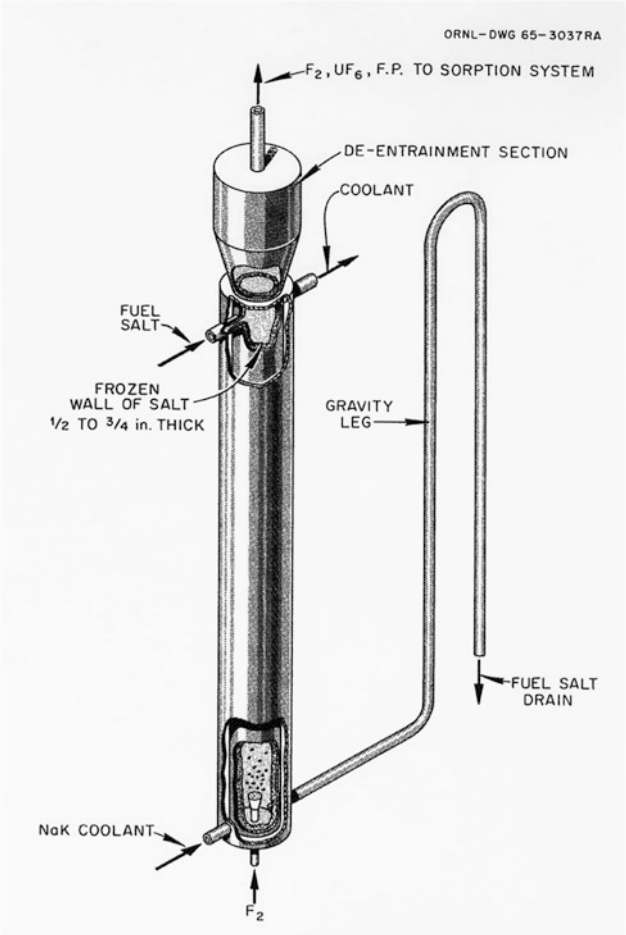

Fig. 2 Fluorination vessel [9]

After separation from the liquid salt, the gaseous elements are absorbed by NaF traps. By heating the NaF traps at given temperatures, the actinides (U, Np) are separated from the FPs. Then, the elements are reduced by a chemical reaction with H_2 before being introduced back into the core (for the actinides) or sent to waste storage (for the FPs).

Step 2: Reductive Extractions (2a and 2b)

The steps 2a and 2b are dedicated to the extraction, respectively, of (non-gaseous) actinides (Pu, Am, Cm, etc.) and lanthanides. These two steps are performed by the same method; reductive extraction, which consists of contacting the fuel salt with a liquid metal. The liquid metal is a mixture of Bi and Li. Bi is the solvent metal and is Li the reductant element, which allows the transfer of the elements dissolved in the salt to the liquid metal. The chemical reaction is given by:

$$\mathrm{AnF_{3(fuel\ salt)}}\left(\text{or } \mathrm{LnF_{3(fuel\ salt)}}\right) + 3\mathrm{Li_{(Bi)}} \rightarrow \mathrm{An_{(Bi)}}\left(\text{or } \mathrm{Ln_{(Bi)}}\right) + 3\mathrm{LiF_{(fuel\ salt)}} \quad (2)$$

The efficiency of the extraction of An and Ln and the selectivity of the separation An/Ln depend on the composition of the two metallic phases involved in steps 2a and 2b. These compositions have been optimized by using an extraction model established recently [10] and applied by combining bibliographic and thermodynamical data.

The composition of the solvent metal for the actinides extraction is Bi–Li(0.1 mol%) and for the second extraction (lanthanides) the composition is Bi–Li(10 mol%). Under these conditions, the number of stages required to reach 99 % efficiency can be determined for each element.

Step 3: Back-Extraction of Lanthanides

The back-extraction of lanthanides is performed by anodic oxidation of the Bi pool, which contains the lanthanides transferred during step 2b. The back-extraction is realized in a molten salt constituted of the eutectic mixture LiCl–LiF (70:30 mol%), the melting point of which is 500 °C. After oxidation of the lanthanides and their dissolution in the molten salt, the lanthanides are precipitated as oxides. An Ar–H_2O gas mixture is introduction in the molten salt to precipitate solid Ln_2O_3. The chemical reaction is given by:

$$2\mathrm{LnF_3}\left(\text{or } \mathrm{LnCl_3}\right) + 3\mathrm{H_2O(g)} \rightarrow \mathrm{Ln_2O_3} + 6\mathrm{HF(g)}\ \left(\text{or } 6\mathrm{HCl(g)}\right) \quad (3)$$

The products of this reaction are Ln_2O_3, which is easily separated from the salt, and HCl or HF, which are gases and are easily removed from the salt. Therefore, the composition of the salt is not modified by this reaction. The precipitation of lanthanide oxides has been demonstrated and a patent related to this method has been filed [11].

Step 2c: Back-Extraction of Actinides

After steps 2a and 2b, the fuel salt is clean and is constituted of LiF–ThF_4. By contacting the metallic phase, which contains the actinides (after step 2a) and the cleaned fuel salt, actinides are transferred from the metal to the salt by an anodic oxidation.

Step 4: Introduction of ^{233}U and Redox Control of the Fuel Salt

At this point, the fuel salt contains LiF–ThF_4 and all the actinides introduced by step 2c. During step 4, the actinides removed during the fluorination step (step 1) are introduced back into the fuel salt after reduction in presence of hydrogen gas. The amount of ^{233}U introduced in this step is optimized for the proper operation of the reactor.

Introduction of uranium also permits control of the redox potential of the fuel salt. The redox potential of the salt is a main parameter that prevents the chemical corrosion of the structural material [12].

Step 5: Recovery of Salt and Metallic Phase Composition

During steps 2a, 2b, 2c, and 3, the composition of several phases, both salts and metals, have been modified. Indeed, the extraction of lanthanides and actinides is accompanied by an increase in the amount of LiF in the fuel salt (LiF–ThF_4) and a decrease of metallic Li in the Bi solvent. Therefore, recovery of the original compositions of all the phases is required to have a balance of the reprocessing equal to zero. Electrochemical techniques are envisaged to perform this step.

The Behavior of the Elements During Reprocessing

The behavior of the elements depends on the redox and acid–base properties of the elements in the fuel salt, LiF–ThF_4. Knowledge of thermodynamic and solvation data is required to predict the behavior of the elements during each step. Table 1 summarizes the state of our knowledge today. For some elements, no technique is currently proposed for their removal from the fuel salt. That is due to the lack of thermodynamic and/or solvation data.

Aqueous Reprocessing of Solid Thorium Oxide Fuel [13, 14]

The Thorex process was originally developed at Knolls Atomic Power Laboratory (KAPL) and Oak Ridge National Laboratory (ORNL) [15, 16]. As in the Purex process, after dissolution of the solid oxide in a nitric acid solution, a solution of tributyl phosphate (TBP) in an inert solvent is used as an extractant. Contrary to the Purex process, whereby the several valences of Pu and U are used to perform the dissolution and the separation, thorium is constantly and naturally in the +IV valency.

A simplified process flow chart, the so-called INTERIM-23 process, omits a partitioning of the Th, which is passed into the waste stream together with the fission products. Uranium is selectively extracted with a 5 % TBP solution [17].

The two main steps of the Thorex process can be briefly described as follows.

Solid Fuel Dissolution

The dissolution of the fuel is realized in an aqueous solution of HNO_3 (13 M), HF (0.05 M), and $Al(NO_3)_3$ (0.1 M). HF is added to form complexes with thorium. To prevent corrosion of the equipment by HF, aluminum nitrate is added as a buffer. The dissolution of thoria can take up to 35 h under these conditions. Agitation through air-lift circulation has been tested with good results; the time of dissolution under these conditions is about 10–12 h. However, it seems that a residual blue solid (so-called blue thorine) is not dissolved. Another drawback is the partial dissolution of Zr (about 5 %) in HF solutions. This has to be taken into account for the rest of the process.

The sonochemistry technique, which is based on using ultrasound to improve the agitation process, is currently being studied.

Liquid–Liquid Extraction

The liquid–liquid extraction is performed by contacting the aqueous solution of dissolution with a solvent phase of dodecane and TBP (30 %).

Contrary to the Purex process in which the Pu/U separation is possible by drawing on the Pu(IV)–Pu(III) valence change, the separation in the Thorex process depends solely on the different distribution ratios in the Th/U system. The distribution ratios depend on the concentration of HNO_3 in the aqueous phase and on the concentration of TBP in the organic phase. A relatively high acid concentration in the scrubbing part of the extraction process ensures almost quantitative Th extraction. But, on the other hand, high acid concentrations favor the extraction of some FPs and, thus, deteriorate the decontamination factors achievable.

Th(IV) tends to form a third phase much more readily than U(VI). This phase consists of TBP and thorium and perturbs the function of the extraction as a result of its physical properties. Therefore, the maximum Th loading of the organic phase should be limited to roughly 30 % of the theoretical capacity. The concentration of thorium is, thus, limited to 1 M, whereas it is possible to feed 1.4 M solutions into the Purex process.

A high acid concentration also favors radiolysis of the TBP and produces dibutyl phosphate (DBP), which forms very stable complexes of Th–DBP in the organic phase (slag formation).

The back-extraction of thorium is performed with diluted HNO_3 and, finally, the back-extraction of U is realized with very dilute HNO_3.

Conclusion

Studies are currently being pursued both in the field of the pyrochemical reprocessing of liquid thorium fuel, particularly in the areas where thermochemical and solvation data are missing, and also in the realization of experiments to validate the different steps of the reprocessing.

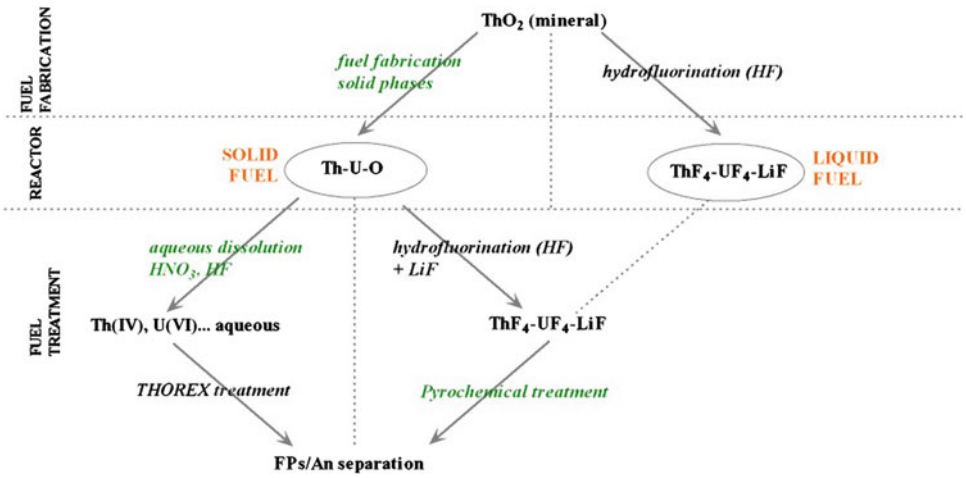

Fig. 3 Pyrochemical treatment of solid and liquid fuels based on thorium

Sonochemistry is being studied to improve the dissolution step of (Th, U)O_2 fuel. Results will be published shortly.

Combining pyrochemistry and solid oxide fuel could be an alternative to the Thorex process, as is shown in Fig. 3.

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Paul Scherrer Institute's Studies on Advanced Molten Salt Reactor Fuel Cycle Options

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Abstract

Nuclear reactors operated with liquid fuel may have several remarkable advantages and features. The most developed reactor system in this category is the molten salt reactor (MSR). It represents an old concept, but its properties qualify it for advanced utilization; these include inherent safety, excellent neutron economy, continuous or batch reprocessing possibilities without fuel fabrication. The aim of this study is to characterize the MSR physics, highlighting its unique fuel cycle advantages in several different neutron spectra by using the ERANOS-based EQL3D procedure and ECCO-MATLAB- or Serpent-MATLAB-based EQL0D procedures.

Introduction

The molten salt reactor (MSR) is an old concept [1, 2], which is presently re-viewed because of its appealing features:

1. Excellent neutron economy of the molten salt and of the thorium fuel cycle;
2. Liquid fuel does not need fabrication and enables, for example, curium recycling;
3. Flexibility of the fuel cycle thanks to the previous two advantages;
4. Inherent safety based on negative feedback coefficients and continuous salt clean-up from highly mobile gaseous and volatile fission products.

Contemporary research reopens the discussion about the advantages and disadvantages of the technology. The main challenges related to MSRs are:

1. Structural material corrosion and irradiation embrittlement;
2. Specific thermal-hydraulics, dynamics, and limited graphite lifespan, if applied;
3. Demanding molten salt reprocessing techniques;
4. Fuel salt selection, treatment, and possible proliferation risks.

The advantages may enable the design, from the neutronics point of view, of an inherently safe MSR with flexible fuel cycle and may represent a strong motivation to overcome the challenges. Furthermore, the advantages may be partly sacrificed to reduce the challenges. This is the general idea adopted by many research teams and also in our research. It is based mainly on MSc students', PhD students', and post-docs' works, which can be integrated into a students' reactor design project. It may be, in the future, extended into other relevant fields (thermal-hydraulics, salt chemistry, materials behavior, etc.) and supported also by senior researchers.

Appealing MSR Features

Excellent Neutron Economy

MSR fuel consists of a molten carrier salt mixed with the salts of actinides and fission products. Generally, two basic

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salts types, foreseen for two different applications, are considered in MSR research:

1. Fluoride salts for the iso-breeding, low-waste-production Th–U cycle;
2. Chloride salts for the high-conversion U–Pu cycle.

The fluorides have, in general, lower parasitic capture probability and provide, thanks to the high inelastic scattering cross section of ^{19}F, a relatively softer spectrum. As ^{232}Th has lower fissionability in the fast spectrum, and because ^{233}U as a main fissile isotope produces only 2.5 neutrons per fission in any spectrum, fluorides are well suited for the Th–U cycle and its high neutron efficiency. This conclusion is also supported by the high solubility of Th and U tetrafluorides in the carrier salt, whereas the solubility of Pu, which forms trifluorides, is limited. Furthermore, under certain conditions, U, Np, and partly also Pu, may form volatile hexafluorides and can be easily separated from the rest of the fuel, by so-called volatilization. Accordingly, fluorides are usually proposed for a clean Th–U cycle in thermal or fast spectra.

The chlorides, on the other hand, have higher parasitic capture probability; however, the high inelastic scattering cross section of ^{19}F is absent and a relatively hard spectrum can be achieved. In such a spectrum, the ^{238}U high fissionability may already play an important role. The main fissile isotope, ^{239}Pu, of the U–Pu cycle produces up to 2.9 neutrons per fission in the fast spectrum and so the higher parasitic capture probability of the chlorides may be acceptable. Furthermore, as the solubility of both U and Pu is high, the chlorides are more suitable for the fast spectrum U–Pu cycle. These conclusions may change in cases where natural Cl is replaced by enriched ^{37}Cl.

The excellent neutron economy of the Th–U fuel cycle is predominantly based on the low neutron capture probability of the main fissile isotope, ^{233}U. It is present at around 10 % in both thermal and fast MSR reactors (Fig. 1). Furthermore, the secondary fissile isotope, ^{235}U, created subsequently from the 10 % of parasitic captures, also has a low capture probability. Moreover, if compared with the secondary fissile isotope, ^{241}Pu, in the U–Pu cycle, ^{235}U is stable, whereas ^{241}Pu decays and worsens the neutron economy. Figure 1 also illustrates that if the ^{239}Pu fission fails, ^{240}Pu, a gateway to minor actinides (MA) buildup, is created. In the Th–U cycle, ^{239}Pu is created through the ^{237}Np and ^{238}Pu path, whereas in the U–Pu cycle it is created directly.

The excellent neutron economy of the Th–U cycle in both thermal and fast spectra was also proven by a spectral study [3]. The results of the ERANOS-based EQL3D procedure [4] and ECCO-MATLAB-based EQL0D procedure [5] are ex-tended here for the U–Pu cycle and chlorides (Figs. 2 and 3).

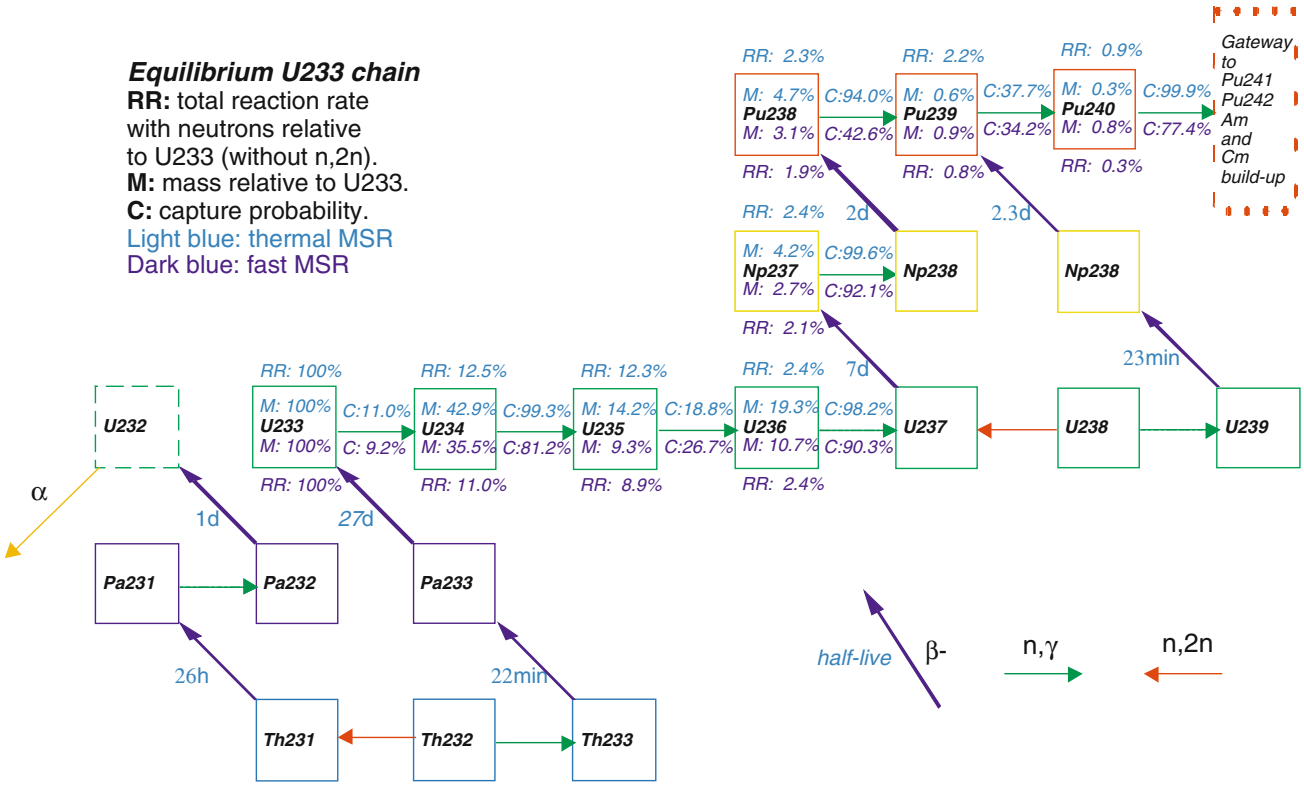

Fig. 1 Evolution of the isotopes under neutron irradiation in the Th–U cycle using fast and thermal spectra MSR reactors with fluoride salts

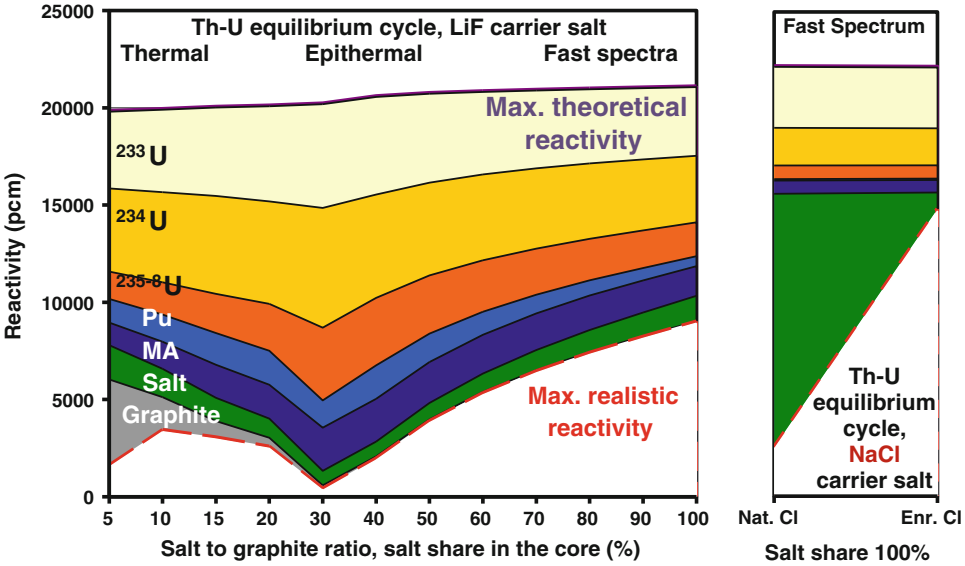

Fig. 2 MSR flexibility in the Th–U cycle using fluoride (*left*) and chloride (*right*) salts; break-down of the infinite-lattice reactivity reduction by the equilibrium fuel composition

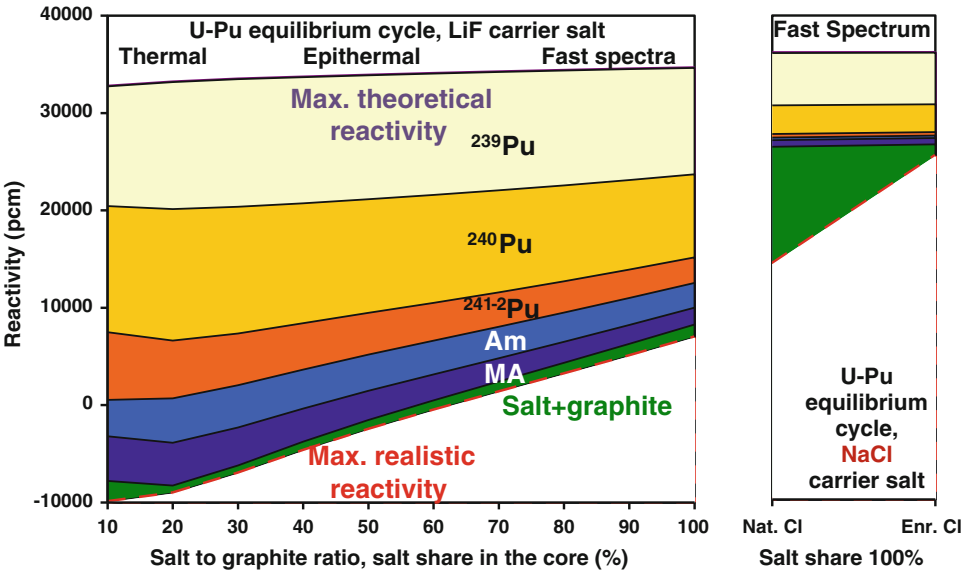

Fig. 3 MSR flexibility in the U–Pu cycle using fluoride (*left*) and chloride (*right*) salts; break-down of the infinite-lattice reactivity reduction by the equilibrium fuel composition

Figures 2 and 3 show the break-down of reactivity reduction by each nuclide from the equilibrium fuel composition, for an infinite lattice. As can be seen, in all cases the maximal realistic reactivity is reached for the fast spectrum. In both the Th–U and U–Pu cycles, enriched ^{37}Cl provides the highest reactivity, thanks to its low absorption and harder spectrum. Natural Cl shows higher reactivity than fluorides in the U–Pu cycle; however, it shows lower reactivity in the Th–U cycle. Accordingly, the hard spectrum of chloride salts is more suitable for the U–Pu cycle [6]. The fluorides are suitable for the Th–U cycle and can be used in both thermal and fast spectra.

No Need for Fabrication

Both fluoride and chloride salts are in liquid form and do not need fabrication. This simple characteristic may help to recycle the U from the Th–U cycle. As can be seen in Fig. 1, the U vector in Th–U cycle includes some amount of ^{232}U. This isotope produces the same decay chain as ^{232}Th, with several hard gamma emitters (e.g., ^{208}Tl); however, its half-life is much shorter. Accordingly, to process any irradiated Th-based fuel will be demanding in terms of radiation protection. The previously mentioned volatilization of mixed fluoride salts may help to overcome this issue,

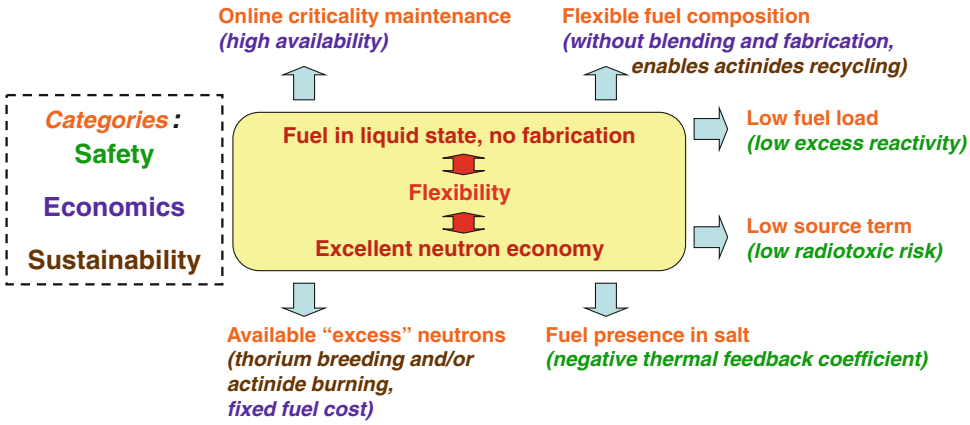

Fig. 4 Flexibility of the MSR fuel cycle and its possible applications

because it is a robust and tested method, which can be operated remotely.

In the U–Pu cycle, the recycling of Pu is not a technological issue and has been industrially mastered. On the other hand, the MAs formed in the U–Pu cycle (Am and Cm) produce considerable decay heat and Cm also emits neutrons. The fabrication of a solid fuel may be, therefore, very complicated. Also in this case, liquid fuel can help to overcome this issue and easily accommodate these actinides.

Flexibility of the Fuel Cycle

Thanks to the combination of excellent neutron economy and of the liquid fuel not requiring fabrication, the MSR fuel cycle can be very flexible. This statement is illustrated in Fig. 4. This flexibility may help overcome historical obstacles for MA burning and ^{233}U breeding related to the heat release and radiation hazards mentioned above. The possibility of on-line refueling and reprocessing might also help the reactor to operate with high availability and virtually eliminate reactivity swings and the need for an initial excess reactivity. If the reactor is to be operated as an iso-breeder with fuel recycling, the fuel cost should be almost independent from the price of the resources. Furthermore, some MSR designs may also enable the recycling of its own or of legacy MA. Accordingly, MSRs have the potential to serve as a sustainable and clean energy source with high utilization of resources and low waste production.

Inherent Safety

MSRs may be designed as inherently safe reactors thanks to the negative temperature feedback coefficients and continuous salt clean-up of highly mobile gaseous and volatile fission products. The removal of gaseous and volatile fission products is a technological requirement, which may simultaneously represent one of the biggest advantages from a safety point of view. Even though it may increase the frequency of small localized incidents, at the same time, it significantly reduces the source term and may act as the strongest pseudo-barrier against the release of fission products during a severe accident. By using the Farmer graph from Fig. 5 as an illustration, it can be seen that the risk related to MSRs with liquid fuel is of a different nature than the risk related to solid fuel reactors.

Any hypothetical severe accident possibly may not cause substantial contamination outside of the power plant as the driving forces, for example, water pressure or chemical reaction of the cladding, are missing in MSRs and as the most mobile fission products are continuously decoupled from the major decay heat source and from the fission chain reaction. This promising qualitative statement will need confirmation by stringent evaluation and quantification of the risks. After several MSR projects dedicated to technological issues and reactor-physics designing, this may be the main direction for further MSR research.

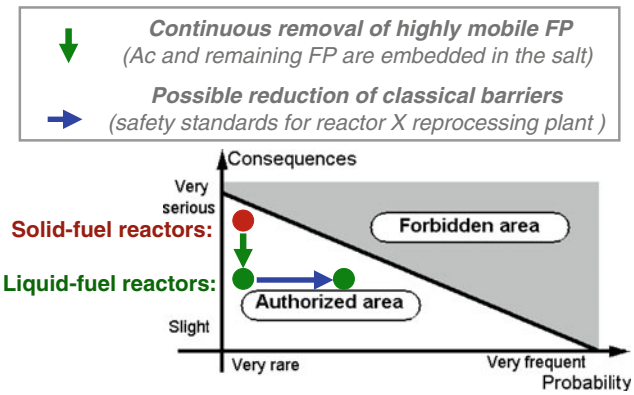

Fig. 5 Relationship between probability and consequences of risks (Farmer graph) illustrated in a log–log scale. In the forbidden area, the product of frequency and consequences is not acceptable (Background chart: http://www.iaea.org/ns/tutorials/regcontrol/assess/assess3212.htm)

Direction of Own MSR Research: Reduction of Challenges

As mentioned in the introduction, MSR advantages may be partly sacrificed to help to reduce the challenges. This is the general idea adopted by many research teams and also in our research, which is currently based mainly on MSc students', PhD students', and post-docs' works. The two main technological challenges are the structural material corrosion and irradiation embrittlement and the demanding molten salt reprocessing techniques.

The mitigation of corrosion of the structural materials will require strict redox potential and impurities control. Nevertheless, the modified Hastelloy-N, proposed at the end of MSR research at the Oak Ridge National Laboratory (ORNL), seems to have been confirmed as a suitable material for long-term operation after recent tests in Russia.

The structural material irradiation embrittlement is related to the fact that the materials compatible with fluoride salts consist mainly of Ni. The natural isotope ^{58}Ni can be transmuted by two predominantly thermal neutron captures to ^{59}Fe and simultaneously produce one He nuclei. This leads to embrittlement of the material. Accordingly, the He-based embrittlement can be reduced by:

1. Finding new resistant materials without ^{58}Ni;
2. Reduction of the thermal fluence to the material;
3. Shortening of the irradiation time.

Reduction of Structural Material Irradiation Embrittlement

One of the ideas to reduce the thermal fluence, is the hybrid spectrum MSR [7]. It is an old idea already applied to the Molten Salt Breeder Reactor (MSBR) at the ORNL. It is based on a two-zone or multi-zone core with one fuel salt, where the moderator determines the criticality in each core zone (Fig. 6). The outer zone is usually subcritical to reduce leakage from the core and, thus, the fluence on the structural materials. The hybrid spectrum MSR is nothing more than an extreme case of this idea, where the small critical thermal zone around the graphite acts as a driver for the big, subcritical, fast zone in the salt. In the pool-type MSR it may also help to localize the fission reaction in the center.

The idea of a hybrid spectrum MSR was preliminarily evaluated for a simple axially infinite reactor design from Fig. 7 (left) and the resulting radial and spectral flux distribution can be seen in Fig. 7 (right). This figure also justifies the title "hybrid spectrum" MSR. The initial results are promising. Nevertheless, the fission products, which are not accounted for in the presented results, may completely change the conclusion about the advantages and disadvantages of the hybrid spectrum MSR. Another solution to prevent irradiation embrittlement may be the shortening of irradiation time. This solution can be applied to the two-fluid MSR (Fig. 8), where the wall between the salts is exposed to a high fluence. The reactor should then be designed to allow simple exchange of the wall every few years, at best, together with the fuel salt.

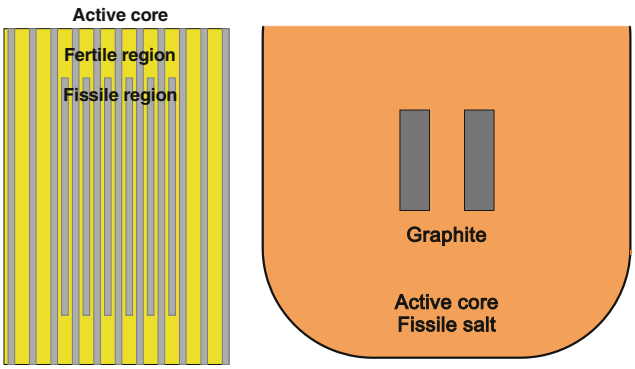

Fig. 6 Illustration of the two-zone MSR core in the thermal case (*left*) and in the extreme hybrid spectrum case (*right*). (*Yellow* or *orange* corresponds to salt, *gray* to graphite)

Simplification of the Reprocessing Strategy in the Th–U Cycle

The ultimate aim of almost every fuel reprocessing is to separate the fission products and recycle, at best, everything else. The irradiated MSR fuel consists of:

1. Carrier salt (LiF, LiF–BeF_2, NaF–BeF_2, etc.);
2. Fertile actinides (^{232}Th);
3. Fuel (U vector);
4. By-products (Np and Pu);
5. Fission products.

From a sustainability point of view, in the Th–U cycle it may be important to recycle the main fertile element, Th, and possibly also some rare elements (Li, Be).

From an economical point of view, it may be interesting to recycle all components. Nevertheless, it will depend on their price and on the reprocessing costs. In some cases their direct disposal, for example, by vitrification, may be cheaper.

From a reactor physics point of view, it is important to recycle U as the main fissile element of the Th–U cycle; the other components are not substantive.

The fluorides foreseen for MSRs have one important feature. Some elements, such as U, Np, and to certain extent Pu, form volatile hexafluorides and can be separated/recycled

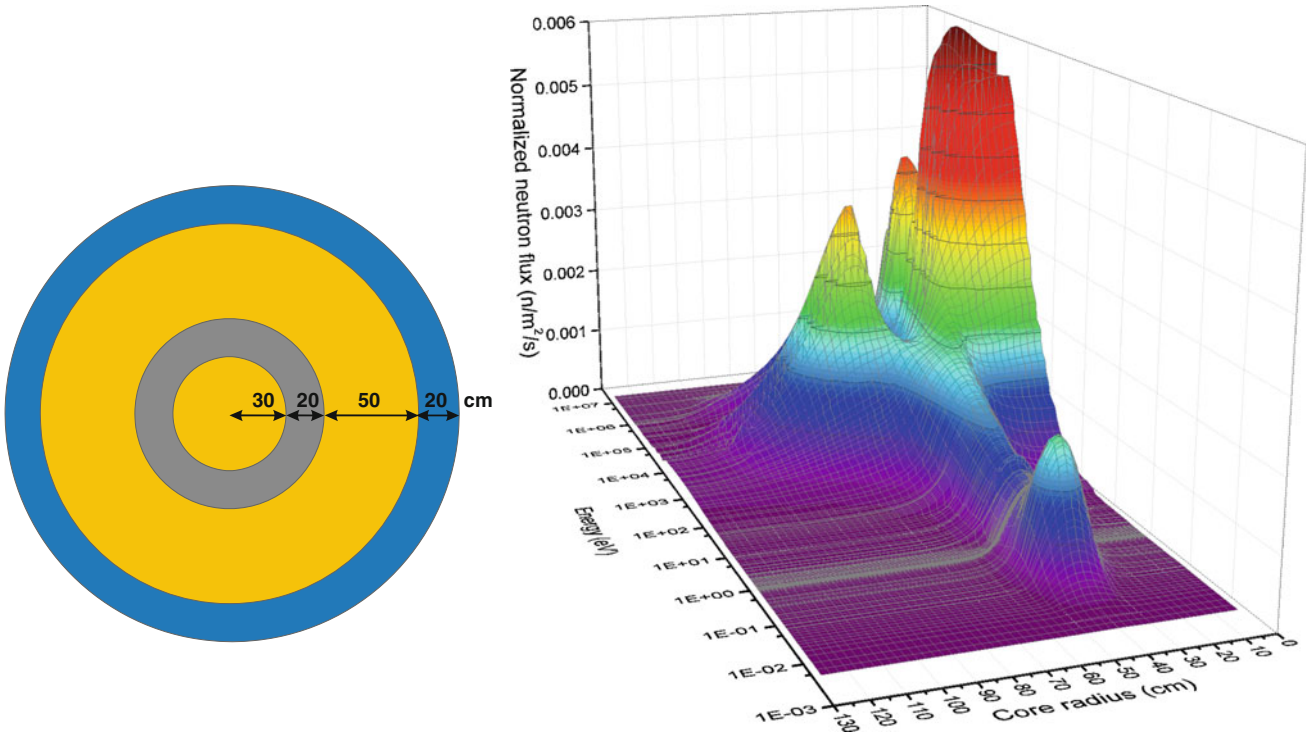

Fig. 7 Simple axially infinite reactor design (*left yellow* = salt, *gray* = graphite, *blue* = hastelloy) and the resulting distribution of the normalized neutron flux (*right*) in the energy spectrum and space in the radial direction) in the hybrid spectrum MSR

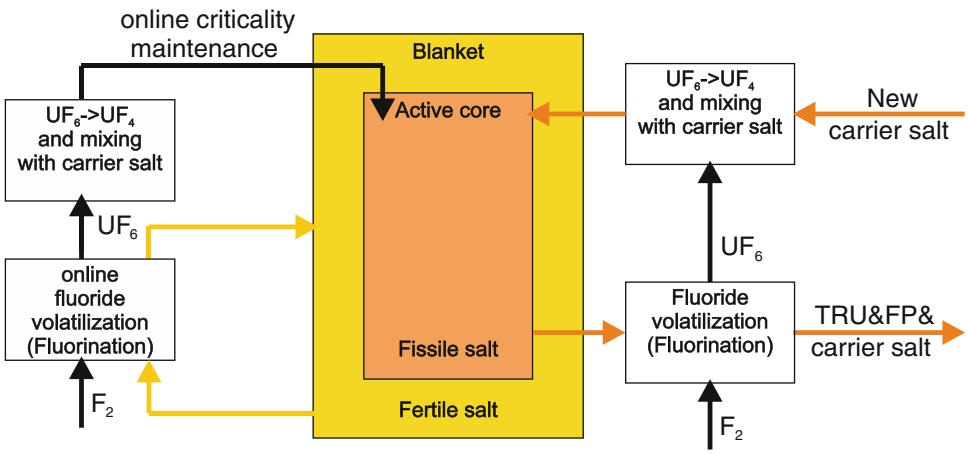

Fig. 8 Simplified in situ fuel recycling by the volatilization in the two-fluid MSR. The remaining carrier salt can be directly disposed or reprocessed later and probably ex situ

by the volatilization technique or fluorination. The volatilization of fluoride salt mixtures is a robust and tested reprocessing method, which was patented in the US as early as 1958 (patent nr. US 2833617 A). It enables relatively simple separation of the selected elements from the irradiated carrier salt. As it can be remotely operated, it is also suitable for ^{232}U recycling. As mentioned earlier, this isotope has in its decay chain several hard gamma emitters, namely, ^{208}Tl and ^{212}Bi. The idea of simplified reprocessing is illustrated in Fig. 8. It is based on in situ fuel recycling by volatilization and delayed, and probably ex situ, carrier salt clean-up or its direct disposal by vitrification [8].

The need to postpone the salt treatment may be also based on the radiological protection of the reprocessing facility. The activity of the fission products and of the ^{232}U decay chain left in the salt may require storage time from 10–20 years. ^{228}Th is a direct product of ^{232}U decay and has a half-life of 1.9 years; the other nuclides in the chain decay fast, for example, ^{208}Tl and ^{212}Bi; thus, the activity and the required storing time may be determined by the 1.9-year half-life of ^{228}Th (Fig. 9).

The volatilization technique has a feature that may make separation of the elements easier; that is, that as a result of the chemical potentials, UF_6 is removed from the salt mixture

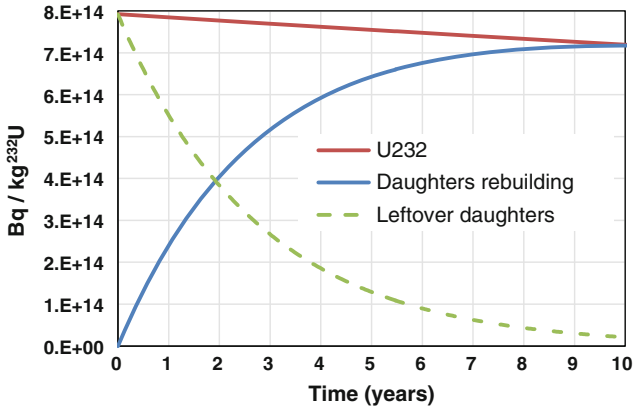

Fig. 9 Activity evolution for the separated ^{232}U and its daughter products in the *leftover* salt

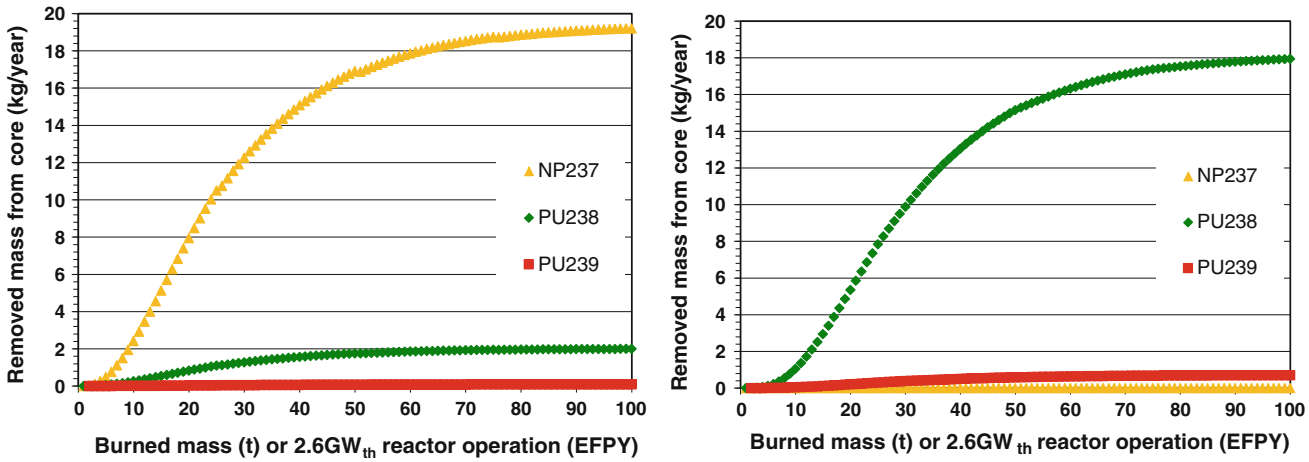

Fig. 10 Yearly removed amount of actinides from a 2600 GW_{th} fast MSR reactor in the cases of U (*left*) or U and Np (*right*) recycling by volatilization

first, followed by NpF_6 and PuF_6. Accordingly, which mixture of the three elements will be separated can be selected by the duration of the volatilization process. Furthermore, U and Np can be removed from the salt with very high efficiency, whereas Pu only to a certain extent. Two ideal scenarios were simulated here: (1) removal of 100 % U only and (2) 100 % U + Np separation by volatilization. The results in Fig. 10 show the mass of other actinides left in the irradiated salt after separation. In both scenarios, the mass will stay below 20 kg/year for a 2600 GW_{th} fast MSR reactor. However, the actinide composition will strongly depend on the recycling strategy [9]. In the case of U recycling, the main actinide left in the irradiated salt will be ^{237}Np (Fig. 10, left); in the case of U and Np recycling, the main actinide left in the irradiated salt will be ^{238}Pu (Fig. 10, right). As the neutronic importance of these two nuclides is small, other criteria may be used for recycling strategy selection.

Conclusion

It can be concluded that MSRs belong to a class of prospective advanced technologies that can help to cover future energy demand, minimize the hazards of severe accidents, and reduce the amount of radioactive waste produced. Their appealing properties may possibly enable design of an inherently safe reactor with a flexible fuel cycle and represent a strong motivation to overcome the difficulties related to structural material corrosion and chemical technology.

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Part V
Invited Speakers: Thorium-Reactor Physics

Nuclear Data Development Related to the Th–U Fuel Cycle in China

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Abstract

In recent years, the Th–U fuel cycle, which is regarded as a supplement to the U–Pu fuel cycle, has caught the nuclear industry's attention. Therefore, to meet the requirements of neutronics studies, nuclear data research related to the Th–U fuel cycle has been carried out in China since 2010. Fission product yields of ^{232}Th induced by 14 MeV neutrons had been measured at the China Institute of Atomic Energy (CIAE). In addition to nuclear data evaluation, evaluation of the fission product yields of ^{232}Th was also accomplished. A complete set of neutron-induced reaction data for ^{233}U and ^{232}Th has been evaluated based on feedback from benchmark testing. Thermal and intermediate spectra benchmark testing results show that the resonance data of ^{233}U and ^{232}Th still needs to be improved.

Introduction

In recent years, the Th–U fuel cycle, which is regarded as a supplement to the U–Pu fuel cycle, has drawn the nuclear community's attention. To meet the requirements, limited research activities have been carried out in China since 2010, including:

- measurement of fission product yields for ^{232}Th as induced by 14 MeV neutrons;
- evaluation of neutron-induced fission product yields (nFPY) for ^{232}Th;
- evaluation of the nuclear reaction data for n + ^{233}U, n + ^{232}Th;
- validation of the latest evaluations for n + ^{233}U, n + ^{232}Th with criticality benchmarks, and so on.

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Measurement of nFPYs for ^{232}Th

To clarify the discrepancies that existed in current evaluations of the nFPYs for ^{232}Th, the nFPYs for ^{232}Th induced by 14 MeV neutrons were measured by the direct γ spectrum method. After corrections of γ-ray absorption in foils, low-energy neutrons, and independent yield, the yields of 46 chains were obtained. All mass yields were normalized by 200 %, of which measured yields summed up to 138 %. In the mass regions of $80 \leq A \leq 100$ and $130 \leq A \leq 150$, the relative uncertainties are less than 5 % for most of the products measured. The comparison of the measured chain yields with the evaluated ones from ENDF/B-VII.0 [1], JEFF-3.1.1 [1], and JENDL-3.3 [2] is shown in Fig. 1. Differences larger than 20 % between the measured and the evaluated are found for mass chains A = 84, 113, 119, 130, 134, 151, and 152.

Evaluation of nFPYs for ^{232}Th

The nFPYs and the mass distributions of ^{232}Th induced by fission and 14 MeV neutrons, were evaluated though comparison of B-VII.0, JEFF-3.1.1, JENDL-3.3, and experimental data. The evaluation work mainly focused on ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{147,149,150,151,152}Sm, ^{143,145}Nd, and ^{153}Eu, all of which are interesting for burnup

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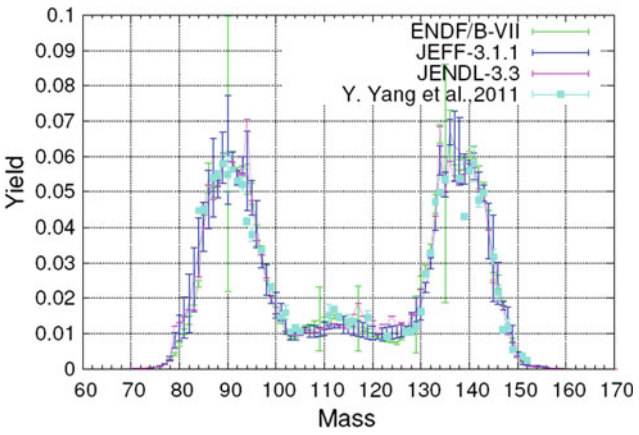

Fig. 1 Comparison of the new measured fission product chain yields

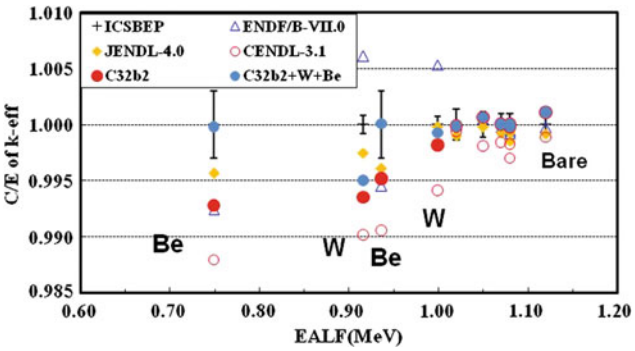

Fig. 3 Comparison of C/E values (calculated over experimental) of k_{eff} for the UMF system

calculations. The final recommendation is a combination of B-VII.0 and JEFF-3.1.1. For example, JEFF-3.1.1 is recommended for the cumulative yield of ^{133}Cs; whereas, for ^{152}Sm, B-VII.0 is recommended. Additionally, the yields of ^{152}Sm and ^{153}Eu, as well as the yields on the same chains, were adjusted.

Re-evaluation of Nuclear Reaction Data for ^{233}U and ^{232}Th

The nuclear reaction data for ^{233}U were re-evaluated based on CENDL-3.1 [3]. According to feedback from ^{233}U benchmark testing [4], (n, el), (n, inl) cross sections and angular distributions have been revised. Figure 2 shows the comparison of the original and revised (denoted as C32b2) (n, inl) cross sections. B-VII.0, JENDL-4.0 [5], and JEFF-3.1.1 are also compared in Fig. 2. As shown in Fig. 3, for the Be- and W-reflected UMF (^{233}U-Metal-Fast) systems, the calculated k_{eff} with CENDL-3.1 was improved significantly when C32b2 was used instead. If the data for Be and W was replaced with CENDL-2.1 [6] and ENDF/B-VII.1 [7], respectively, further improvements were obtained (Fig. 3).

The nuclear reaction data for ^{232}Th were re-evaluated based on CENDL-3.1. As shown in Fig. 5, over-predictions of k_{eff} of more than 2000 pcm were found by CENDL-3.1 in the fast and intermediate spectra benchmarks, which is caused by under estimation of (n, γ) cross sections from 4 keV to 1 MeV. Therefore, in Fig. 4, the experimental data [8–13] in the unresolved resonance region were reviewed and the data measured by Aerts et al. [13] at CERN/n_TOF (neutron time-of-flight facility) were adopted. The benchmark results based on C32b1 have been significantly improved by replacing the ^{232}Th data.

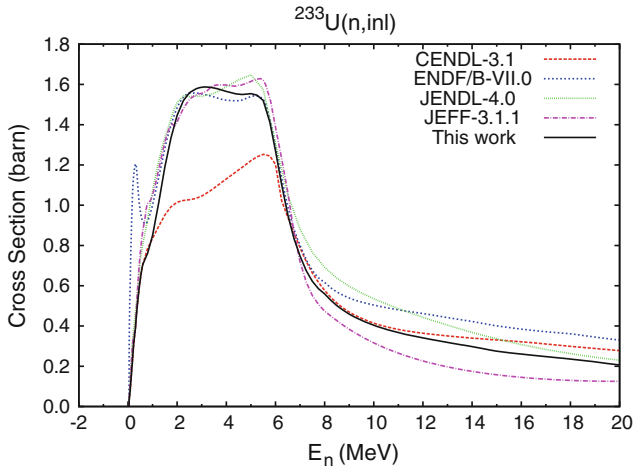

Fig. 2 Comparison of ^{233}U(n, inl) cross sections

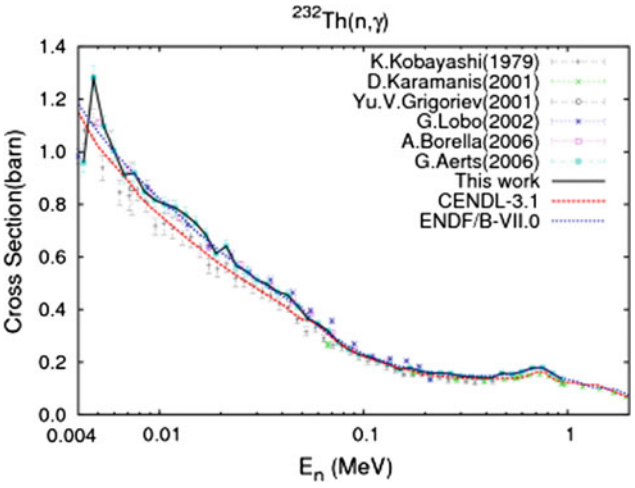

Fig. 4 Comparison of ^{232}Th(n, γ) cross section data

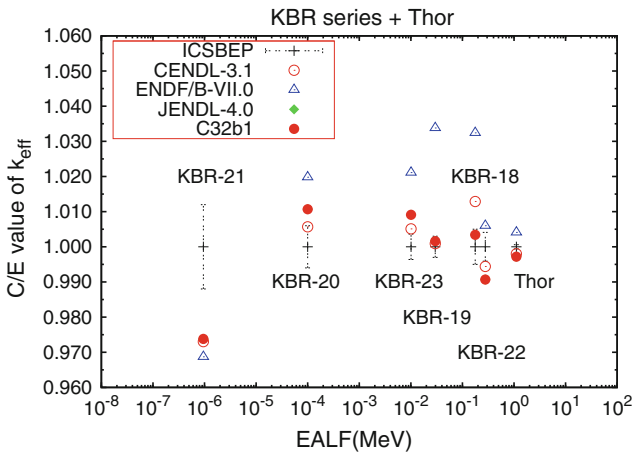

Fig. 5 Comparison of C/E values for the KBR and Thor reactors

Nuclear Data Validation for ^{233}U and ^{232}Th

To improve the neutron reaction data for ^{232}Th and, the evaluations of CENDL-3.1, B-VII.0, JENDL-3.3, and 4.0 were tested with criticality benchmarks from International Criticality Safety Benchmark Evaluation Project (ICSBEP) [14]. The criticality calculations were done with the MCNP5 code [15]. The ACE libraries used in the calculations were prepared by using the NJOY99 code [16]. Significant under-predictions of k_{eff} in relation to the spectra index in the USI (^{233}U-Solution-Inter-001) system were observed (Fig. 6). Bias against the spectra index of all libraries was also found in thermal benchmark results for ^{232}Th. The ^{233}U (n, f), ^{233}U (n, γ), and ^{232}Th (n, γ) cross sections in thermal and resolved resonance regions are thought to be improved by sensitivity analysis.

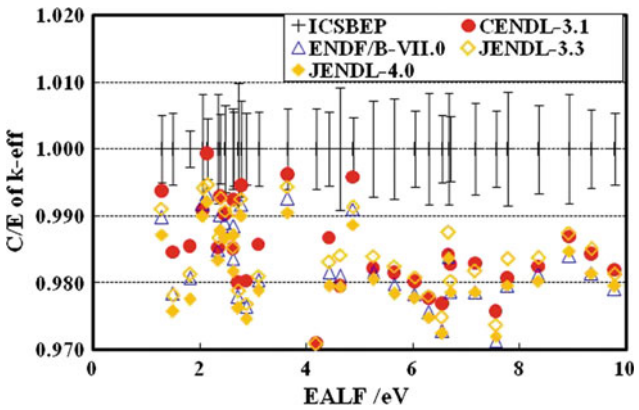

Fig. 6 C/E values of k_{eff} for the USI system

Summary

To improve the nuclear data related to the Th–U fuel cycle, some key data have been measured or re-evaluated in China. The nFPYs for ^{232}Th were measured and evaluated; discrepancies larger than 20 % were found. Based on the feedback from benchmark testing and new experimental data, nuclear reaction data for ^{232}Th and ^{233}U in CENDL were revised and show improved performance in UMF and KBR benchmarks. However, for thermal and intermediate spectra system, k_{eff} biases against the spectra index were found. Resonance parameters of ^{233}U and ^{232}Th are the data that need to be improved in the future.

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Nuclear Data Development Related to the Th–U Fuel Cycle in India

S. Ganesan

Abstract

This paper aims to share the excitement, challenges, and progress in the indigenous Indian efforts in nuclear data science in a generic context, in particular, towards the utilization of the thorium fuel cycle. To meet all the nuclear data needs in India for energy and non-energy applications, a Nuclear Data Physics Centre of India (NDPCI) was successfully formed in 2009. The NDPCI operates in a virtual mode at this time. Efforts are underway to create a sustainable physical center for the NDPCI. This paper also presents the interesting results of calculations that show a highly positive temperature coefficient for the BeO reflector in the KAMINI reactor, the only operating ^{233}U-fuelled reactor in the world.

Introduction

The nuclear data needs for the thorium fuel cycle are not only for the main fissile and fertile isotopes of thorium fuels, such as those that have already been discussed [1] (viz., ^{230}Th, ^{232}Th, ^{231}Pa, ^{233}Pa, ^{232}U, ^{233}U, and ^{234}U), but also inevitably include all the range of actinides for multiple fuels, those of coolants, structural materials, fission products, and minor actinides that are associated with the use of thorium-fueled reactors.

The Indian approach to nuclear energy, including committed efforts towards thorium utilization, involves the development of innovative reactor designs by using a multiphysics, multiscale modelling approach. India is energy-hungry, wanting to increase her human development index with her huge population, which is presently over 1.28 billion. Thorium fuel has a great potential to serve as a significant source of low-carbon electricity in India as part of a viable energy mix of options designed to last for several centuries. For the interested reader, a broad overview on current Indian perspectives on using thorium is nicely described in the contributions by Vijayan et al. [6], Wattal et al. [7], and Degweker et al. [8] in this ThEC13 proceedings. The official websites of the Department of Atomic Energy (DAE) [2] presents, to the interested reader, all the details of various on-going activities of the Bhabha Atomic Research Centre (BARC) three-stage program that, by design, inevitably involves closed fuel cycles with multiple fuels (U–Pu and Th–U) and with long burnup. India recognizes that nuclear data science is an essential base technology effort that forms part of the big data science in meeting all the Indian nuclear data needs for thermal, fast, fission–fusion, and accelerator-driven subcritical systems [3–5], which involve multiple fuel cycles with closed fuel cycles. It is recognized [3–5] that long-term nuclear data needs of closed fuel cycles with multiple fuels (U–Pu and Th–U), with high burnup, are demanding in the Indian context of the BARC three-stage nuclear power program.

Thorium Utilization Studies Need New and Improved Nuclear Data

The author believes that the best nuclear reactor design—one that would not be even remotely accident prone during its entire fuel cycle, that would create the minimum radioactive waste, would be fully proliferation resistant, would have the maximum tolerance of normal (and even remotely possible operator errors), man-made, and natural disasters—is yet to be made. Research associated with evolving new nuclear

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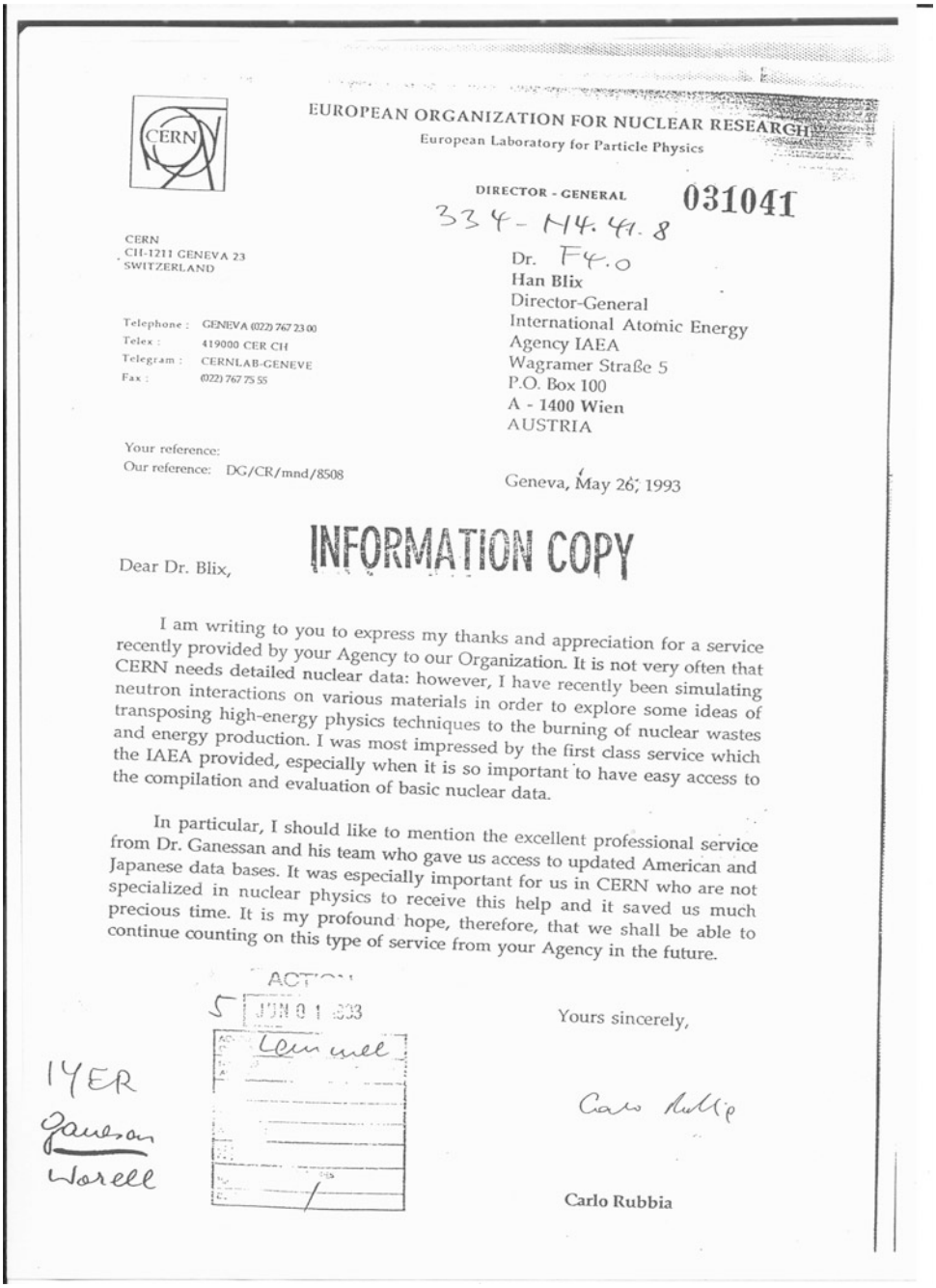
energy systems using thorium within the context of the BARC three-stage program needs a significant amount of new and improved nuclear data, both at the differential and integral levels. The role of multiphysics multiscale modeling (MMM3) with "big" data science, including detailed nuclear data, in attempts to evolve the best reactor design cannot be over emphasized.

The letter from Prof. Carlo Rubbia, CERN, to Dr. Hans Blix, International Atomic Energy Agency (IAEA), in 1993 (Fig. 1) is an apt illustration of the fact that the strong need for basic nuclear data is automatically and naturally felt when serious studies of new physics designs (here, in the case of the Energy Amplifier by Carlo Rubbia and his team for utilizing thorium) are performed.



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Your reference:
Our reference: DG/CR/mnd/8508

Geneva, May 26, 1993

INFORMATION COPY

Dear Dr. Blix,

I am writing to you to express my thanks and appreciation for a service recently provided by your Agency to our Organization. It is not very often that CERN needs detailed nuclear data: however, I have recently been simulating neutron interactions on various materials in order to explore some ideas of transposing high-energy physics techniques to the burning of nuclear wastes and energy production. I was most impressed by the first class service which the IAEA provided, especially when it is so important to have easy access to the compilation and evaluation of basic nuclear data.

In particular, I should like to mention the excellent professional service from Dr. Ganessan and his team who gave us access to updated American and Japanese data bases. It was especially important for us in CERN who are not specialized in nuclear physics to receive this help and it saved us much precious time. It is my profound hope, therefore, that we shall be able to continue counting on this type of service from your Agency in the future.

ACTION
5 JUN 01 1993

Yours sincerely,

Carlo Rubbia

Carlo Rubbia

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Fig. 1 Letter from Prof. Carlo Rubbia on nuclear data needs for the Energy Amplifier

Initial Indian Efforts at Kalpakkam on Nuclear Data for the Thorium Fuel Cycle

The author was involved in the nuclear data science efforts in India as early as the seventies and eighties, when a detailed look at some of the data statuses of the thorium fuel cycle was attempted. For instance, the evaluation of unresolved resonance parameters for ^{232}Th and ^{233}U was attempted [9, 10] in Kalpakkam in the late seventies. Attempts were also made [11] from the seventies onwards to create Indian processing code systems to use the freely available basic evaluated nuclear data libraries. Partial nuclear data evaluations were also attempted as the first exercise and India's first basic evaluated nuclear data file in ENDF/B format was on the ^{232}Th isotope, which provided evaluated nuclear data [12] above the resolved resonance region under the IAEA Nuclear Data Library (INDL) Project in the early eighties. The interesting fast reactor sensitivity study performed at Kalpakkam [13] in the late eighties by using different data sets for ^{232}Th and ^{233}U illustrated that the doubling time of a conceptual thorium-fueled fast reactor changed by a factor of nearly two owing to use of different nuclear data sets by changing only the data for ^{232}Th and ^{233}U and leaving the nuclear data of all other isotopes unaltered.

Nuclear Data Physics Experiments for the Thorium Fuel Cycle

The nuclear data available in the IAEA EXFOR database [14] clearly indicates increased experimental nuclear data activities in India in the area of nuclear data measurements in recent years. Due to space constraints, the detailed EXFOR entries and discussion of nuclear data, as well as graphs of nuclear data related to the thorium fuel cycle that have been generated in India will not be reproduced here, however, they can be found in [14].

Surrogate techniques are being employed [15, 17–19] by BARC to measure neutron-induced nuclear reaction data of unstable actinide nuclei. Consider the situation when we do not have an unstable target and also do not have a neutron beam: how can we get the neutron-induced cross sections in such cases? Indian nuclear physicists have been specializing in complex heavy-ion nuclear fusion reactions and associated multi-parameter measurements. This in-house, specific expertise helped BARC to enter the area of surrogate methodologies to generate neutron-induced nuclear reaction data. BARC has successfully obtained [15, 17–19], experimentally, by using surrogate nuclear techniques, the neutron-induced fission cross-section data of unstable minor actinides such as ^{233}Pa ($t_{1/2}$ = 26.975 ± 0.013 days), ^{234}Pa ($t_{1/2}$ = 6.70 ± 0.05 h), ^{239}Np ($t_{1/2}$ = 2.356 ± 0.003 days), ^{240}Np ($t_{1/2}$ = 61.9 ± 0.2 min), and ^{241}Pu ($t_{1/2}$ = 14.325 ± 0.006 years). In all these efforts [15, 17–19], nuclear–model-based predictions using standard nuclear model codes, such as TALYS and EMPIRE, are also being performed.

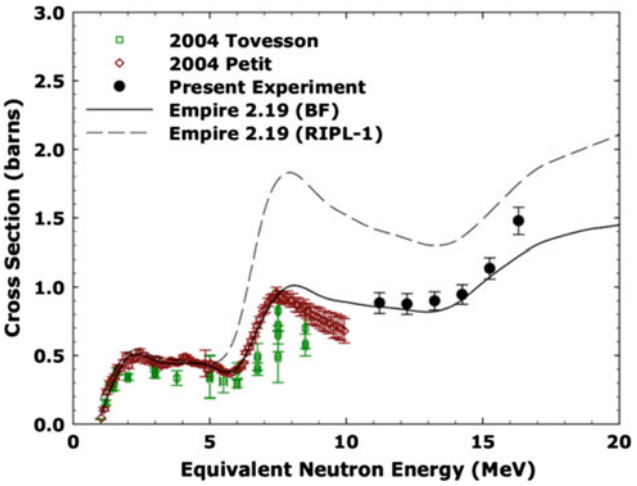

Fig. 2 Experimental ^{233}Pa(n, f) cross section along with EMPIRE-2.19 predictions [15]

The new hybrid surrogate ratio method (HSRM) approach [15] is being used to determine the ^{233}Pa(n, f) cross section in the excitation energy range of 11.5–16 MeV (Fig. 2).

In this BARC hybrid approach, a weakly bound projectile nucleus, ^{7}Li, was employed as a beam hitting a single target of ^{232}Th. We produce, thereby, in situ, the two nuclei, ^{234}Pa and ^{236}U, by two different direct reactions:

$$^{232}\mathrm{Th}(^{6}\mathrm{Li},\alpha) \rightarrow {}^{234}\mathrm{Pa} \text{ and } {}^{232}\mathrm{Th}(^{6}\mathrm{Li},\mathrm{d}) \rightarrow {}^{236}\mathrm{U}$$

The ^{234}Pa and ^{236}U nuclei are treated as surrogates for the reactions:

$$\mathrm{n} + {}^{233}\mathrm{Pa} \rightarrow {}^{234}\mathrm{Pa} \text{ and } \mathrm{n} + {}^{235}\mathrm{U} \rightarrow {}^{236}\mathrm{U}$$

The present BARC measurement [15] is unique in that the two residues are formed with an overlapping excitation energy spectrum. In the HSRM, the same target is used to populate the two element systems and, thus, by taking a ratio of the two reaction rates on the same target, systematic uncertainties due to target thickness, beam current, and dead time in the determination of the ratio of fission decay probabilities corresponding to 'desired' and 'reference/monitor' reactions, are eliminated. The EXFOR entry for this surrogate measurement of the ^{233}Pa(n, f) cross section [15] was completed. The EXFOR entry numbers 33,023 and D6075 for the above-mentioned data took into account two inadvertent errors in the analysis and publication. One error was taking the ratio of 233/234 as unity and the other was the usage of an outdated value of neutron separation energy for ^{234}Pa. The corrected plots from the EXFOR database [16] are comparable with the uncorrected versions, as shown in Fig. 3.

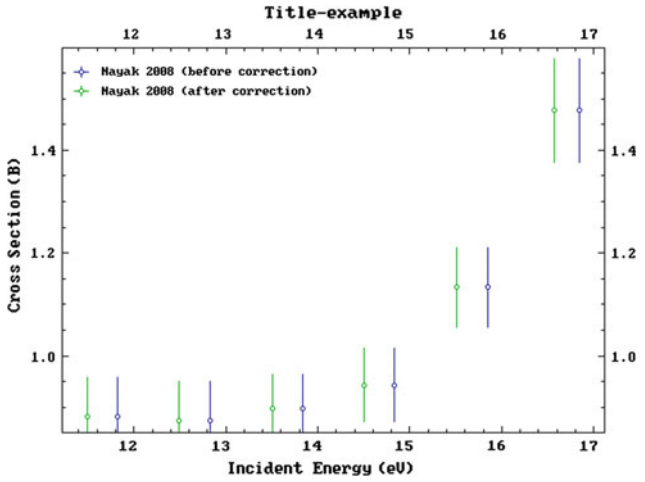

Fig. 3 The ^{233}Pa(n, f) cross section as a function of the equivalent neutron energy. The publication [15] had two inadvertent errors: *1* Skipping of center of mass to laboratory transformation and *2* adoption of 5.45 MeV as the Sn(^{234}Pa) value

The BARC team has also employed HSRM to determine the ^{239}Np(n, f) and ^{240}Np(n, f) cross sections in the equivalent neutron energy range 10.0–16.0 MeV [17] by using ^{7}Li +^{232}Th and ^{6,7}Li+^{238}U transfer–fission coincidence measurements. The ^{241}Pu(n, f) cross sections have been determined [18] by the surrogate ratio method in the equivalent neutron energy range 11.0–16.0 MeV by using ^{238}U(^{6}Li, d) → ^{242}Pu and ^{232}Th(^{6}Li, d) → ^{236}U transfer reactions at E_{lab} = 39.6 and 39.0 MeV, respectively. The BARC team has also carried out [19] measurements of the ^{234}Pa(n, f) cross section in the MeV energy region by employing the respective reactions, ^{232}Th (^{7}Li, α) → ^{235}Pa and ^{232}Th(^{7}Li, t) → ^{236}U. Generating the covariance error matrix for nuclear data obtained in such surrogate-based approaches is desirable.

In the thermal region, the ^{234}Pa(n, f) cross section was deduced experimentally [20] in the form of the ^{233}Pa(2nth, f) cross section by using a fission track technique by irradiation of ^{233}Pa in the APSARA thermal reactor. The cross section value was found to be 4834 ± 57 b. The ^{233}Pa(2nth, f) cross section was calculated theoretically by using the TALYS computer code and was found to be in good agreement with the experimental value after normalization with respect to ^{241}Am(2nth, f). It may be noted that ^{234}Pa has a half-life of 6.7 h. Of course, it remains to be determined under what neutron flux conditions reactions with large cross sections become important. Additionally, whether such reactions play a role in practical applications, for instance, in exotic thorium systems with unusually high neutron flux and transients, also needs to be investigated.

The cross sections and fission yield data of actinides were measured by BARC teams by using activation methods with neutrons from the spectrum of neutrons arising from ^{7}Li(p, n) reactions, in D-T (14 MeV) neutron facilities. Photon–nuclear reaction data using Bremsstrahlung gamma sources with electron accelerators have been experimentally generated for a range of targets and electron energies. An illustrative reference list of recently performed nuclear data physics experiments is presented in [15, 17–26].

Mirror Websites for Nuclear Data

The online nuclear data services (NDS) at BARC (http://www-nds.indcentre.org.in/) mirrors the nuclear data website of the Nuclear Data Section of the IAEA, Vienna (http://www-nds.iaea.org). Under this arrangement, online updating is performed every 12 h in the mirror with the IAEA website through a 2 MB direct link. The server is being maintained, with manpower and machinery, by the BARC Computer Division. It offers faster downloads. The India mirror site is getting increased usage.

Indian EXFOR Compilations of Nuclear Data

India became a member of the international nuclear reaction data compilation network (NRDC) by invitation in 2008. India has thus far contributed more than 264 EXFOR entries as accepted by the IAEA [27] at the time of writing. Introduction of the EXFOR culture [28] to people in India, including those in the area of basic nuclear physics, has become a relatively simple task with the new managerial initiatives of the NDPCI, which have included holding EXFOR workshops in India. Indian EXFOR compilation workshops in nuclear data compilations have become a role model [29] and have become a useful value addition to the nuclear physics community in India, providing greater visibility for the experimental measurements and research work done by India. The Indian EXFOR workshops have been phenomenally successful in a systemic sense, with the incentives towards sustainable viability arising from the demographic advantages in India. The importance of such highly focused training courses on EXFOR is well recognized by the BARC scientific community and is deliberately kept separate from the national nuclear physics conferences. The NDPCI is trying to form a permanent EXFOR compilation team at BARC. The NDPCI is responsible for all EXFOR compilations of experimental nuclear physics data generated in and published by nuclear physics facilities [28].

IAEA Coordinated Research Projects (CRPs)

India actively participated in and benefitted greatly from the IAEA CRP on "Evaluated Data for the Thorium–Uranium Fuel Cycle," 2003–2006 [30]. Under the scope of this CRP, India contributed, for instance, the KAMINI benchmark [31] that was published in the International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook. For more information on ICSBEP benchmarks, see: http://icsbep.inl.gov/.

Interestingly, under the scope of the IAEA WIMS library update project (WLUP) [32], improved nuclear data for thorium nuclide chains became available for the users of the WIMSD lattice cell code. The Indian reactor physics community use the updated WIMSD nuclear data libraries [32] provided by the IAEA in all design calculations. Using the XnWLUP software [33], we generated over 100,000 inter-comparison plots of multigroup data sets (including, number of nuclides, partial reactions, evaluated files, ratio of discrepancies, zoomed portions in energies) in WIMSD format to have a look at the discrepancies in nuclear data for various isotopes. The KAPS-1 over-power transient could be explained [34] only with the use of the new WLUP libraries. The safety and operational requirements of power plants are engineered with a number of one-to-one mock-up experiments providing adequate and conservative safety margins. However, the KAPS-1 over-power transient incident sent out a strong signal to the Indian nuclear community that in general, whether the U–Pu or Th–U fuel cycle is used, the design manuals of operating nuclear systems should be updated every few years as and when improved nuclear data becomes available. Indian scientists are aware of the limitations of the use of WIMSD conventions in advanced reactor designs. For instance, the fission spectra effect in WIMSD conventions owing to fissions in isotopes of the thorium fuel cycle was studied in a recent research paper [35].

Analyses of Irradiation of Thorium Bundles in PHWRs

The Indian post-irradiation experiment (PIE) of thorium [3, 31, 36] spanned two decades and involved hundreds of experts, as is the case with such PIE experiments associated with operating reactors. Identical loading of thorium bundles [36] was used in the 220 MW_e power stations, KAPP-1&2, KAIGA-1&2, and RAPS-3&4, to attain flux flattening in the initial core. The thorium oxide in all the 35 bundles put together in a reactor is about 400 kg. The bundles loaded in KAPP-1&2, KAIGA-1&2, and RAPS-3&4 went through over 500 days of full power operation, amounting to about 12,000 MWd/tonne burnup and these bundles have already been discharged from the core. After a cooling period of five

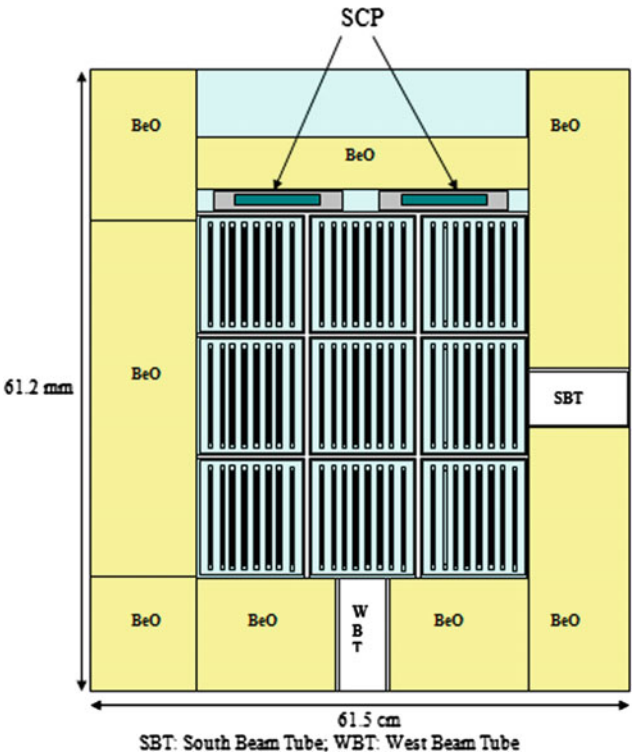

Fig. 4 Horizontal cross-section view of KAMINI core–reflector assembly. (*After* http://www.igcar.ernet.in/benchmark/science/26-sci.pdf). See [39] for full specifications as used in the calculations

years, samples were obtained from one of the irradiated ThO_2 bundles and have been analyzed experimentally by alpha spectrometry for ^{232}U and by thermal ionization mass spectrometry for ^{233}U, ^{234}U, ^{235}U, and ^{236}U by two different groups at BARC. The previous analyses by the two teams at BARC gave an under-prediction factor of 6–8 for the production of ^{232}U. The discrepancy was resolved and traced to the fact that the nuclear data of effective one-group values for cross sections of isotopes of the thorium fuel cycle and the use of assumptions (e.g., self-shielding) in the ORIGEN code package are applicable to traces of thorium in natural uranium rods, but not for the irradiation of bulk thorium rods used for power flattening in our pressurized heavy water reactors (PHWRs). Such discrepancies in the estimation of ^{232}U are not acceptable as it leads to large errors [37] in shielding back-end facilities for the thorium fuel cycle. As a general remark, it may be noted that the $^{232}Th(n, 2n)$ reactions that occur above 6.5 MeV are the main contributor to the production of ^{232}U, in this case of thorium loading in PHWRs. We should note that it is a challenge to characterize the uncertainty in such threshold activation rates in thermal reactors as the fraction of neutron flux above 6 MeV calculated by lattice cell codes in the super cell model, is in the range of 0.05 % and the flux above 6.5 MeV is further very uncertain as the fission neutron spectrum, that is, the amount of virgin fission neutrons above 6.5 MeV, have large

Table 1 Calculated reflector BeO temperature dependence of k_{eff} of KAMINI

Temperature (K)	Density (g/cc)	k_{eff}	Std. deviation	Δk
293	2.9299	0.99007	0.00025	0
600	2.9058	1.00131	0.00025	0.01124 (11.2 mk)
800	2.8903	1.00648	0.00025	0.01641 (16.4 mk)
1200	2.8598	1.01385	0.00025	0.02378 (23.8 mk)

uncertainties to begin with. The on-going IAEA CRP on improving nuclear data of prompt fission neutron spectra [38] is important also in this perspective.

Positive Temperature Coefficient of the Reflector in KAMINI

The KAlpakkam MINI (KAMINI) reactor is a ^{233}U-fueled, light water moderated, natural convection cooled, beryllium oxide reflected, zero power research reactor (Fig. 4). The-KAMINI reactor is located at the Indira Gandhi Centre for Atomic Research at Kalpakkam, India, and is the only operating ^{233}U-fueled reactor in the world. Because of the highly efficient reflector material, BeO, it has a very low fuel inventory ($\sim$612 g). The reactor is designed to operate at a nominal power of 30 kW. The built-in maximum excess reactivity of the reactor is restricted to 300 pcm (3 millik since 1 mk = 0.001 = 100 pcm), and this reactivity is apportioned to maintain various effects, such as coolant temperature load, irradiation sample load, operating margin, xenon load, and long-term burnup losses.

India contributed KAMINI [39] to the ICSBEP handbook in 2005. We used the ICSBEP specifications [39] for the KAMINI calculations. In the calculations, we set the reflector at various higher temperatures, keeping the temperature of the core part at room temperature. We briefly present the results of these calculations in Table 1. The reactivity jumps by several dollars when only the reflector is heated, while keeping the temperature of the core unchanged. Our calculations show a sizable positive temperature coefficient of the BeO reflector in KAMINI reactor at Kalpakkam.

We understand why the reflectivity worth goes up with temperature in the case of KAMINI. KAMINI is a highly reflected system. The use of massive BeO as a reflector in KAMINI contributes as much as 0.45 to the k_{eff} value. An increase in temperature of the BeO reflector results in increase in the scattering cross sections as illustrated in the WLUP multigroup comparison graphs (Fig. 5). Below 4 eV, neutron scattering cross sections in BeO are affected by chemical binding (Be in BeO). The density effect due to increase in temperature reduces reactivity, as we would expect, but this is not the dominant factor in these results.

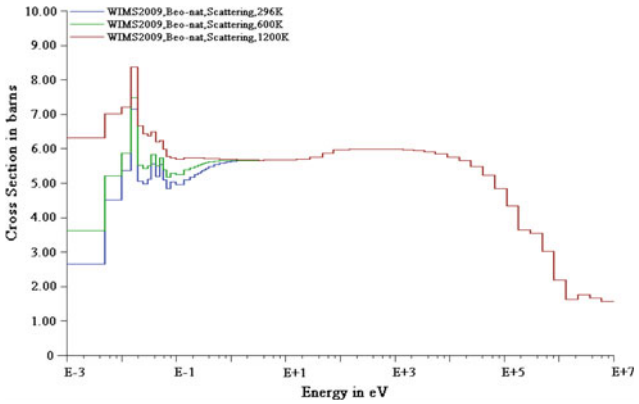

Fig. 5 Typical scattering cross sections of Be in BeO, generated by using the XnWLUP package [33] at 296, 600, and 1200 K. Note the very high temperature dependence below 4.6 eV

The author has informally suggested to the team at the Indira Gandhi Centre for Atomic Research (IGCAR) to experimentally confirm this new effect.

Formation of the Nuclear Data Physics Centre of India (NDPCI)

The great importance of nuclear data for the BARC three-stage program was institutionally recognized by the Department of Atomic Energy in 2004 as a result of the KAPS over-power transient incident [3, 5, 34]. A change of mindset took place in a systemic context, moving towards recognizing the importance of nuclear data physics for energy and non-energy applications [3, 5]. The DAE has declared nuclear data physics as a thrust area.

Activities of the Nuclear Data Physics Centre of India include the following actions towards satisfying the needs of the U–Pu and Th–U fuel cycles:

- Measurement of neutron- and charged-particle-induced cross sections as related to the Th–U fuel cycle, ADSS, AHWR, CHTR, shielding, fast reactors, and other programs of the DAE [2, 6–8], also medical isotopes production;
- Generation and use of co-variances as part of nuclear data evaluation and usage;

- Compilation and evaluation of nuclear reactions and nuclear structures; EXFOR compilations, ENSDF-related compilations;
- DAE-BRNS sponsored NDPCI theme meetings and national conferences on topics in nuclear data physics;
- Advanced reactor applications to enable use of updated nuclear data libraries in plug-in formats, such as for discrete ordinates and Monte Carlo codes;
- Coordination on nuclear data physics involving IAEA NDS and to be a single window from India to IAEA NDS;
- International collaborations with: the CERN neutron time-of-flight facility (n_TOF) under the BARC memorandum of understanding, Korea, IAEA CRPs;
- Identification of faculty and support for formation of useful local neutron data centers in Indian universities and institutes.

The NDPCI, despite its phenomenal progress since 2004, in the opinion of the author, is still in the lower part of its learning curve. NDPCI recognizes that a large amount of work will be essential to create ENDF/B formatted evaluated nuclear data files using new experimental data. These tasks include follow up with "raw" nuclear data compilations, critical evaluation using inference and data evaluation/assimilation methodologies, production of new ENDF/B formatted libraries, including co-variances extending to higher energies, and quality assured nuclear data processing activities to provide the designers/users of innovative systems with "ready to plug-in" processed data that has been integrally validated for use in applications. Nuclear data sensitivity studies such as in [13, 40] are part of activities promoted by the NDPCI.

Concluding Remarks

The indigenous Indian efforts in nuclear data science, especially those towards utilization of the thorium fuel cycle, are expected to continue with their associated excitement and challenges. Many topics mentioned during the talk are not covered here due to space constraints. The formation of the NDPCI and India's successful activities [41] in nuclear data science, since 2004, has helped India in her efforts to graduate from the previous status of India as a user of freely available plug-in multigroup nuclear data and processed evaluated nuclear data files to the status of a contributor of improved/new basic nuclear data. The NDPCI is currently operating in virtual mode. Efforts by BARC are underway to create a sustainable physical center for the NDPCI.

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Nuclear Data for the Thorium Fuel Cycle and the Transmutation of Nuclear Waste

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Abstract

Neutron-induced reaction cross sections play an important role in a wide variety of research fields, ranging from stellar nucleosynthesis, the investigation of nuclear level density studies, to applications of nuclear technology, including the transmutation of nuclear waste, accelerator-driven systems, and nuclear fuel cycle investigations. Simulations of nuclear technology applications largely rely on evaluated nuclear data libraries. These libraries are

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based both on experimental data and theoretical models. An outline of experimental nuclear data activities at CERN's neutron time-of-flight facility, n_TOF, will be presented.

Introduction

An important aspect of the design and safe operation of novel and existing nuclear reactors depends on sophisticated calculations and simulations involving nuclear reactions. These calculations are largely based on so-called nuclear data, a generic notion comprising the physical properties related to nuclear structure and nuclear reactions.

The nuclear data of nuclear reactions needed for such calculations are usually based on evaluated nuclear data libraries, such as JEFF [1], ENDF [2], JENDL [3], CENDL, BROND, and several others.

Nuclear data in general, and neutron-induced reactions in particular, are important for a number of research fields. They play a key role in the safety and criticality assessment of nuclear technology, not only for existing power reactors but also for medical applications [4], radiation dosimetry, the transmutation of nuclear waste, accelerator-driven systems, future reactor systems such as Generation IV reactors, and the thorium-based nuclear fuel cycle [5–7]. Other applications of nuclear data are related to research fields such as the study of nuclear level densities [8, 9] and stellar nucleosynthesis [10–12].

Evaluated nuclear reaction data are intended to be complete and to contain all reactions and all energy regions, even where experimental data are missing, insufficient, or inconsistent with other experimental data sets. Part of the tasks of the evaluator is to make choices between inconsistent

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datasets, find optimum theoretical models describing experimental data, and provide data for "missing" gaps, combining input from available experiments and theory into a single recommended dataset, the evaluation. Both theoretical models and experimental data are the fundamental ingredients in evaluated data.

Experimental nuclear reaction data are compiled on a worldwide basis by the international network of nuclear reaction data centers (NRDC) in the EXFOR database [13]. The EXFOR database forms an important link between nuclear data measurements and the evaluated data libraries. To preserve experimental nuclear data for future use and, notably, for evaluations, measured nuclear data as close as possible to measured quantities should be stored in EXFOR, including all experimental information needed to exploit these data.

One of the most striking features of neutron–nucleus interactions is the resonance structure observed in the reaction cross sections at low incident neutron energies. As the electrically neutral neutron has no Coulomb barrier to overcome, and has negligible interaction with the electrons in matter, it can directly penetrate and interact with the atomic nucleus, even at very low kinetic energies on the order of electron-volts. The cross sections can show variations of several orders of magnitude on an energy scale of only a few eV. The origin of the resonances is well understood. They are related to the excitation of nuclear states in the compound nuclear system formed by the neutron and the target nucleus, at excitation energies lying above the neutron-binding energy of typically several MeV. As an example, in the lower panel of Fig. 1, several neutron-induced cross sections for a typical nucleus are shown as a function of the neutron kinetic energy ranging from meV to GeV. One can clearly distinguish the resolved resonances in the total, fission, and capture cross sections, corresponding to excited states in the compound nucleus formed by the neutron and the target nucleus. At higher energies, threshold reactions such as (n, nγ) and (n, xn) are shown as well. In the upper panel, the typical energy distributions of neutrons are shown on the same energy scale. For the examples given, these distributions closely follow a Maxwell–Boltzmann distribution. At low energy, the neutron spectrum of fully thermalized neutrons at room temperature is shown. On the high-energy side, a typical energy distribution of neutrons following fission induced by thermal neutrons is shown, as well as an accelerator-driven system (ADS) neutron spectrum [14]. In between, we find neutron spectra for stellar environments in the kT = 5 – 100 keV range (with k the Boltzmann constant and T the temperature) where the astrophysical s-process takes place.

The Neutron Time-of-Flight Facility (n_TOF) at CERN

The neutron time-of-flight facility, n_TOF, was constructed after an idea proposed by Rubbia et al. [15] and became fully operational with the start of the scientific measurement program in May 2002. The facility is based on the 6-ns wide, 20 GeV/c pulsed proton beam from CERN's proton synchrotron (PS) with typically 7 × 10^{12} protons per pulse, impinging on a lead spallation target, yielding about 300 neutrons per incident proton. A layer of water close to the spallation target moderates the initially fast neutrons down to

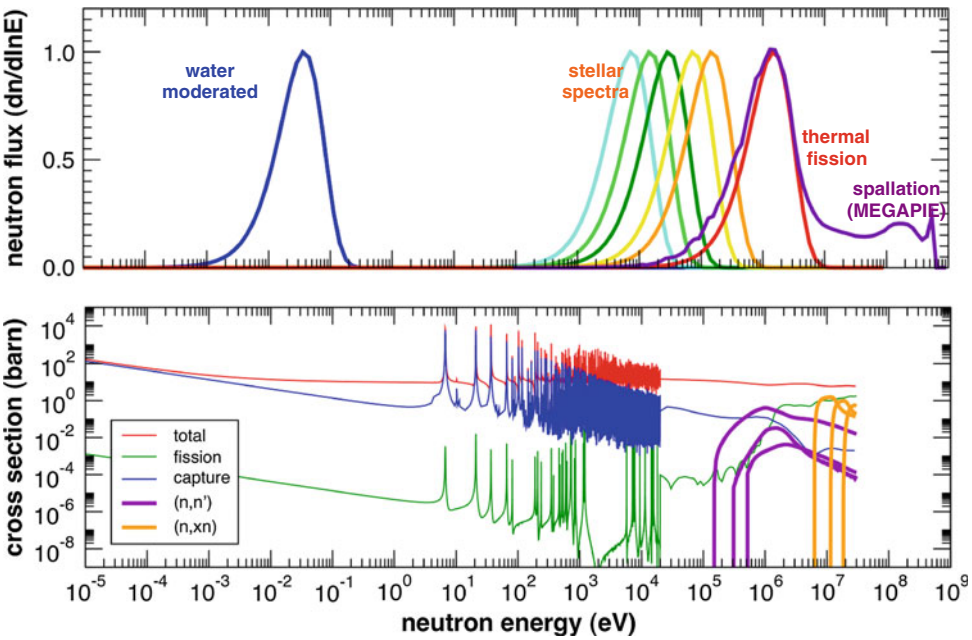

Fig. 1 *Top* Several typical neutron energy distributions, normalized to unit height. *Bottom* Different types of neutron-induced reaction cross sections on the same energy scale

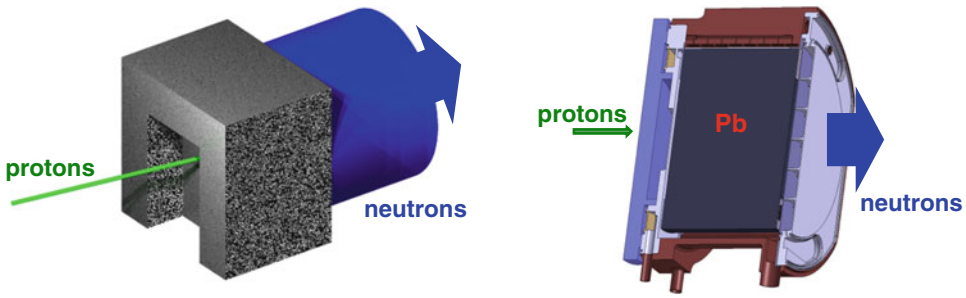

Fig. 2 The two lead spallation targets used as a neutron source at n_TOF. *Left* The first target with a 80 × 80 × 60 cm^3 lead block, in use from 2001 to 2004. The surrounding cooling and moderator water is not shown. *Right* A view of the second target, a cylindrical block of 40 cm height and 60 cm diameter, in use from 2009 with different moderators

a white spectrum of neutrons covering the full kinetic energy range between meV and GeV neutrons. The minimal time between two proton pulses is a multiple of 1.2 s, related to the operation cycle of the PS, which allows the neutron energy range to be covered down to subthermal energies without overlap of slow neutrons from previous cycles. The neutron beam is collimated and guided through a vacuum neutron tube over a distance of approximately 185 m to an experimental area (EAR1) where samples can be mounted in the beam and neutron-induced reactions can be studied. The kinetic energy of the neutrons is determined by the time-of-flight, which, combined with the known travel distance, gives the neutron velocity. A more detailed description of the facility can be found in [16] and references therein.

During phase-I, when the first spallation target was used from 2001 to 2004, the water coolant also served as the moderator. The spallation target was a block of lead of dimensions 80 × 80 × 60 cm^3. During phase-II, after the installation in 2008 of an upgraded cylindrical lead spallation target 60 cm long and 40 cm in diameter, the target was enclosed with a separate cooling circuit resulting in 1 cm water layer in the beam direction, followed by an exchangeable moderator with a thickness of 4 cm. As moderator, normal water has been used as well as water with a saturated ^{10}B solution to reduce the number of 2.23 MeV gamma rays from hydrogen capture, which otherwise forms an important contribution to the background owing to in-beam gamma rays. This influences the energy distribution of the neutron flux noticeably only below 1 eV. A schematic view of the two spallation targets is shown in Fig. 2. More details on the phase-II facility can be found in [16].

Contributions from the n_TOF Facility to Nuclear Data

Until recently, the facility was recording data for two main periods: phase-I and phase-II, using a single flight path with an experimental area (EAR1) at about 185 m. A second, vertical flight path leading to a second experimental area (EAR2) has been constructed recently and is currently under commissioning. In addition to nuclear data measurements, the collaboration has put effort into developing instrumental, detection, and beam imaging techniques.

Nuclear Data Measurements During Phase-I

During the first phase from 2001 to 2004, capture and fission data for a number of isotopes have been taken. Capture measurements with C_6D_6 liquid scintillator detectors included $^{24,25,26}Mg$, ^{56}Fe, $^{90,91,92,93,94,96}Zr$, ^{139}La, ^{151}Sm, $^{186,187,188}Os$, ^{197}Au, $^{204,206,207,208}Pb$, ^{209}Bi, and ^{232}Th. A 4π calorimeter consisting of 40 BaF_2 crystals (**TAC**) was used for neutron capture measurements of ^{197}Au, ^{233}U, ^{234}U, ^{237}Np, ^{240}Pu, and ^{243}Am. Fission cross sections were measured with the **FIC-0** fission detector containing the isotopes ^{232}Th, ^{234}U, ^{235}U, ^{236}U, ^{238}U, and ^{237}Np. A different detector, **FIC-1**, which was ISO-2919 compliant, was used to measure neutron-induced fission cross sections of the isotopes ^{233}U, ^{235}U, ^{238}U, ^{241}Am, ^{243}Am, and ^{245}Cm. A different development was the fission detectors based on parallel plate avalanche counters (PPACs), with which the fission cross sections of ^{nat}Pb, ^{209}Bi, ^{232}Th, ^{237}Np, ^{233}U, ^{234}U, ^{235}U, and ^{238}U have been measured. A list of measured isotopes and reactions, together with the final or most relevant publication, is given in Table 1.

As an example, the resolved resonance region up to 4 keV of the $^{232}Th(n, \gamma)$ measurement is shown in Fig. 3. The datapoints (in black) with their uncertainties are shown with a density of 5000 bins per energy decade. The reconstructed capture yield from R-matrix resonance parameters, including Doppler and resolution broadening, is shown on the same figure (in red). One can clearly distinguish the many resonances as a result of the high resolution. A total of 391 resonances were resolved. More details on this experiment can be found in [17, 18].

Table 1 The measurements performed at n_TOF during phase-I from 2001 to 2004

Isotope	Reaction	Detector	References
^{24}Mg	(n, γ)	C_6D_6	[19]
^{26}Mg	(n, γ)	C_6D_6	[19]
^{90}Zr	(n, γ)	C_6D_6	[20]
^{91}Zr	(n, γ)	C_6D_6	[21]
^{92}Zr	(n, γ)	C_6D_6	[22]
^{94}Zr	(n, γ)	C_6D_6	[23]
^{96}Zr	(n, γ)	C_6D_6	[24]
^{129}La	(n, γ)	C_6D_6	[25]
^{151}Sm	(n, γ)	C_6D_6	[26–28]
^{186}Os	(n, γ)	C_6D_6	[29, 30]
^{187}Os	(n, g)	C_6D_6	[29, 30]
^{188}Os	(n, g)	C_6D_6	[29, 30]
^{204}Pb	(n, γ)	C_6D_6	[31]
^{206}Pb	(n, γ)	C_6D_6	[32, 33]
^{207}Pb	(n, γ)	C_6D_6	[34]
^{208}Pb	(n, γ)	C_6D_6	–
^{209}Bi	(n, γ)	C_6D_6	[35]
^{232}Th	(n, γ)	C_6D_6	[17, 18]
^{233}U	(n, γ)	TAC	–
^{234}U	(n, γ)	TAC	–
^{237}Np	(n, γ)	TAC	[36]
^{240}Pu	(n, γ)	TAC	–
^{243}Am	(n, γ)	TAC	[37]
^{233}U	(n, f)	FIC	[38, 39]
^{234}U	(n, f)	FIC	[40]
^{236}U	(n, f)	FIC	[41]
^{241}Am	(n, f)	FIC	[42]
^{243}Am	(n, f)	FIC	[43]
^{245}Cm	(n, f)	FIC	[44]
^{234}U	(n, f)	PPAC	[45, 46]
^{237}Np	(n, f)	PPAC	[45]
^{209}Bi	(n, f)	PPAC	[47]
^{nat}Pb	(n, f)	PPAC	[47]
^{232}Th	(n, f) ang.	PPAC	[48]

Nuclear Data Measurements During Phase-II

During phase-II, from 2009 to 2012, mostly capture measurements were performed. In addition to these measurements, several other techniques have been tested for the first time at this facility. A first experiment aimed at resonance spin assignments was performed on a sample of ^{87}Sr. A first test measurement with a MicroMegas detector (MGAS) was performed to get a fission measurement on ^{240}Pu and ^{242}Pu. The results on ^{240}Pu were not conclusive owing to the high radioactivity. Another reaction that was investigated was the (n, α) reaction, using a MicroMegas detector with ^{33}S, and using a chemical vapor deposition (CVD) diamond detector with ^{59}Ni. Finally, the ^{12}C(n, p)^{12}B reaction was exploited by in-beam activation. References for the phase-II measurement results are given in Table 2.

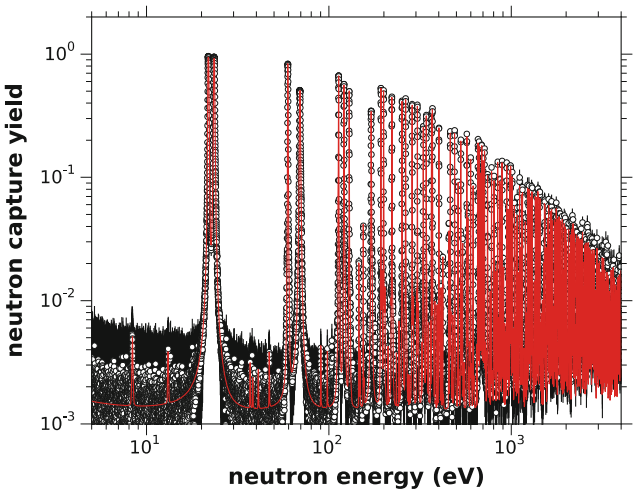

Fig. 3 The ^{232}Th(n, γ) capture yield data (*black*) as measured at the n_TOF facility at CERN together with the reconstructed yield (*red*) from resonance parameters

Conclusion and Outlook

The measurement of neutron-induced reaction cross sections is one of the major goals of the n_TOF collaboration. Reaching this goal goes together with a continuous development of detectors and analysis techniques, the design of feasible experiments, and the training of a new generation of physicists working in neutron physics. Sample preparation is an increasingly important topic that needs to be addressed. To preserve the measured data for the future, a close collaboration with the community of experimental and evaluated nuclear data libraries is mandatory.

The final construction of EAR2, at a ten-times shorter distance from the spallation target than EAR1, after a period of design and construction marks the start of a third phase of data taking at n_TOF, allowing us to push the limits of feasibility of neutron-induced reaction measurements. The ongoing commissioning aims at measuring and validating its

Table 2 The measurements performed at n_TOF during phase-II from 2009 to 2012

Isotope	Reaction	Detector	References
^{33}S	(n, α)	MGAS	–
^{59}Ni	(n, α)	CVD diamond	[49]
^{25}Mg	(n, γ)	C_6D_6	[50]
^{54}Fe	(n, γ)	C_6D_6	[51]
^{56}Fe	(n, γ)	C_6D_6	–
^{57}Fe	(n, γ)	C_6D_6	[51]
^{58}Ni	(n, γ)	C_6D_6	[52]
^{62}Ni	(n, γ)	C_6D_6	[53]
^{63}Ni	(n, γ)	C_6D_6	[54]
^{87}Sr	spins	C_6D_6	[55]
^{92}Zr	(n, γ)	C_6D_6	–
^{93}Zr	(n, γ)	C_6D_6	[56]
^{197}Au	(n, γ)	C_6D_6/TAC	[57, 58]
^{235}U	(n, γ)/(n, f)	TAC/MGAS	[59]
^{236}U	(n, γ)	C_6D_6	[60]
^{238}U	(n, γ)	C_6D_6	[61]
^{238}U	(n, γ)	TAC	[62]
^{241}Am	(n, γ)	C_6D_6	[63]
^{241}Am	(n, γ)	TAC	[64]
^{242}Pu	(n, f)	MGAS	[65]
^{12}C	(n, p)	Activation	[66]

characteristics in terms of flux, beam profile and backgrounds, and making a detailed comparison between the two experimental areas EAR1 and EAR2.

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Fast Reactor Physics

K. Mikityuk

Abstract

A very simplified model is used to simulate the equilibrium fuel cycle in a sodium-cooled fast reactor, considering (for the sake of comparison) two feed fuels: natural uranium and natural thorium, assuming recycling of all actinides under conditions of constant power density and constant fuel mass. The balance of reaction rates, equilibrium fuel composition, neutron balance, main safety parameters, as well as radiotoxicity and decay heat level of the equilibrium fuel are presented. The paper is a shortened version of the lecture given at FJOH'2013 summer school (Mikityuk, Equilibrium closed fuel cycle, 2013, [1]).

Introduction

A fast neutron critical reactor is a category of nuclear reactor in which the fission chain reaction is sustained by fast and resonance neutrons. Such a reactor needs no neutron moderator and typically uses stainless steel as a cladding and sodium as a coolant. Typical neutron spectra as well as the unit cell schematics are compared in Fig. 1 for a sodium-cooled fast reactor (SFR) and a pressurized water reactor (PWR).

The materials loaded in the reactor core exhibit nuclear transmutations, that is, conversions of one element or isotope into another due to reactions with neutrons (including fission, (n, γ), (n, 2n), (n, α) reactions, etc.) and due to radioactive decays (including spontaneous fission, α- and β-decays, electron capture). A transmutation of actinides with low fission probability (e.g., fertile U-238 or Th-232) into actinides with high fission probability (e.g., fissile Pu-239 or U-233) is called breeding or conversion. Owing to the nuclear properties of actinides in a high neutron energy region, a fast-spectrum nuclear reactor can be designed to enable efficient indirect burning of actinides available in nature (U-238 and Th-232) and recycling of all actinides, producing as final waste fission products only.

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In general, the nuclear fuel cycle is the progression of nuclear fuel from mining to final disposal through enrichment, fabrication, irradiation, cooling, and reprocessing (which is, in fact, separation of elements) [2]. The actinides periodically added to the fuel cycle are called feed fuel. The feed fuel can include natural uranium and natural thorium, enriched and depleted uranium, and weapons-grade actinides from nuclear weapons. The fuel cycle based on U-238 as a feed fuel is called the uranium fuel cycle, whereas the fuel cycle based on Th-232 as a feed fuel is called the thorium fuel cycle.

An open (or once-through) uranium fuel cycle does not include fuel reprocessing and is currently used at nearly all nuclear power plants. To reduce the radiotoxicity and decay heat load on the final repository, all actinides, which mostly contribute to these parameters, could be separated and recycled in the fast reactor. Diagrams of the corresponding fuel cycles are shown in Fig. 2a and b for U and Th fuel cycles, respectively. In this option, only fission products and reprocessing losses go to the final repository.

Owing to the unavailability of the thorium cycle fissile element (U-233), fissile material (plutonium) should be borrowed from the uranium cycle to start the thorium fuel cycle until enough U-233 is generated.

In this paper, the equilibrium fuel cycle, which is closed on all actinides for both the U and Th fuel cycles (EQL-U and EQL-Th), is considered. The equilibrium cycle is a cycle with a sufficiently large number of repetitive cycles, after

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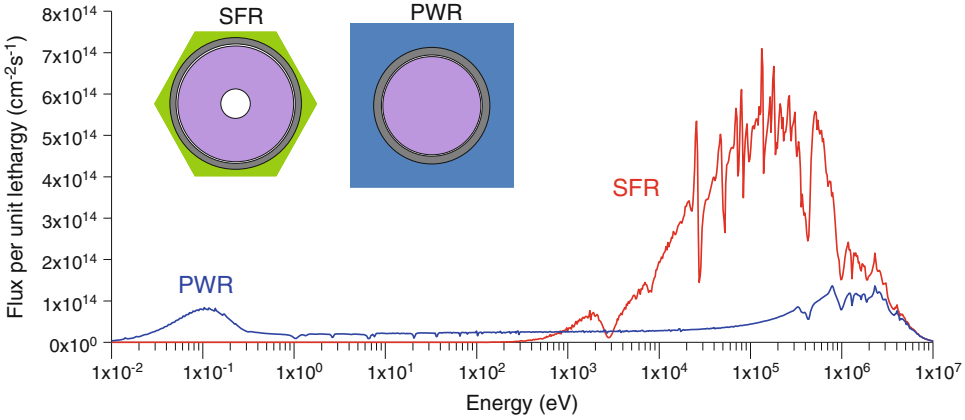

Fig. 1 Typical neutron spectrum of fast (SFR) and thermal (PWR) reactors as well as the schematics of the SFR and PWR unit cells [1]

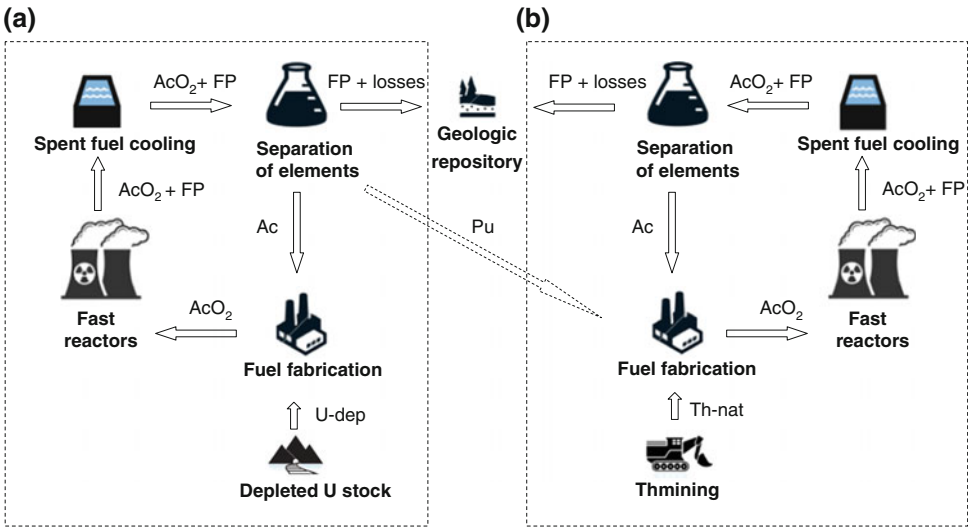

Fig. 2 Uranium (**a**) and thorium (**b**) fuel cycles closed on all actinides. *U-dep* depleted uranium; *Th-nat* natural thorium; *Ac* all actinides of the corresponding cycle; *FP* fission products

which the composition of actinides, averaged over the whole cycle duration from cycle to cycle, remains unchanged and the resulting reactor parameters can be considered as asymptotic characteristics of the reactor with imposed power, feed fuel, and reprocessing parameters. In this context, any reactor operation in repetitive fuel cycles can be considered as in transition from beginning-of-life to the asymptotic equilibrium state.

Calculation Tools, Assumptions, and Models

To study the mass flows, balance of reaction rates, neutron balance, radiotoxicity, and decay heat level of the equilibrium fuels, a simplified model of the equilibrium fuel cycles was developed, using the following assumptions and simplifications:

- The neutron spectrum is calculated for a unit cell, simulating the infinite fuel lattice of typical SFRs. No neutron leakage is considered;
- The power density and the total mass of actinides remain constant;
- The cooling and reprocessing times are assumed to be zero, which is equivalent to the assumptions of on-line feeding and reprocessing;
- The losses are assumed to be negligible and, therefore, only fission products (FPs) are removed from the system and replaced by the feed fuel to keep the mass of the actinides constant;
- The fission products are not considered in the neutron balance, mass evolution, evaluation of safety parameters, radiotoxicity, and decay heat.

We used the Serpent 1.1.16 Monte-Carlo code [3] for solving the eigenvalue problem and finding the infinite multiplication factor, the one-group cross sections, and integral flux for fuel region, by using the unit-cell model and an initial guess for fuel composition. After this step, we solved for all actinides a set of the steady-state Bateman equations, describing the evolution (transmutation) of actinides under the given irradiation conditions, characterized

by the set of one-group cross sections and integral flux as well as by decay constants for radioactive isotopes. The resulting fuel composition was then given to the unit-cell model and the Serpent calculation was repeated. When the iterations on k_∞ converged, the safety parameters as well as radiotoxicity and decay heat were calculated. The details of the procedure can be found in reference [1].

More detailed studies, in which neutron leakage as well as reprocessing and cooling times were explicitly simulated, can be found in references [4, 5].

Results for EQL-U and EQL-Th Closed Fuel Cycles in SFR

Reaction Rates

The balance of reaction rates in the EQL-U and EQL-Th fuel cycles closed on all actinides are shown in Fig. 3a and b, respectively. All reaction rates are normalized to the feeding rate, equal to 1000 U-238 and Th-232 atoms added to the system per unit time.

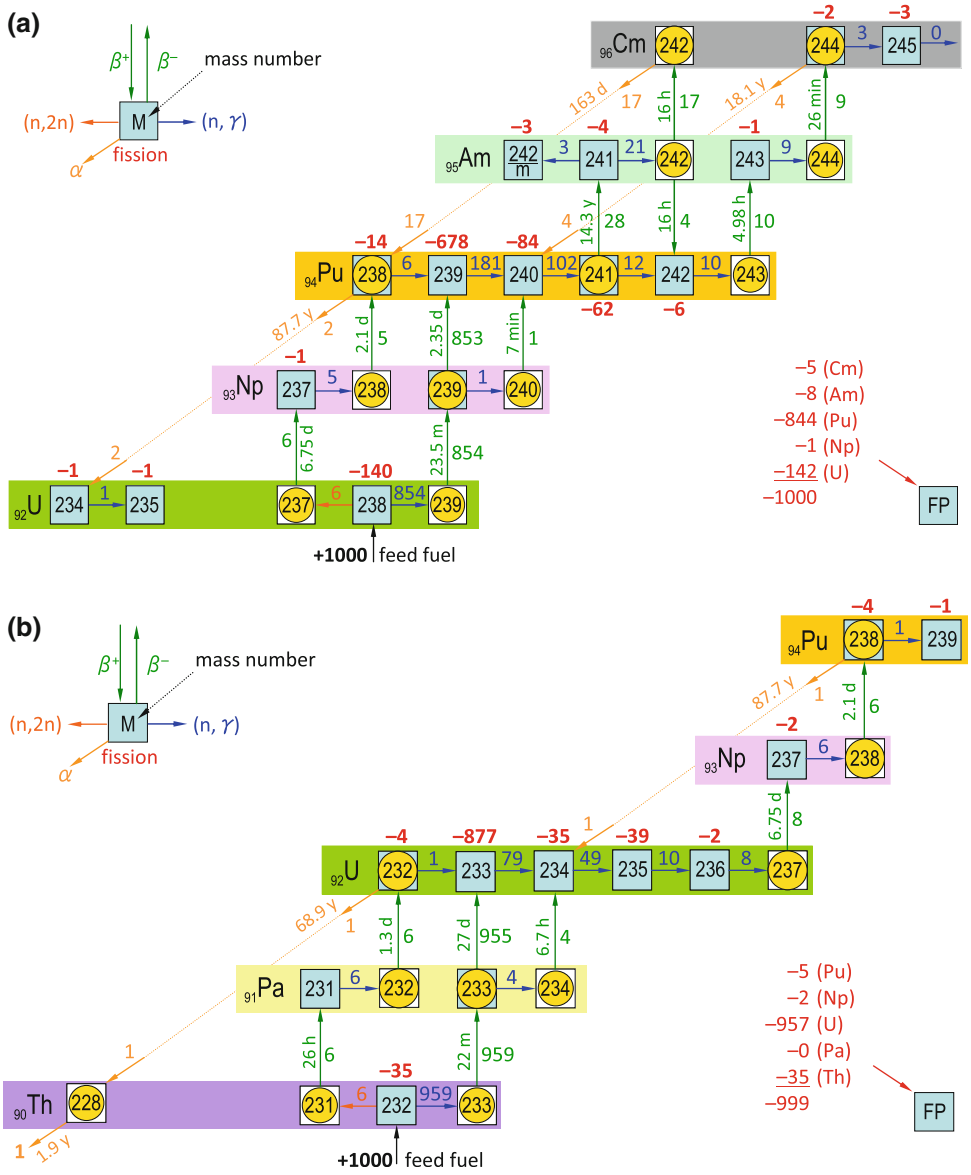

Fig. 3 Balance of reaction rates in the EQL-U (**a**) and EQL-Th (**b**) closed fuel cycles [1]

Table 1 Microscopic cross sections, fission-to-destruction ratios, and mass fractions for the main actinides of the EQL-U and EQL-Th fuel cycles in SFR [1]

	σ_{fc}(b)	σ_c (b)	λ/Φ (b)	F/(A + D)	EQL-U (wt%)	EQL-Th (wt%)
Th^{228}	0.02	0.30	8.69	0.00		0.04
Th^{230}	0.03	0.14		0.18		0.04
Th^{232}	0.01	0.31		0.04		85.6
Pa^{231}	0.25	2.72		0.08		0.06
Pa^{233}	0.07	0.86	224.8	0.00		0.12
U^{232}	2.04	0.59	0.24	0.71		0.05
U^{233}	2.51	0.23		0.92		9.56
U^{234}	0.31	0.50		0.38	0.07	2.98
U^{235}	1.85	0.53		0.78	0.01	0.60
U^{236}	0.10	0.39		0.21	0.04	0.63
U^{238}	0.04	0.27		0.14	81.59	
Np^{237}	0.32	1.49		0.17	0.10	0.13
Np^{239}	0.44	1.86	2474.7	0.00	0.00	
Pu^{238}	1.21	0.50	0.18	0.64	0.31	0.10
Pu^{239}	1.73	0.46		0.79	10.17	0.02
Pu^{240}	0.37	0.46		0.45	5.78	0.01
Pu^{241}	2.46	0.46	1.12	0.61	0.66	
Pu^{242}	0.26	0.46		0.36	0.55	
Am^{241}	0.27	1.74	0.04	0.13	0.36	
Am^{242m}	3.01	0.47	0.11	0.84	0.02	
Am^{243}	0.20	1.55		0.11	0.15	
Cm^{242}	0.62	0.45	35.8	0.02	0.01	
Cm^{244}	0.41	0.82	0.88	0.20	0.11	
Cm^{245}	2.65	0.51		0.84	0.03	
Cm^{246}	0.28	0.50		0.36	0.02	

In equilibrium, every feed fuel nucleus added to the system is burned: either as feed fuel or as another nucleus after one or several transmutations. The mass flow rate between actinides and the distribution of the fission rates over the actinides are determined by the ratio between fission, capture, and decay probabilities, characterized by a one-group fission cross section (σ_f), one-group capture cross section (σ_c) and the decay constant divided by the integral flux (λ/Φ). The fission-to-destruction ratio, $F/(A + D) = \sigma_f/(\sigma_f + \sigma_c + \lambda/\Phi)$, of an actinide characterizes its fissile quality. The higher this value is, the higher the fraction of the fission in the total reaction rate for a given isotope, and the lower the transmutation of this isotope to the higher actinides. As a rule, a lower equilibrium amount of higher actinides is beneficial for the neutron balance, safety parameters, decay heat, and radiotoxicity. In particular, the fuel composition for neutron balance, which is "closer" to the feed fuel composition, means that fewer neutrons are lost for captures. Microscopic cross sections and fission-to-destruction ratios for the main actinides in the EQL-U and EQL-Th fuel cycles are given in Table 1.

From the viewpoint of the source of penetrating radiation from actinides, a property that requires the protection of workers during fuel reprocessing, in the EQL-U cycle this is mainly a neutron source from spontaneous fissions of minor actinides, whereas in the EQL-Th cycle this is mainly a hard gamma source from the products of U-232 decay. It is worth remembering that the fission products are not considered in this study; thus, in the short-term, the gamma source from the fission products dominates [6].

Fuel Composition

The resulting fuel compositions in wt% are shown in Fig. 4 and Table 1 for the equilibrium uranium and thorium cycles. The first observation is that, in the case of the EQL-Th cycle, the fuel composition is "closer" to the feed fuel (Th-232).

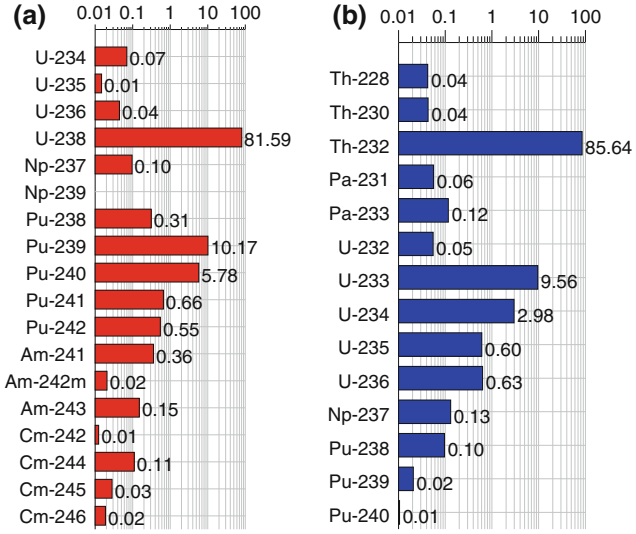

Fig. 4 Fuel composition in EQL-U (**a**) and EQL-Th (**b**) closed fuel cycles in SFR (wt%)

Because of the low mass number of Th-232, a very low amount of minor actinides (Np, Am, Cm, etc.) is generated in the thorium cycle. The second observation is a difference of about 5 % in feed fuel concentration (U-238 and Th-232), which in the equilibrium is determined by the ratio of the macroscopic fission cross section, Σ_f, divided by the microscopic total cross section of feed fuel, σ_t [1]. Although σ_t is about the same for U-238 and Th-232, Σ_f is about 5 % higher in the EQL-Th cycle.

The third observation relates to the ratio between the primary fissile (Pu-239 and U-233) and primary fertile (U-238 and Th-232) concentrations, which is about 12 % and 11 % in the U and Th cycles, respectively. Neglecting small bypass reactions (see Fig. 3a and b), this ratio can be estimated by dividing the capture cross section of the primary fertile by the total cross section of the primary fissile [1, 5].

Neutron Balance

To evaluate the individual contributions of actinides to the multiplication factor, k_∞, we use the following simple neutron balance equation:

$$k_\infty = P/A = \sum_i P_i / \left(\sum_i A_i + A_{CC} \right) \qquad (1)$$

where P is the total neutron production rate and A is the total neutron absorption rate. A can be presented as a sum of the absorption rates of individual actinides plus A_{CC}—the rate of neutron absorption by cladding and coolant (for the sake of simplicity we neglect the absorption by oxygen in fuel). Similarly, P can be presented as the sum of the neutron production rates of individual actinides. If A is normalized to 1, then P equals k_∞. Using this isotope-wise decomposition, we can consider the value P_i/A as a contribution of the isotope, i, to the total multiplication factor.

The isotope-wise decomposition of the numerator (red bars) and denominator (blue bars) of (1) is shown in Fig. 5. The main observation is a significantly lower k_∞ in the EQL-Th cycle compared with the EQL-U cycle, mainly explained by a lower fission rate of the primary fertile isotope Th-232 compared with U-238.

Safety Parameters

The inherent safety of fast-spectrum systems is based on the negative (leakage) component of the void reactivity effect,

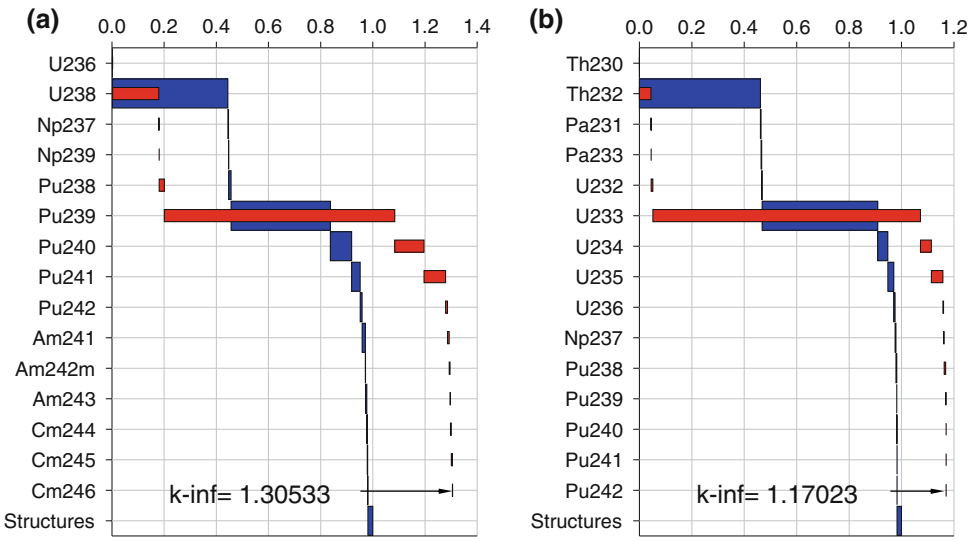

Fig. 5 Isotope-wise decomposition of neutron absorption and production rates normalized to a total absorption rate equal to 1.0 for the EQL-U and EQL-Th fuel cycles. *Blue bars* absorption; *red bars* production

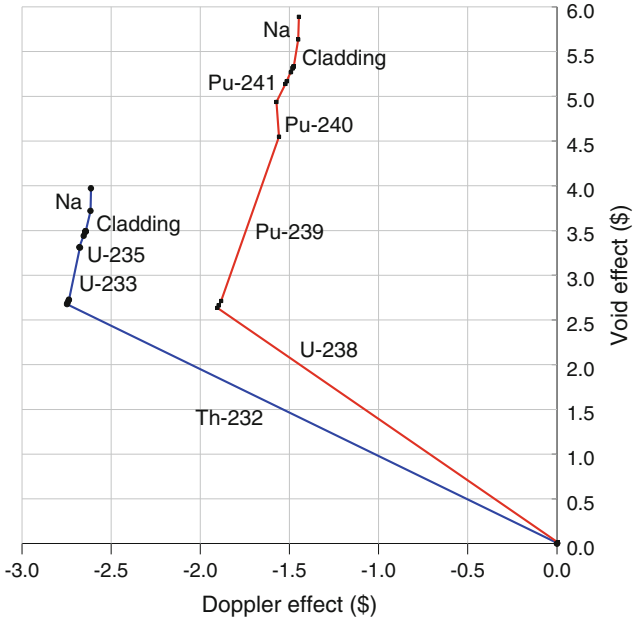

Fig. 6 Decomposition of Doppler and void effects for the EQL-U and EQL-Th fuel cycles

the negative fuel Doppler reactivity effect, and negative reactivity effects from thermal expansions of the in-reactor structures.

We consider the two most important safety parameters: Doppler and void effects.[1] To understand the contribution of individual actinides to the total effect, we made a simple actinide-wise decomposition assumption: the actinide contribution is evaluated as the change of the rate of absorption by the given actinide divided by the total neutron production rate, assumed constant. The results of this evaluation are shown in Fig. 6.

Radiotoxicity and Decay Heat

Radiotoxicity and decay heat for 1 g of the EQL-U and EQL-Th fuel were calculated, by using coefficients derived under framework of another study [7] (Fig. 7). Mainly, all Pu isotopes, Cm and higher actinides as well as their progenies determine the radiotoxicity and decay heat of the fuel in the EQL-U cycle, whereas U-232, Pu-238, and their progenies are responsible for radiotoxicity and decay heat of the EQL-Th fuel for the first millennium. After 1000 years, the progenies of U-233 and U-234 become the dominating isotopes in terms of radiotoxicity and decay heat.

[1]Doppler and void effects are evaluated as changes of the eigenvalue caused by the changes of fuel temperature from nominal to the melting point and of the coolant density from nominal to zero, respectively.

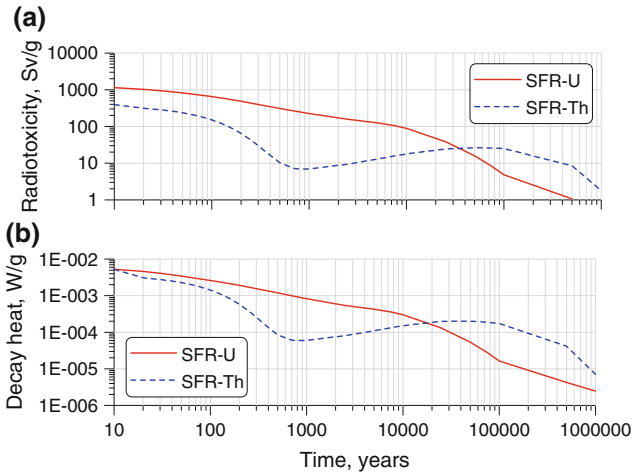

Fig. 7 Radiotoxicity (**a**) and decay heat (**b**) of the EQL-U and EQL-Th fuels

Summary

The advantages and disadvantages of implementation of the thorium cycle in fast reactors in comparison with the uranium cycle can be summarized as follows:

1. Past and current fast reactors were/are based on the uranium cycle. Operational experience with thorium fuel is very limited. Experience in thorium fuel manufacturing and reprocessing is significantly lower compared with that with uranium fuel. This is one of the main obstacles in introducing the Th cycle in fast reactors.
2. An important disadvantage of the Th cycle is that the fissile fuel for this cycle (U-233) is available neither from nature nor from previous reactor operation. Therefore, in order to start the Th fuel cycle, fissile fuel should be borrowed from the U cycle (Pu-239 or U-235).
3. Nevertheless, calculational analysis with state-of-the-art tools shows that fast reactors can operate in an equilibrium thorium cycle closed on all actinides. However, mainly owing to the lower fast fission probabilities of Th-232 compared with U-238, k_∞ of the EQL-Th fuel is significantly lower than k_∞ of the EQL-U one. This means that blankets of fertile material can be required in the thorium fast reactor, which is unfavorable from a non-proliferation viewpoint.
4. Owing to the low mass number of the feed fuel in the Th cycle (Th-232), a very low amount of minor actinides (Np, Am, Cm, etc.) is generated in this cycle and, therefore, the long-lived neutron source, decay heat, and radiotoxicity associated with minor actinides are very low. On the other hand, U-232—precursor of hard gamma emitters—is produced in the Th cycle via the (n, 2n) reaction of Th-232.

5. From a safety viewpoint, the Th cycle has a number of advantages. In particular, because of the nuclear cross sections of the primary fissile and fertile isotopes, the Doppler effect is stronger and the void effect is weaker in the EQL-Th fuel cycle compared with the EQL-U cycle. This trend can be weakened or even reversed for the start-up conditions, when Pu-239 has to be used as the fissile fuel in the Th cycle.
6. From the viewpoint of proliferation risks, the misuse of U-233 is, on one hand, prevented by presence of U-232 (predecessor of hard gamma emitters), but, on the other hand, the precursor of the primary fissile in the Th cycle—Pa-233—has a significantly higher half-life (27 days) compared with the precursor of the primary fissile in the U cycle—Np-239 (2.35 days). This potentially gives time for separation of protactinium, which decays to weapons-grade U-233 practically without the presence of U-232 (due to the low amount of Pa-232 in the equilibrium fuel).
7. Radiotoxicity and decay heat of the equilibrium Th fuel are lower for about the first ten thousand years of cooling compared with the equilibrium U fuel. However, they become higher after ten millennia due to the build-up of decay products.

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Introduction to the Physics of Thorium Molten Salt Fast Reactor (MSFR) Concepts

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Abstract

Recent conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts have kindled renewed interest in molten salt reactors. This concept, operated in the thorium fuel cycle, may be started either with ^{233}U, enriched U, and/or transuranic elements as the initial fissile load. This paper describes some studies and developments around the MSFR concept based on the thorium fuel cycle. MSFRs are seen as a long-term alternative to solid-fueled fast neutron systems thanks to their unique potential, which includes large negative temperature and void coefficients, lower fissile inventory, no initial criticality reserve, a simplified fuel cycle, waste reduction, etc. They have been selected as one of the reference reactors of the Generation IV International Forum.

Introduction

The molten salt fast reactor (MSFR) was chosen by the Generation IV International Forum (GIF) in 2008 as a representative molten salt reactor fitting the Gen IV criteria [1] because of its fast spectrum, sustainability, and waste minimization, and the use of thorium as fertile element owing to its proliferation resistance [2–7]. In such a homogeneous reactor, the main safety characteristics are due to the absence of any moderator or construction materials in the core, which contains only the liquid fuel salt components. Thermal dilation of the liquid fuel salt gives it a thermal feedback coefficient of about −5 pcm/K, which allows power tuning by heat extraction. Because of a negative void feedback coefficient, draining the liquid fuel salt in geometrically subcritical tanks allows long term stalling with passive cooling for decay heat removal. Two advantages of having the fissile and fertile isotopes in a liquid fuel are: (1) the possibility of fuel composition adjustment without stopping the reactor and (2) the circumvention of the difficulties of solid fuel fabrication with large amounts of transuranic elements (TRU). Indeed, this reactor may be operated with a variety of fissile and fertile elements but is most efficient with ^{233}U, Pu, and Th.

This type of reactor is still at a conceptual level, based on numerical modeling. However, in the 1950s and 60s, experimental studies were conducted at the Oak Ridge National Laboratory (ORNL) in the USA. This provided a very valuable experimental base to assess the feasibility of such reactors. In 1958, a water-based liquid fuel was used in a 5 MW_{th} homogeneous reactor experiment called HRE-2, which demonstrated the auto-stability of homogeneous reactors. From 1966–1969, an 8 MW_{th} experimental graphite-moderated molten salt reactor was operated for four years without any trouble, demonstrating that using a molten fluoride salt at 650 °C was possible. However, this molten salt reactor experiment (MSRE) only tested fissile isotopes (^{233}U, ^{235}U, and Pu), not thorium, for breeding. Later, ORNL studied in detail a power reactor called the molten salt breeder reactor (MSBR), which was never built. This design was a thermal reactor with a graphite-moderated core that needed intense chemical salt treatment with about a 30-day removal time for soluble fission products, a draw-back that is eliminated with a fast spectrum.

This paper describes some studies and developments around the MSFR concept and illustrates the contemporary

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interest in fast reactor concepts based on the thorium fuel cycle, which is seen as a long-term alternative to solid-fueled fast neutron reactors.

Description of the MSFR Concept

Core and System Designs

Conceptual design activities are currently (2013) underway so as to ascertain whether MSFR systems can satisfy the goals of Generation IV reactors in terms of sustainability (Th breeder), non-proliferation (integrated fuel cycle, multi-recycling of actinides), resource savings (closed Th/U fuel cycle, no uranium enrichment), safety (no reactivity reserve, strongly negative feedback coefficient), and waste management (actinide burner). Calculation results presented here were obtained for a somewhat arbitrarily chosen reactor called "reference MSFR". This is not to be taken as an optimized reactor, but as a basis for interdisciplinary studies.

The reference MSFR is a 3 GW_{th} reactor with a total fuel salt volume of 18 m^3, operated at a maximum fuel salt temperature of 750 °C [8, 9]. More recently, thermal-hydraulic studies have been performed in the frame of the EVOL (evaluation and viability of liquid fuel fast reactor system) FP7 project, resulting in a torus shape of the core [10, 11]. As shown in Fig. 1, the fuel salt flows from the bottom to the top of the core cavity (note the absence of in-core solid matter). After exiting the core, the fuel salt is fed into 16 groups of pumps and heat exchangers located around the core. The salt traveling time through the circuit is 3–4 s [12]. The fuel salt considered in the simulations is a molten binary fluoride salt with 77.5 mol% lithium fluoride; the other 22.5 mol% consists of a mix of heavy nuclei fluorides. This proportion, maintained throughout the reactor evolution, leads to a fast neutron spectrum in the core. The total fuel salt volume is distributed half in the core and half in the external part of the fuel circuit. This MSFR system thus combines the generic assets of fast neutron reactors (extended resource utilization, waste minimization) with those associated with a liquid-fueled reactor.

In preliminary designs developed in relation to calculations, the core of the MSFR is a single compact cylinder (2.25 m high × 2.25 m diameter) and the nuclear reactions occur within the liquid fluoride salt, which acts both as fuel and as coolant. The external core structures and the fuel heat exchangers are protected by thick reflectors made of nickel-based alloys, which are designed to absorb more than 99 % of the escaping neutron flux. These reflectors are themselves surrounded by a 20-cm thick layer of B_4C, which provides protection from the remaining neutrons. The radial reflector includes a fertile blanket (50-cm thick; red area in Fig. 1) to increase the breeding ratio. This blanket is filled with a LiF-based fertile salt with initially 22.5 mol% $^{232}ThF_4$.

The fuel circuit is connected to a salt draining system, which can be used for a planned shut down or in case of any incident/accident leading to an excessive temperature being reached in the core. In such situations, the fuel salt geometry can be passively reconfigured by gravity-driven draining of

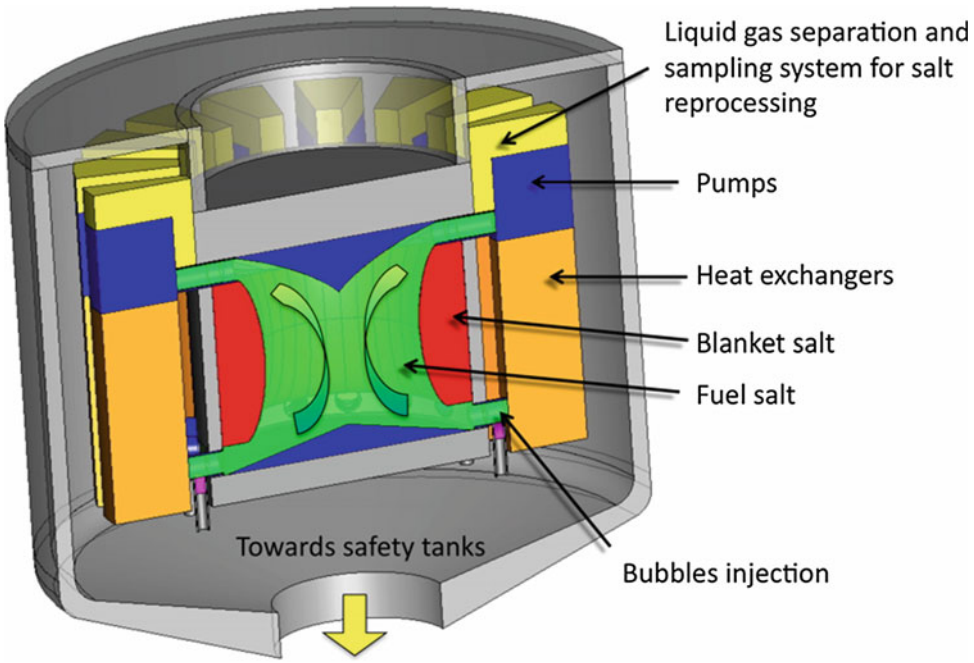

Fig. 1 Schematic drawing of the MSFR design. Fluoride-based fuel salt is *green*, fertile blanket salt is *red*

the fuel salt into tanks located under the reactor and where a passive cooling and adequate reactivity margin can be implemented.

Figure 2 is a general view of what a reactor could look like, with its elements represented as generic boxes for the various functions because they have not yet been studied in detail.

The first barrier (pink) includes three zones. The upper zone contains the fuel circuit (green) and the neutral gas reprocessing (yellow). A collector for salt draining is represented (funnel and vertical tube), leading the drained salts to containers with subcritical geometry (not detailed) situated in a large water pool. This large water pool acts as a thermal buffer in case of high temperature emergency draining. At the bottom of this pool is located a layer containing a dilution salt that can passively mix with the fuel salt in case of a large first barrier failure. This can provide neutron poisons to the fuel and create a large salt-wall interface for passive cooling in the event of a severe accident. Heat pipes (dark blue) are used to transfer the decay heat to the atmosphere. This means that decay heat can be removed into the atmosphere in case of a heat sink failure.

Salt Cleaning and Reprocessing

The fuel salt undergoes two types of treatment: on-line neutral gas bubbling in the core and remote mini-batch reprocessing on-site [13]. These salt treatments aim to remove most of the fission products without stopping the

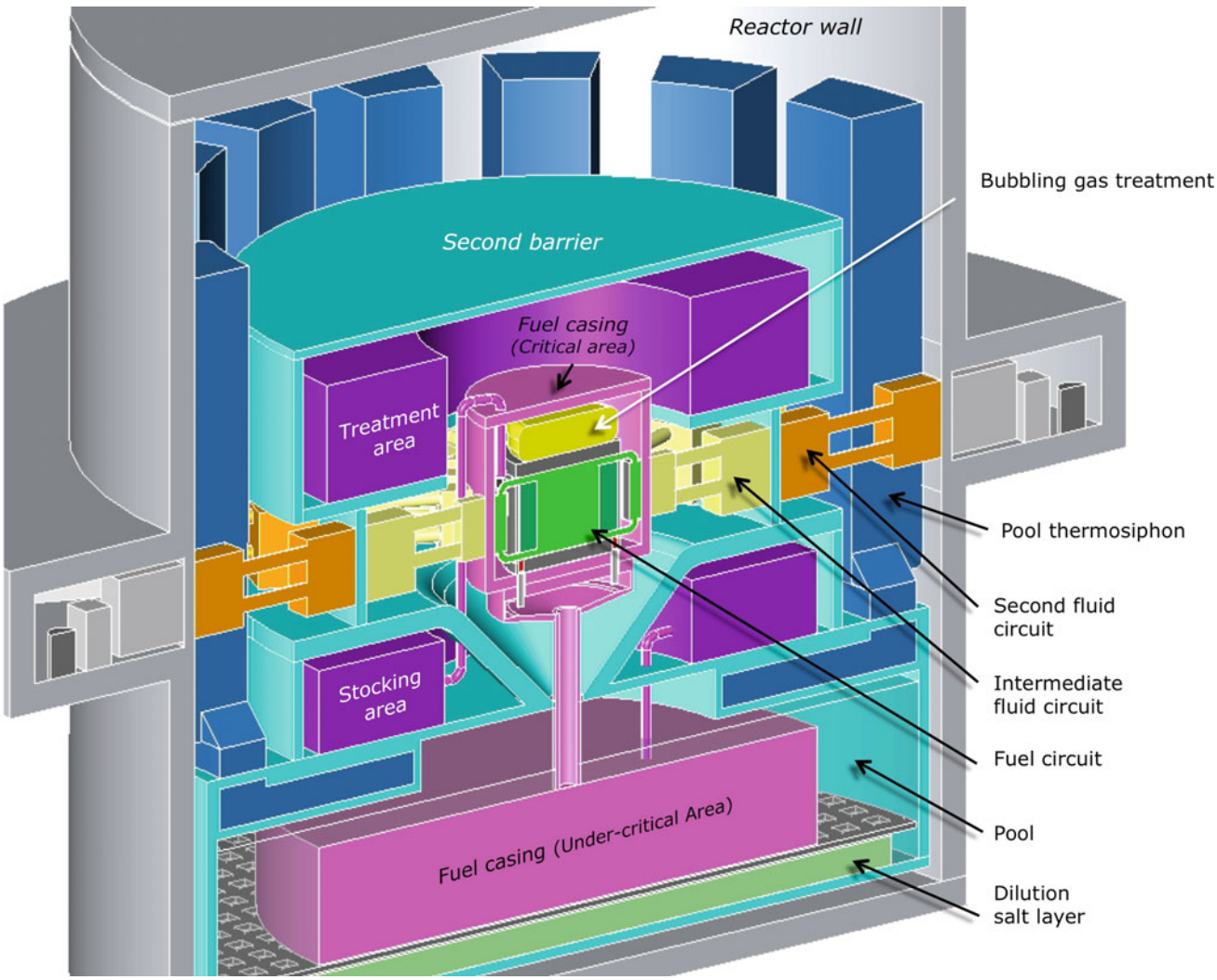

Fig. 2 Illustration of the main functions associated with the MSFR operation. In the middle is the *green* fuel salt circuit surrounded by a *pink* envelope representing the first confinement barrier. The *cyan* envelope represents the second barrier, including storing and chemical salt processing units in *violet*. The third barrier is in *gray*. Two heat transfer circuits between the three barriers are represented as loops in *yellow* and *orange*

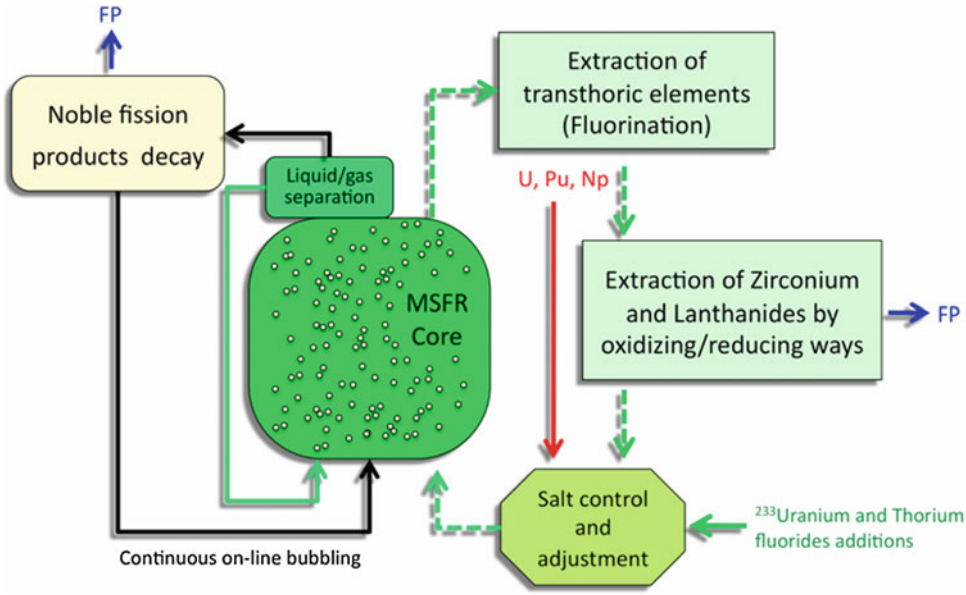

Fig. 3 Schematic representation of the fuel salt treatment with two loops

reactor and, thus, secure a rather small fissile inventory outside the core compared with present-day light water reactors (LWRs). The reprocessing rate itself is assumed to be equivalent to the present LWR rate; although, it could be possible to reprocess the fuel salt every ten years, but to the detriment of economical yield.

The salt treatment is schematically presented in Fig. 3. It consists of two circuits. One is a continuous gas bubbling in the core to extract the gaseous fission products (FP) and the metallic particles present in the salt (metallic FP and corrosion products). The gaseous stream is sent to a provisional storage area, where most of the Kr and Xe decay into Rb and Cs, preventing their accumulation in the fuel salt. The remaining gas is recycled.

On the left is the on-line treatment with gas bubbling in the core to extract noble gases and metallic particles (FP). On the right is the mini-batch on-site reprocessing with two objectives: removing FP (Zr, Ln) and adjusting the fuel content in fissile and fertile isotopes.

The other is a semi-continuous salt reprocessing at a rate of about 10 L per day to limit the lanthanide and Zr concentrations in the fuel salt. The sampled salt is returned to the reactor after purification and after addition of ^{233}U and Th as needed to adjust the fuel composition. This is also an opportunity to tune the oxide reduction potential of the salt by controlling the U^{4+} to U^{3+} ratio.

These two processes are aimed at keeping the liquid fuel salt in an efficient physical and chemical state for long time periods (decades). The gas bubbling has two objectives: removing metallic particles by capillarity (floating) and extracting gaseous fission products before they decay in the salt. The pyrochemical salt batch reprocessing avoids the accumulation of large quantities of lanthanides and zirconium in the fuel salt, a process that could be detrimental to several properties such as Pu solubility or salt volatility. Conversely to the thermal molten salt reactor, none of these processes are vital to the fast reactor operation. If they were interrupted for months or years, the MSFR would not stop, but it would have a poorer breeding ratio and could suffer from partial clogging of the heat exchangers, leading to poorer efficiency. The effect of the batch pyro processing rate is shown in Fig. 4. Notice that with the reactor configuration used for the calculation, the core is an under-breeder. Breeding is reached for the reprocessing of a full load up to 4000 days owing to the addition of the fertile blanket.

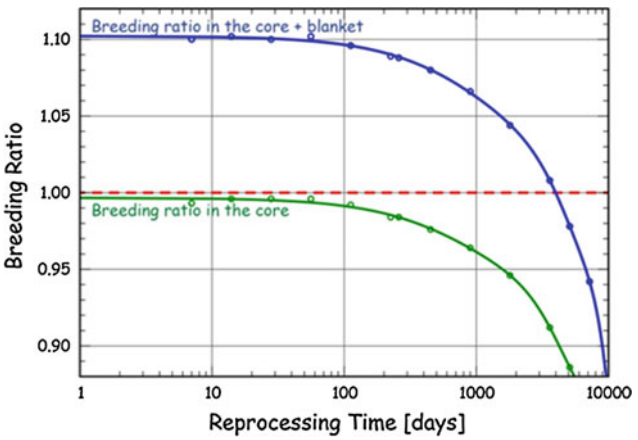

Fig. 4 Influence of the batch reprocessing rate on the breeding ratio in the core and in the whole MSFR system (core+fertile blanket)

MSFR Fuel Cycle Scenarios

To produce power, a fission nuclear reactor requires fissile material. Generation 2 or 3 reactors (PWR, CANDU, EPR), being under-breeder systems, that is, using more fissile material than they produce, need to be regularly re-fueled with fissile material throughout their operation time. Conversely, breeder generation IV reactors (SFR, MSFR, GFR) require only one (or two in the case of solid-fuel reactors) initial loading of fissile material. They then produce at least the amount of fissile material they need to be operated during their entire lifespan. Molten salt reactors require only one fissile load as no fuel re-fabrication is necessary and the fuel salt composition is controlled on-line without stopping reactor operation, whereas two loads are necessary for solid-fuel reactors, with one fissile load inside the reactor and the other in the reprocessing/fuel manufacturing process.

According to our simulations results, the thorium-based MSFR can be started with a variety of initial fissile loads [15, 16]:

- ^{235}U, the only natural fissile material on earth (0.72% of natural uranium). It can be used directly to start MSFRs with enriched uranium as the initial fissile material, with an enrichment ratio of less than 20% due to proliferation resistance issues;
- MSFRs can be directly started with ^{233}U as the initial fissile material, assuming that this ^{233}U can be produced in fertile blankets of other reactors (SFR, etc.) or by irradiating ^{232}Th in an accelerator-driven system (ADS), for example. Once an initial park of MSFRs based on the Th–^{233}U cycle is launched, ^{233}U will also be produced in MSFRs that are breeder reactors, allowing the deployment of such ^{233}U-started MSFRs in a second phase even if no ^{233}U is produced elsewhere;
- Using the plutonium produced in current pressurized water reactors (PWRs) or in future EPRs as the initial fissile material. An even better scenario would be the use of mixtures of TRU produced by these Generation II or III reactors.
- A mixture of these starting modes. For example, ^{233}U may be produced by using special devices containing thorium and Pu–MOX in cur-rent PWRs or in future EPRs.

Figures 5 and 6 present comparisons of fuel composition evolutions of a "3 GW_{th} reference MSFR" reactor started with ^{233}U, TRU, Th–MOX, or enriched U and TRU.

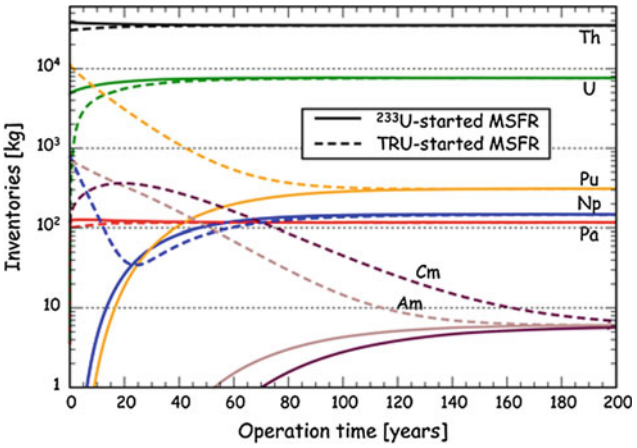

Fig. 5 Time evolution up to equilibrium of the heavy nuclei inventory for the ^{233}U-started MSFR (*solid lines*) and for the TRU-started MSFR (*dashed lines*). Operation time is given in equivalent full power years (EFPY) [14]

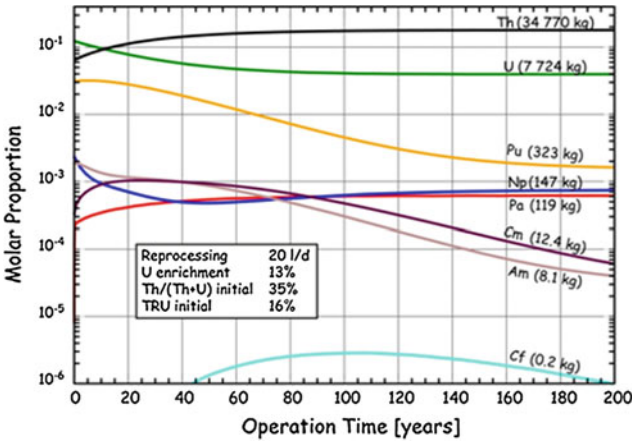

Fig. 6 Time evolution up to equilibrium of the heavy nuclei inventory for the optimized MSFR configuration started with enriched uranium and TRU elements. Operation time is given in EFPY

Safety Issues

A molten salt reactor has some specific safety features because the fuel salt geometry can be modified quickly and passively by draining to subcritical tanks. It is possible to design the system with a maximum of passive devices to cool the fuel in all circumstances and for long times without attendance. Moreover, for the MSFR, reactor stability is strengthened by its large negative feedback coefficients. Some of these features are discussed below, but not all safety provisions are detailed.

Safety Approach and Risk Analysis for a Liquid-Fueled Reactor

The unique characteristics of a liquid-fueled reactor strongly influence its design and safety analyses. For example:

1. The principle of defense in depth and multiple barriers has to be adapted as the conventional barriers (such as cladding, primary circuit and containment in a PWR) are no longer applicable;
2. Diversity and independence of the MSFR reactivity control mechanisms have to be demonstrated (no control or shutdown rods or burn-able poisons);
3. New safety criteria to evaluate reactor response during normal, incidental, and accidental conditions are needed as the MSFR fuel is in the liquid state, which is not an acceptable situation for the LWR fuel;
4. In the evaluation of severe accident scenarios with leakage to the environment, any interactions between the fuel salt and groundwater should be investigated in detail and the source term be determined;
5. Evaluation of the risk posed by the residual decay heat and the radioactive inventory in the reprocessing unit is also necessary.

A novel methodology for the design and safety evaluations of the MSFR is needed. Nevertheless, it would be desirable that the MSFR methodology rely on currently accepted safety principles such as the principle of defense in-depth, the use of multiple barriers, and the three basic safety functions: reactivity control, fuel cooling, and radioactive product confinement. In addition, owing to the limited amount of operation experience and some of its novel features, any new methodology should be robust and comprehensive, and integrate both deterministic and probabilistic approaches. To fulfill these objectives, a MSFR design and safety analysis methodology is currently being developed [17] according to the following steps:

1. Systemic modeling of all reactor components by using a model-based risk analysis tool;
2. Identification of the safety functions, to be defined from the components' functional criteria;
3. Identification of reactor abnormal events (including failure modes and dangerous phenomena);
4. Risk evaluation based on evaluation of probability and severity.

The design and safety criteria should ensure that all the reactor components adequately perform the safety functions in order to meet the requirements defined for each plants' operating conditions. With MSFR development being at its early stages, the idea is to adopt an inherent safety-by-design approach.

Decay Heat Removal

The decay heat generation versus time is represented in Fig. 7. Based on the concept described above, fission products are present in two different places when the reactor is stopped. Some are in the liquid fuel salt and some in the gas processing unit. About a third of the heat is produced in the gas processing unit and two thirds in the liquid fuel. The power of both heat sources decreases rapidly (by a factor of ten in about one day) from the value at shut down, which depends on the history of the power generation. The total amount of power at shut down is about 5 % of the nominal power. This value is lower compared with solid-fuel reactors because fission products are continuously removed in this concept.

In case of cooling problems, the fuel salt and the fluid containing fission products (salt or metal) of the gas processing unit can be drained into a subcritical tank placed in a water pool. A large amount of water is used as a decay heat thermal buffer so as to reduce the heat-to-cold-sink transfer rate by a factor of ten, for instance. This heat transfer is achieved by passive thermosiphons or heat pipes to the atmosphere through the reactor building walls (the third barrier). If unattended for a very long time, the fuel salt will solidify.

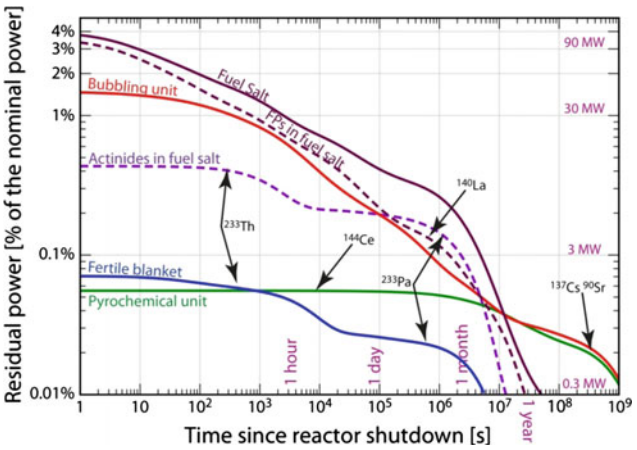

Fig. 7 Residual heat in the different radioactive fluids of the MSFR, after the total fission shut-down of the reactor previously under steady-state conditions [12, 17]

Issues and Demonstration Steps of the Concept Viability

Despite the status of preconception design of MSFRs, several limiting factors can be identified in the development of the concept.

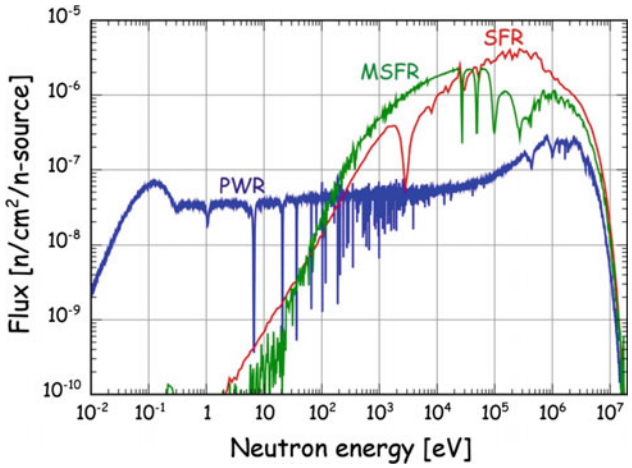

Fig. 8 Fast neutron spectra of the reference MSFR (*green curve*) and of a sodium-cooled fast neutron reactor (SFR, *red curve*) compared with the thermalized spectrum of a pressurized water reactor (PWR, *blue curve*) [14]

The first, obvious, issue is materials resistance to high temperatures, if the reactor is to be operated with a reasonably high power density. A first temperature limit is given by the fuel salt melting point (565 °C) to which a safety margin should be added to avoid local solidification (50 °C, for instance). To this, add 100–150 °C for in-core temperature heating corresponding to a salt circulation period of 3–4 s, so as to satisfy heat-transfer dynamics in the heat exchangers without incurring an excessive pressure drop within these. This leads to a temperature of about 750 °C at the core outlet to the gas–salt separation device and the pump (hot leg). Those devices may be maintained at 700 °C by cooling, that is, the same temperature as the heat exchanger plates during the heat transfer, the intermediate coolant salt being at about 650 °C. Although it seems that there are current alloys that can withstand such temperatures for a long time, this could still be a limit unless the material is replaced regularly, as is done with solid-fuel cladding.

The second issue is resistance to the neutron flux at high temperature, unless low power density operation is chosen. Calculations of the maximum displacement per atom (dpa) of the core walls yield 7.5 dpa/year for a power density of 330 W/cc. This is less than expected for solid-fuel fast reactors because of the neutron spectrum difference that is due to neutron inelastic scattering on fluorine nuclei, as shown in Fig. 8, and the absence of solid material in the core.

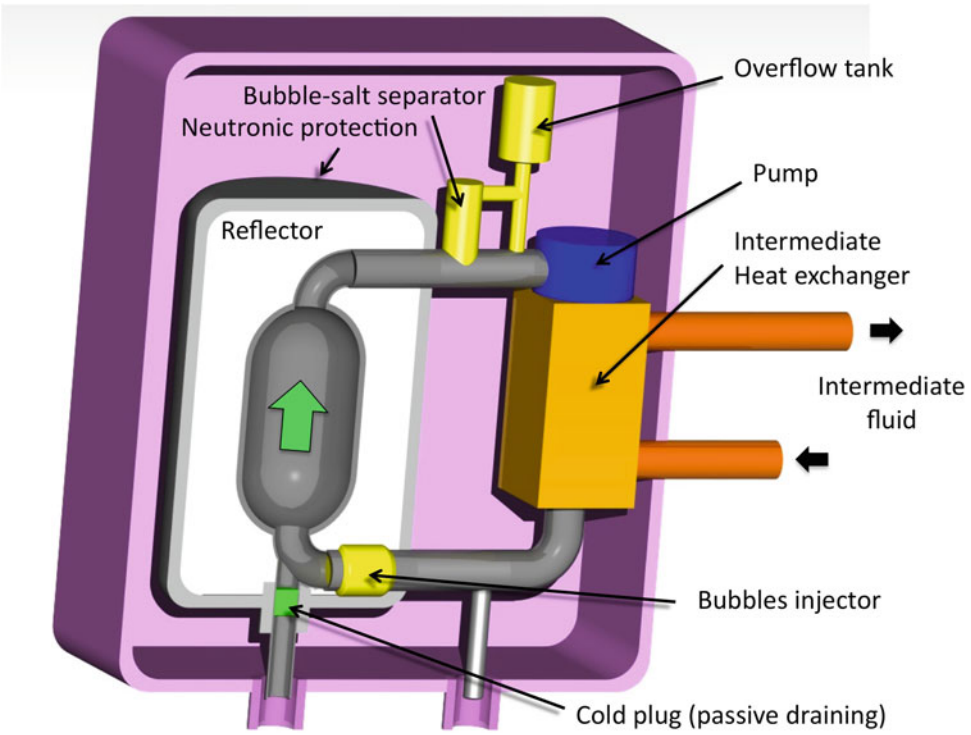

Fig. 9 Sketch of a single liquid fuel loop reactor for demonstration purposes or modular conception. The fuel volume (1.8 m^3) is reduced by a factor of ten from the 3 GW_{th} reactor and the power (200 MW_{th}) by a factor 15 in order to use the same intermediate heat exchanger

The third issue appears when trying to limit the per GW fissile inventory. This means restricting as much as possible the proportion of fuel salt out of the core, that is, in the tubing, pumps, and heat exchangers. It is technically challenging to reduce this "useless" amount of salt to less than 50 % of the total load and 30 % appears as a limit.

The fourth issue is a question more than a real limit: the safety evaluation. Indeed, present-day safety evaluation techniques are suitable for solid-fuel water reactors but partly irrelevant for liquid-fuel reactors. A new way of tackling the problem should find a consensus before any national safety authority can approve a liquid-fuel reactor design and this will take time and resources.

The size of the reactor liquid-fuel loop is not a limit, as shown by the calculation of a single-loop 200 MW_{th} reactor instead of a 16-loop 3 GW_{th} reactor. A low power demonstration version [18] is sketched in Fig. 9, but a regenerator version could be implemented by replacing the reflectors with a blanket. The size of this fuel loop assembly is about 2.5 m in diameter and 3 m high (core: 1.1 m diameter and 1.1 m high). The power is limited by the intermediate exchanger size, which is assumed to be the same as for the 3 GW_{th} reactor.

From the parametric studies that were carried out on the MSFR, no stumbling blocks appeared and the various limits can all be circumvented by reducing the power density.

Conclusion

Since 2005, R&D on molten salt reactors has been focused on fast spectrum concepts (such as the MSFR), which have been recognized as a long-term alternative to solid-fuel fast neutron reactors as MSFRs have attractive features such as very negative feedback coefficients, smaller fissile inventory, and a simplified fuel cycle. Experimental research on basic data is being conducted by a European network supported by EURATOM and ROSATOM to confirm the validity of the theoretical advantages of this concept. No insurmountable obstacles have been identified thus far, but almost all the technology remains to be tested, and demonstration experiments will have to be conducted to continue to assess the potential advantages of fast spectrum molten salt reactors, regardless of whether they are based on the thorium fuel cycle or are used as TRU burners.

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Part VI
Invited Speakers: Accelerator-Driven Systems

Accelerator-Driven Systems (ADS) Physics and Motivations

Jean-Pierre Revol

Abstract

The physics of Accelerator-Driven Systems (ADS) includes accelerator physics, the physics of neutron production by spallation from a particle beam, the physics of core nuclear reactions, as well as, the physics of subsystems such as heat extraction and production of electric power. This presentation will cover mainly the characteristics of the ADS starting from the particle beam providing the external neutron source to the nuclear reactor part. I will simply show how particle beam properties and the requirement for efficient electric power generation can be naturally used to define the parameters of such systems. I will only consider the case of a fast neutron core using thorium, since the main advantage of an ADS system is that it allows the use of thorium in a rather straightforward way. I will describe the basic differences between critical and subcritical (ADS) systems, and show that the physics of the system is naturally suited to the use of thorium as fuel, whether the system is optimized to destroy nuclear waste or to produce electric power. I will conclude that the physics of Accelerator-Driven Systems is well known, it has been simulated and checked in dedicated experiments, in particular at CERN with FEAT and TARC. These physics properties are used to design Accelerator-Driven Systems that are inherently safe.

A Brief History of ADS

The basic process in accelerator-driven systems (ADS) is nuclear transmutation, first observed by Ernest Rutherford in 1919 while bombarding nitrogen atoms with α particles, producing ^{17}O in the reaction: $^{14}N_7 + {}^4He_2 \rightarrow {}^{17}O_8 + {}^1p_1$. In his case, the particle accelerator was radioactive polonium (^{210}Po).

In 1929, a major step forward was achieved by Ernest Orlando Lawrence (Fig. 1), who designed the first cyclotron,[1] opening the prospect for intense, controlled particle beams.

In 1940, Ernest Orlando Lawrence in the USA and Nicolay Nicolayevich Semenov in the USSR independently proposed using a particle accelerator as a neutron source. The first practical realization, setting an important milestone, was in 1941, when Glenn T. Seaborg and co-workers produced the first μg of plutonium (^{238}Pu) by using the 60-inch cyclotron at U.C. Berkeley to bombard natural uranium (mostly ^{238}U) with 6 MeV deuterons.

Industrial-scale applications started in 1950, when Lawrence launched the Materials Testing Accelerator (MTA) project [1] at the World War II Naval Reserve Training Station at Livermore, California, now known as the Lawrence Livermore National Laboratory, to produce ^{239}Pu from depleted uranium. Two years later, in 1952, Wilfrid Bennett Lewis in Canada proposed using an accelerator to breed ^{233}U fuel from natural thorium for CANDU reactors [2]. This can be considered as the birth of the electro-breeder concept.

[1]Credit for the invention of the cyclotron also goes to Leo Szilard and Rolf Wideröe.

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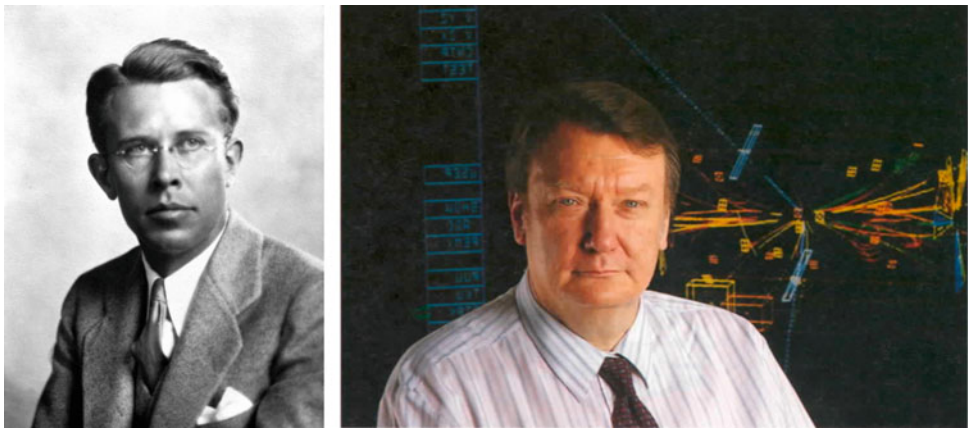

Fig. 1 *Left* Photo of Ernest Orlando Lawrence (freeinfosociety.com); *Right* Photo of Carlo Rubbia in front of a 1983 UA1 display of a proton–antiproton collision showing the first Z^0 particle ever observed. © P. Stumpf/SIPA Press

The MTA was dropped and Lewis' project was slowed down when rich uranium deposits were discovered in the United States, and it was also realized that it would require several hundred MW of beam power for these methods to be practical. At that time, as the aim was enrichment, there was essentially no energy gain in the system. Nowadays, ADS considered either for nuclear waste transmutation or energy production require typically ten times less beam power as nuclear fissions in the core are used to produce neutrons in addition to the neutrons provided by the accelerator.

In the 1960s, Lawrence and collaborators patented the Electronuclear Reactor to provide adequate quantities of materials such as ^{239}Pu and ^{233}U, which can only be produced artificially. This concept of accelerator breeding was also studied by Russian scientists in the 1970s. A neutron yield experiment [3] with depleted uranium was carried out by using the Dubna accelerator. Studies of fertile-to-fissile conversion (FERFICON) also took place in the late 1970s at Los Alamos [4].

Renewed interest in ADS arose again in the 1980s and 1990s, when the USA slowed down the development of fast critical reactors (the Fast Flux Test Facility at Argonne National Laboratory was stopped in 1993) and when it was realized that accelerator technology had made significant progress since Lawrence's time. More realistic concepts of ADS, taking safety issues into account, were developed:

- Hiroshi Takahashi at Brookhaven National Laboratory submitted several proposals for ADS. One of these was the PHOENIX project [5], the purpose of which was to transmute nuclear waste, including minor actinides, with a fast neutron ADS and a high neutron multiplication factor, $k \approx 0.99$;
- Charles D. Bowman at Los Alamos proposed a thermal neutron ADS for the Accelerator Transmutation of nuclear Waste (ATW) with molten thorium-based fuel and chemical on-line extraction of fission fragments and of protactinium (^{233}Pa) [6];
- Japan launched Options for Making Extra Gains from Actinides (OMEGA) [7, 8] at the Japan Atomic Energy Research Institute (JAERI). Today, this program is continued as the Japan Proton Accelerator Research Complex (J-PARC) at Japan Atomic Energy Agency (JAEA), with the main purpose of burning minor actinides.

Major progress was made in the 1990s, when Carlo Rubbia (Fig. 1) became convinced that not only was accelerator technology then mature for a realistic exploitation of the ADS idea, but that new tools existed for optimizing the parameters of such systems. He launched a vigorous research program at CERN based on three main elements:

- Development of an innovative simulation, inherited from particle physics, including the spallation process initiated by a proton hitting a lead or uranium target, producing neutrons that are transported throughout the system, and describing the fuel evolution as a function of burnup;
- Specific experiments at the CERN Proton Synchrotron (PS) to validate basic physics concepts (FEAT [9] and TARC [10]);
- Design, construction, and exploitation of an advanced neutron time-of-flight facility (n_TOF) [11–13] at CERN to acquire precise neutron cross-section data, which is crucial to reliably simulate any advanced reactor configuration built out of new materials. The remarkable performance of n_TOF is the subject of the presentation at this workshop by Frank Gunsing [14].

C. Rubbia named his accelerator-driven system "Energy Amplifier" (EA) [15, 16], as the energy provided by the proton beam appears to be "amplified" through fission reactions in the system's core. His work at CERN triggered a

major R&D wave on ADS in Europe [17] and in the entire world, as shown in this conference.

Basic Elements of ADS

An accelerator-driven system is conceptually simple and consists of two basic elements (Fig. 2):

- A particle accelerator driving a neutron source, which is needed to sustain fission reactions in a subcritical core;
- A core in which both source and fission neutrons are at work.

Note that the spallation neutron energy spectrum is on average not very different from the energy spectrum of fission neutrons (Fig. 3), except for a high neutron energy tail in the spallation spectrum, which turns out to play a visible role in terms of neutron balance, owing to (n, *X*n) reactions on the target and moderator material.

The main areas of physics relevant to such systems, the physics of neutron production by spallation from a particle beam hitting a target and the physics of neutron interaction and transport around the spallation region and in the core, are well understood and can be simulated with precision.

For a practical system, physics also drives the choice of other ADS elements, such as the moderator to obtain an adequate neutron energy spectrum, heat extraction and cooling in general, and the conversion of heat into electric energy, which requires the highest possible operating temperature to improve the efficiency compared with pressurized water reactors.

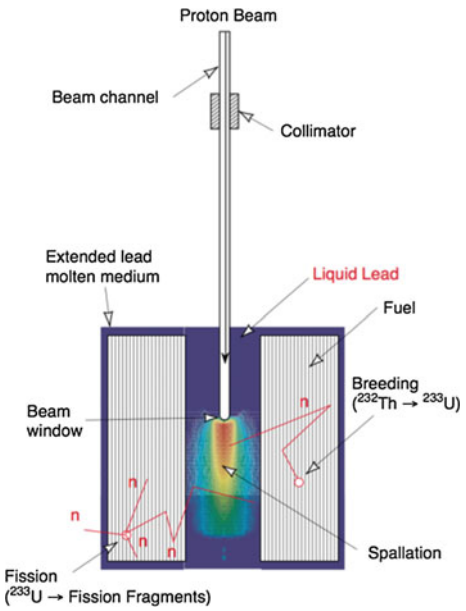

Fig. 2 Schematics of the basic elements of an accelerator-driven system: the proton beam is inserted vertically from the top, neutrons produced by spallation off a target (molten lead in this case) are multiplied through fission in the core surrounding the spallation area, where breeding of ^{233}U takes place

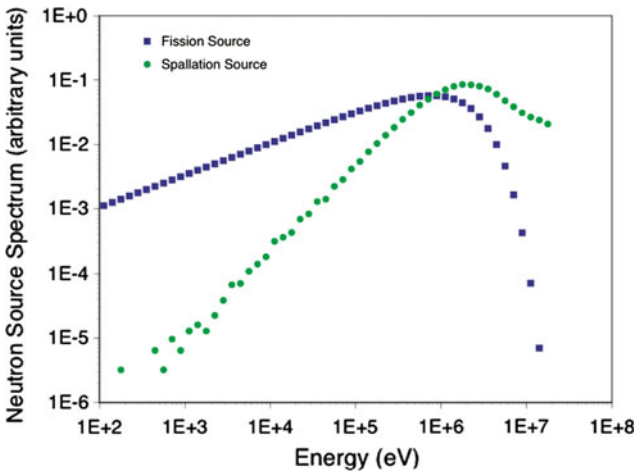

Fig. 3 Comparison of fission and typical spallation neutron energy distributions. The spallation neutron energy spectrum shown here for incident 1 GeV/c protons is characterized mainly by a more pronounced high-energy tail compared with the fission spectrum

Subcritical Core Physics

The cost to pay for running a subcritical system is the addition of an accelerator and its associated neutron source. However, the reward is that the presence of the accelerator allows a great flexibility in the choice of fuel, and in particular, ADS are well suited to the use of thorium-based fuel.

The theory of subcritical systems is interesting in itself and provides insight into their physical properties, which are quite different from those of critical systems. In particular, their response to fast reactivity changes is spectacularly slower and more moderate than in critical systems [15, 16]. Knowledge of the neutron flux geometry is important for calculating the generated power distribution and optimizing the uniformity of fuel burnup. Even though it is nowadays possible to simulate extremely complex systems precisely with Monte Carlo methods, an analytical approach can be used with some simplified assumptions to extract the general properties of such systems and compare with critical systems (for instance, see [18, 19]).

If one assumes a theoretical system, with uniform material distribution, monoenergetic neutrons, and small absorption, such systems can be represented by a simple equation similar to that describing a critical system, but with the addition of an external neutron source term, $C(\vec{r}, t)$, representing neutrons provided by the accelerator:

$$\frac{\partial n(\vec{r},t)}{\partial t} = \nu\,\Sigma_{\mathrm{f}}\,\Phi(\vec{r},t) + C(\vec{r},t) - \Sigma_{\mathrm{a}}\,\Phi(\vec{r},t) + D\,\nabla^2\,\Phi(\vec{r},t)$$

The rate of change of n, the number of neutrons, as a function of space ($\vec{r}$) and time (t) is given by the difference between the rate of neutrons provided by fissions and by the accelerator and the rate of neutrons lost through absorption (fission and capture) and leakage outside the geometrical volume considered. In the above equation, ν is the number of neutrons produced per fission, Σ_{f} and Σ_{a} are the neutron fission and absorption macroscopic cross sections, Φ is the neutron flux, C is the spallation neutron source term, D is the neutron diffusion coefficient, and ∇^2 is the Laplacian. It is interesting to consider such a system at equilibrium:

$$\frac{\partial n(\vec{r},t)}{\partial t} = 0 \Rightarrow \nabla^2\,\Phi(\vec{r},t) + \frac{(k_\infty - 1)}{L_{\mathrm{C}}^2}\,\Phi(\vec{r},t) = -\frac{C}{D}$$

with $k_\infty \equiv \nu\Sigma_{\mathrm{f}}/\Sigma_{\mathrm{a}}$ and where L_{C}, the neutron diffusion length, is given by $L_{\mathrm{C}}^2 \equiv D/\Sigma_{\mathrm{a}}$. There exist two very different regimes corresponding to the two possible signs of the factor $(k_\infty - 1)$:

- $k_\infty < 1$, the system is intrinsically subcritical, as was the case in the FEAT experiment [9] at CERN, with $k_\infty \approx 0.93$. The solution is an exponential function (Fig. 4).
- $k_\infty > 1$, the subcriticality of the system comes from the lack of neutron confinement, it is a geometrical issue. The solution is oscillatory. This is, for instance, the case of Carlo Rubbia's Energy Amplifier, with $k_\infty \approx 1.2$–1.3 [15, 16]. In this case, one can expand the neutron external source term and the neutron flux function, over the complete set of solutions ($\psi_{l,m,n}$) of the kernel equation:

$$\nabla^2\,\Phi(\vec{r},t) + B^2\Phi(\vec{r},t) = 0$$

where

$$B^2 \equiv \frac{(k_\infty - 1)}{L_C^2}$$

Defining the expansion coefficients, $\mathrm{C}_{l,m,n}$ and $\Phi_{l,m,n}$, in the following way:

$$C(\vec{r}) = D\sum_{l,m,n} C_{l,m,n}\psi_{l,m,n}(\vec{r}); \quad \Phi(\vec{r}) = \sum_{l,m,n} \Phi_{l,m,n}\psi_{l,m,n}(\vec{r})$$

one obtains the general solution for the neutron flux, upon substitution of the above expressions into the equilibrium equation:

$$\Phi(\vec{r}) = L_{\mathrm{C}}^2 \sum_{l,m,n} \frac{C_{l,m,n}}{1 - k_{l,m,n}}\psi_{l,m,n}(\vec{r}); \quad k_{l,m,n} \equiv k_\infty - L_{\mathrm{C}}^2 B_{l,m,n}^2$$

where $B_{l,m,n}^2$ are the eigenvalues corresponding to the eigenfunctions ($\psi_{l,m,n}$).

Unlike critical systems in which only the fundamental mode is present, in the ADS case, all the modes are excited.

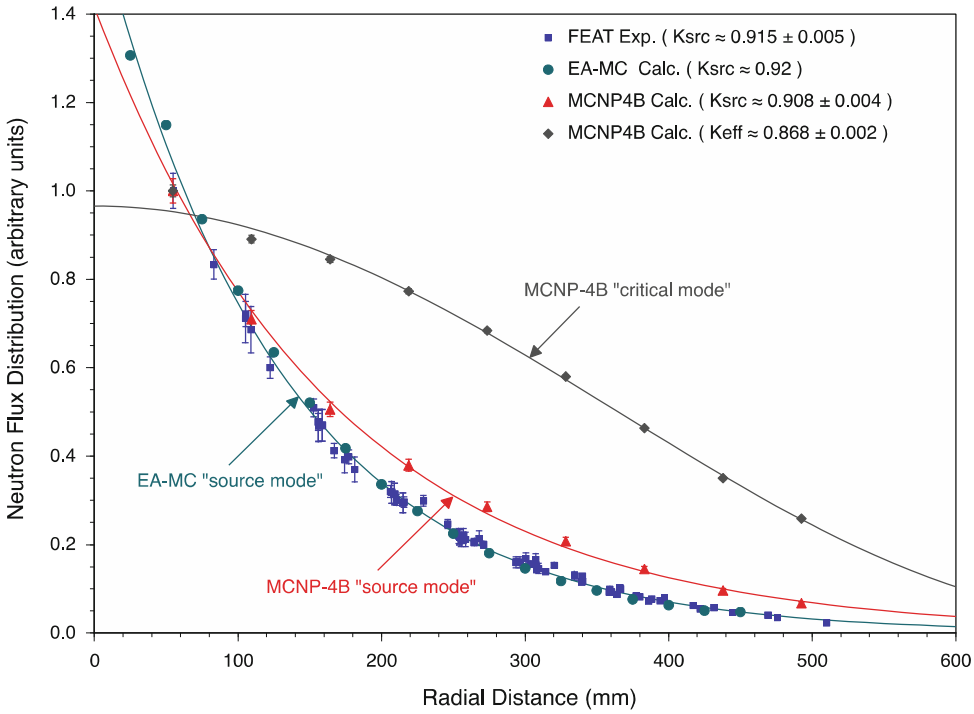

Fig. 4 Neutron flux measured in the FEAT experiment [9] as a function of the radial distance to the beam line, illustrating the exponential behavior of the corresponding solution to the flux equation with $k_\infty < 1$

A general theorem can be easily proven: if the fundamental mode is subcritical, then all the other modes are also subcritical. This is obviously an important and reassuring property of subcritical systems.

An interesting property of subcritical systems is that the neutron multiplication factor changes depending on whether the accelerator is on ($k = k_{source} \equiv k_s$) or off ($k = k_{eff}$). When the accelerator is off, the definition of the neutron multiplication coefficient is similar to that of a nuclear critical reactor:

$$k_{eff} \equiv \frac{\nu\Sigma_f}{\Sigma_a + DB^2}$$

However, when the accelerator is turned on, the presence of the external neutron source and of non-fission neutron multiplication through (n, *X*n) reactions on lead, allowed by the hard energy spectrum of spallation neutrons (Fig. 3) modifies, the neutron multiplication factor:

$$k_s \equiv \frac{\nu\Sigma_f\Phi(\vec{r},t) + C(\vec{r},t)}{\Sigma_a\Phi(\vec{r},t) - D\nabla^2\Phi(\vec{r},t)}$$

The change of geometry of the neutron flux between the fundamental mode, characterizing k_{eff}, and all the modes excited by the source, characterizing k_s, also plays a role in the determination of the neutron multiplication factor. As a consequence, the good news is that k_{eff} is always smaller than k_s, which means that switching off the accelerator, hence the neutron source, not only stops the main power generation, but also moves the system further away from prompt criticality, back to the k_{eff} value. This is an additional safety asset for ADS, for which safety relies on the physical properties of the system as opposed to human control in critical reactors.

Accelerators for ADS

The required accelerator power (P_{beam}) can be expressed as a function of the desired ADS fission power (P_{ADS}), the neutron multiplication factor k_s, and G_0, a constant that depends on the beam energy, the target material, and the detailed geometry of the system:

$$P_{beam} = \frac{(1 - k_s)}{k_s G_0} P_{ADS}$$

Therefore, the choice of the accelerator power is a trade-off between accelerator power and criticality margin (Fig. 5). For given k_s and G_0 values, the ADS power changes linearly with the beam power, allowing the possibility of modulating the power output with the accelerator. This could be a useful feature if ADS were to be associated with fluctuating renewable energy sources.

Compared with uranium, the neutronics with thorium is favorable for ADS power modulation because of the much longer half-life of ^{233}Pa (27 days) compared with ^{239}Np (2.3 days). A problem with the use of thorium in critical reactors may thus become an advantage in the case of ADS.

The Paul Scherrer Institute (PSI) separate-turns cyclotron has achieved a beam power of 1.4 MW [20] (2.4 mA, with 0.59 GeV protons). Such a beam would produce a power P_{ADS} of 130 MW_{th} with k_s = 0.98, if driving an Energy Amplifier as designed by Rubbia [15, 16].

Accelerator Requirements

In principle, it does not matter how the external neutron source is provided. In practice, for industrial applications, there are a number of well-defined requirements for the accelerator:

Beam particle: The preferred choice is protons for their simplicity of production and because they are efficient in producing neutrons by spallation.

Beam energy: Optimum neutron production is obtained for $E_{beam} \geq 900$ MeV, as shown by the FEAT experiment [9]. At lower energy, protons tend to lose more energy by ionization, which does not produce neutrons. Above about 900 MeV, the energy gain shows a plateau. It then slowly decreases with further increases of the beam energy, as pion production increases.

Beam power: The beam power for industrial applications should be typically in the range 1–10 MW, depending on the choice of the k_s value and on the desired power output. A large operational range of beam intensities might be required to follow electrical power demand. The maximum range depends on the core design. Beam power stability is clearly important, as the thermal power output is proportional to the beam power.

Beam spot size (footprint): In a system with a window separating the accelerator vacuum chamber from the spallation target, the beam spot size should be large at the point of impact on the window. Studies at JAEA have shown that densities up to 0.1–0.2 mA/cm^2 are currently achievable [22]. For MYRRHA [21], the design value is 0.07 mA/cm^2.

Beam losses: These have to be controlled so as to minimize irradiation of the accelerator elements and of the environment. For linear accelerators (linacs), the beam losses are spread all along the accelerator structure and the upper limit currently accepted is 1 W/m. For cyclotrons, beam losses are localized at injection (low energy) and at extraction. Beam losses have to be minimized and taken into account for any high-power beam, not only for ADS. They have a direct impact on maintenance and repair.

Reliability: Fatigue of mechanical structures, in particular of solid fuel elements, requires the minimization of beam

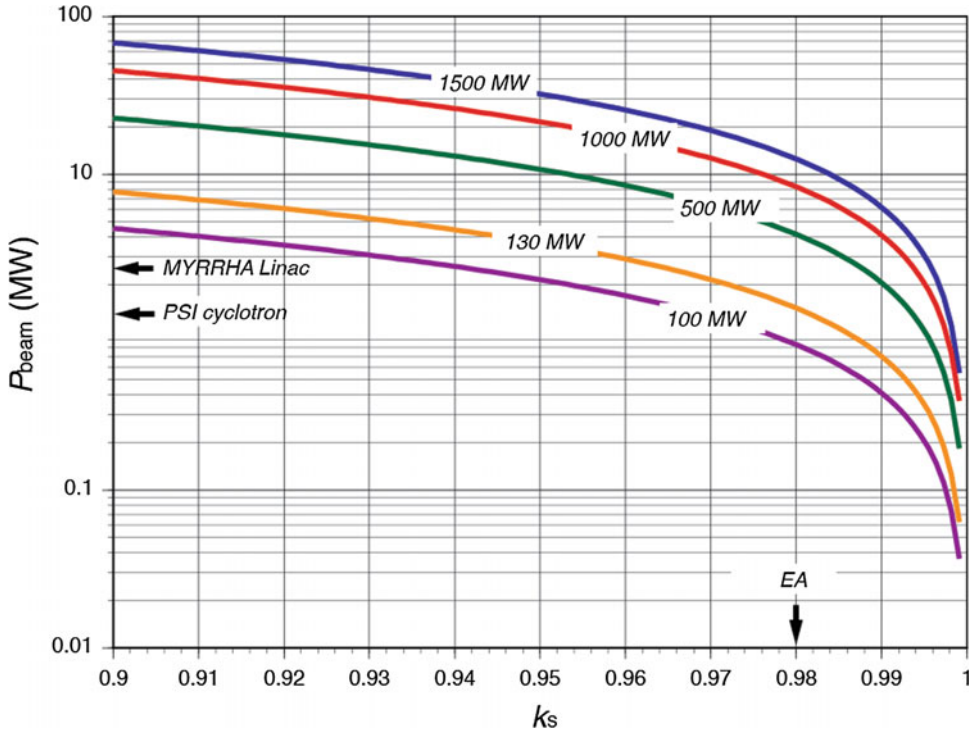

Fig. 5 Beam power versus k_s for 1 GeV/c protons. Curves of constant fission power are shown, labeled in MW_{th}. The k_s choice in C. Rubbia's Energy Amplifier [15, 16] is indicated, as well as the beam power achieved with the PSI cyclotron [20] and that planned for MYRRHA [21]. The 130 MW curve corresponds to 0.6 GeV/c protons used in the MYRRHA linear accelerator and in the PSI cyclotron

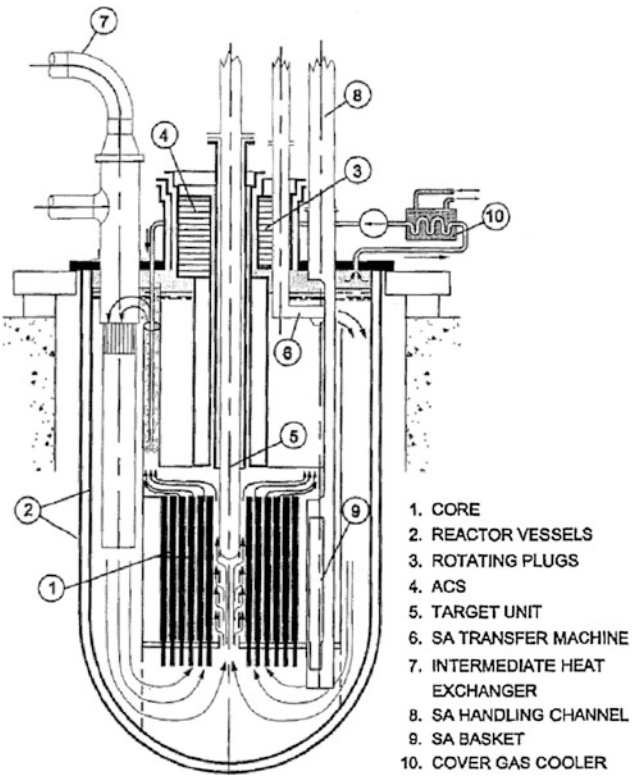

Fig. 6 Ansaldo engineering design for EU proposal by Carlo Rubbia [23]

trips. In the case of MYRRHA, there are no constraints on the number of trips of duration (t_{trip}) of less than 0.1 s. However, no more than 100 trips per day with 0.1 s < t_{trip} < 3 s, and 10 trips in three months with t_{trip} > 3 s are allowed. Such constrains may evolve with the development of better materials or can be relaxed in systems with molten fuel.

Energy efficiency: One must maximize the accelerator electric power efficiency, $\eta = P_{beam}/P_{grid}$. This is relevant to the overall energy efficiency of the system.

Size of accelerator: This might be a feature in favor of cyclotrons as, for nuclear waste elimination, one might prefer to have the accelerator fit within the site of a nuclear power plant to avoid transportation of nuclear waste.

Cost: This is clearly a very important element to be taken into account.

In the end, the solution that is chosen from among the linac, cyclotron, or even fixed-field alternating gradient accelerator (FFAG) technologies, will be the one best fulfilling all of the above requirements.

The accelerator community has always assumed that an adequate accelerator can be built. However, a main criticism of ADS has been that "the accelerator does not exist and will be too expensive". This is obviously a challenge to be taken on by the accelerator community: to build an accelerator able

to deliver a multi MW proton beam with the required characteristics.

ADS Proposals

The first significant ADS proposal in Europe [23] (Fig. 6) was submitted to the European Union in 1999 by Carlo Rubbia in cooperation with ANSALDO, an Italian power engineering company, but was rejected, which retrospectively appears as an unfortunate strategic mistake as Europe would have today 15 years of experience on the industrialization of ADS systems. As director of the Italian ENEA, Carlo Rubbia prepared another proposal named TRADE for "TRiga Accelerator Driven Experiment" [24], based on an existing TRIGA research reactor. In the end, it was not submitted to the EU. Europe, where some of the best experience in nuclear reactors is available, missed an opportunity to take the lead in the development of ADS.

Summary of Recent ADS Developments

Even though R&D on ADS is certainly not at the level required by the increasing importance of current energy issues, a significant R&D effort is taking place worldwide, motivated by the great energy challenge facing developing countries, which are likely to take the lead in this domain.

The PSI cyclotron beam has already reached the power range for industrial applications (1.4 MW) [20]. MEGAPIE [25], the world's first megawatt spallation neutron source ran successfully for three months in 2006 at SINQ, the Swiss Spallation Neutron Source. Currently, MEGAPIE successors operate beyond 1 MW, such as SNS, the 1.4 MW US Spallation Neutron Source [26], at Oakridge National Laboratory.

The MYRRHA project [21] at SCK•CEN, in Mol, Belgium, could be the first ADS prototype of significant power, if funded. It will use a 2.4 MW linac with 0.6 GeV protons. However, one could regret that thorium is not on the agenda and that the system will be transformed into a critical research reactor after only a few years of operation as an ADS.

In Kharkov, Ukraine, the KIPT neutron source [27] has been completed and licensed with low enriched uranium (LEU), but is not yet operational. It is based on the ADS concept with an electron accelerator driving a 100 kW neutron source and with a rather small multiplication factor of 2 for the LEU core.

At Troitsk [28], Russia, and at the Institute of Modern Physics, China (CADS), [29] ADS is being considered for burning minor actinides, and a discussion in India is taking place to use ADS [30] to simplify the Indian thorium utilization scheme [31].

As a consequence of the Fukushima accident, Japan recently re-launched the transmutation experimental facility program at J-PARC, which is now part of the roadmap toward the ADS proposed by JAEA for nuclear waste transmutation. It consists of two activities, under J-PARC:

- Development of the ADS Target Test Facility (TEF-T) to verify the feasibility of the beam window, a challenge for ADS and to consider ADS as a material test facility;
- Further development of the Transmutation Physics Experimental Facility (TEF-P) to study reactor physics issues of subcritical cores and minor actinide loaded cores.

Other ADS concepts were presented at the ThEC13 Conference in Geneva, including molten salt ADS by Rubbia [32], Pyeon (Japan) [33], and Chai (Korea) [34]. Using a molten fuel developed for molten salt reactors (MSRs) could help relax requirements on the accelerator in terms of the number of power trips. There are also relevant studies on corrosion with high-temperature lead or lead–bismuth eutectic mixture, which have resulted in the production of new materials resistant to corrosion in molten lead up to about 550 °C. For instance, reduced activation ferritic martensitic steels, which also have applications in fusion technologies.

Conclusion

The physics of ADS is well known and reliable simulation is available. ADS may offer the best way of using thorium in a nuclear fission system. When taking into account the need for safety, waste management, and non-proliferation, thorium in a fast neutron ADS is very attractive. ADS represent a challenging innovation but there is no roadblock, as the considerable amount of R&D world-wide shows. There is no reason a "demonstrator" of significant power cannot be built, in order to prepare for the industrialization of ADS. Hopefully, this can be achieved soon, preferably by an international collaboration, as the global energy challenge should be surmounted by global responses.

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Review of Accelerators for Accelerator-Driven Systems (ADS)

Alex C. Mueller

Abstract

The present contribution reviews different types of accelerator concepts from a generic point of view. Basic principles of physics and common-sense engineering arguments will be used to investigate what kind of concepts can provide a sound footing for accelerator-driven system (ADS) applications including, but not limited to, thorium-ADS. In particular, we shall revisit the 2002 findings of the OECD-NEA and the 10 years of R&D that have been taken place since, as well as considering the choice of other recent projects that exhibit certain similarities to ADS (neutron spallation sources, radioactive beam and neutrino factories, etc.).

Introduction

As can be seen from the well-known Livingston diagram, Fig. 1, different types of accelerators do not compete too much with each other for a given specification, in this case, the energy. In other words, there is an optimal range for a given concept. This observation quite often holds true for a number of other parameters too.

Underlying to this are the basic principles of beam physics, such as Liouville's theorem, Einstein's relativistic properties (scaling with the factor $\beta = v/c$, which compares the speed, v, of the accelerated particles to that of photons), or the repulsive Coulomb interaction between the charged particles that constitute the beam. Such facts are well discussed in classical books, such as, for example, those in [1–3].

For a given application, it is, therefore, best to first closely examine the desired specifications for the particle beam and then choose the accelerator that may match them "intrinsically" with some ease. This accelerator physics approach is in clear opposition to a more engineering approach, where one tries to shoehorn the requirements into one's, for whatever good or bad reason, beloved machine concept.

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With these caveats in mind, we shall examine in the following sections, in a very generic way, machine concepts for accelerator-driven systems (ADS).

ADS Accelerator Specifications

Basic ADS Characteristic

By somewhat simplifying, we may distinguish between three different types of ADS with regard to the power developed in the subcritical core.

These are:

(1) "Zero-power" experimental devices (P_{therm} < several kW);
(2) Concept demonstrators (P_{therm} < 100 MW);
(3) "Industrial" machines (P_{therm} > 250 MW).

As a mature example of (1), one may consider GUINEVERE at Mol, Belgium. This has been built by SCK·CEN, CNRS-IN2P3, and the Commissariat à l'énergie atomique et aux énergies alternatives (CEA). It is now regularly used by an international collaboration [7].

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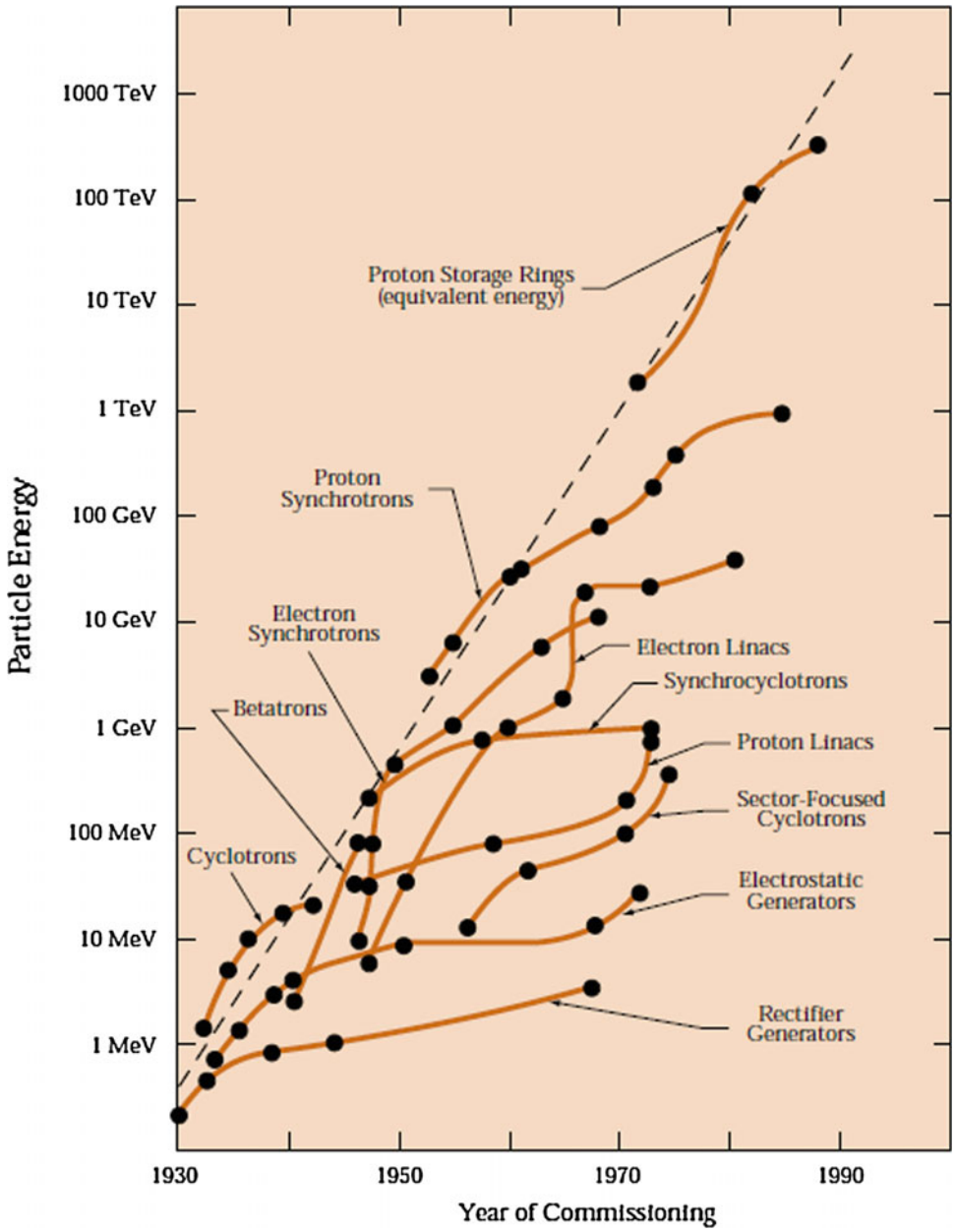

Fig. 1 Livingston chart for accelerators [4]. The version shown here [5] was updated in the 1990s. An up-to-date version, including, for example, the Large Hadron Collider (LHC) can be found in [6] or by a search on the Web

Table 1 compares some characteristics and specifications between ADS of types (2) and (3).

For the second category, various projects have been reported in the past at conferences in the field [8, 9], the most advanced of these is MYHRRA of the SCK, infrastructure of the European Strategy Forum on Research Infrastructures list, and already partially financed by commitment of the Belgian government. MYRRHA is described, with many details, in the present proceedings [10] along with specific aspects for the development of its accelerator [11].

It is, therefore, understandable that only conceptual studies have been reported for an industrial transmutation device, which would require the return of experience from a demonstrator of a more advanced stage.

Basic Accelerator Considerations

Energy

Already related to the beam energy, the Livingston chart gives an ab initio impression of what kind of accelerators could be considered: type (2) and (3) ADS, as described in the preceding section, require spallation neutron sources, induced by protons exceeding 500 MeV or so; hence, synchrotrons, linear accelerators (linacs), cyclotrons, and synchrocyclotrons could be chosen.

The much lower neutron flux for class (1) experiments can be generated by a low-energy nuclear reaction, such as

Table 1 Typical specifications for ADS of category, (2) and (3) (see text)

	XT-ADS (ADS prototype; type 2)	EFIT (Industrial transmuter; type 3)
Goals	Demonstrate the concept Demonstrate transmutation Provide an irradiation facility	Maximise the transmutation efficiency Ease of operation and maintenance High level of availability
Main features	50–100 MW_{th} power	Several 100 MW_{th} power
	k_{eff} around 0.95	k_{eff} around 0.97
	600 MeV	800 MeV
	2.5 mA proton beam	20 mA proton beam
	Conventional MOX fuel, test locations for MA	Minor actinide fuel
	Eutectic lead–bismuth for coolant and target	Lead coolant and target

The table was extracted from the EUROTRANS study. k_{eff} refers to the (effective) criticality factor as defined in reactor physics for an assembly of fissioning material; note the considerable subcriticality

by a 250 keV deuteron beam impinging on a tritium target. Here, according to the Livingston chart, a rectifier generator is clearly sufficient, for example.

Beam Intensity

According to Table 1, the required proton beam intensities for demonstrators have to exceed several mA, and those of industrial machines require an additional order of magnitude. From that consideration, one immediately can rule out synchrotrons and synchrocyclotrons.

Indeed, conceptually, they are pulsed machines (ramping of magnetic field for the former and of the radiofrequency for latter) and, therefore, exhibit a low duty factor. Thus, although the instantaneous circulating current in a synchrotron may be in the range of Amperes, owing to its strong focusing properties, the average extracted-to-a-target current is generally on the order of μA.

Reliability

A new (and historically ADS-specific) requirement is the one for "reliability". What is meant here is an extreme absence of beam "trips" that last longer than a second or so.

Indeed, frequently repeated beam interruptions induce thermal stress and fatigue on the reactor structures, the target or the fuel elements, with possible significant damages, especially on the fuel claddings. Moreover, these beam interruptions decrease the plant availability, resulting in plant shut-downs in most cases. Therefore, it has been estimated that beam trips in excess of one second duration should not occur more frequently than five times per three-month operation period for a demonstrator category ADS, and three times per year for an industrial one.

The OECD-NEA (Nuclear Energy Agency) Assessment

Based on such considerations and on the experience with existing accelerators, those under construction or planned at the time, a statement was published by an expert working group meeting on the occasion of the OECD-NEA workshop in 2002 [8]. One may quote in particular:

- Cyclotrons of the Paul Scherrer Institute (PSI) type should be considered as the natural and cost-effective choice for preliminary low-power experiments where availability and reliability requirements are less stringent.
- Continuous wave (CW) linear accelerators must be chosen for demonstration facilities and full-scale plants, because of their high value potential regarding availability, reliability, and power upgrade capability.

In the following section, we shall reconsider this statement, which is over a decade old, and take into account the new high-power proton accelerators that, in the meantime, have been planned, are under construction, or are even running.

Linear Accelerators and Cyclotrons for ADS

Cyclotrons

In the "classical" cyclotron, the charged particles move in a constant and perpendicular magnetic field. For a given mass and charge they, thus, exhibit a constant revolution frequency, independent of their energy as can be deduced from the Lorentz force. Thus, one can place very few (and in extreme cases, a single) accelerating cavities inside the cyclotron magnet, and these cavities act over the whole turn

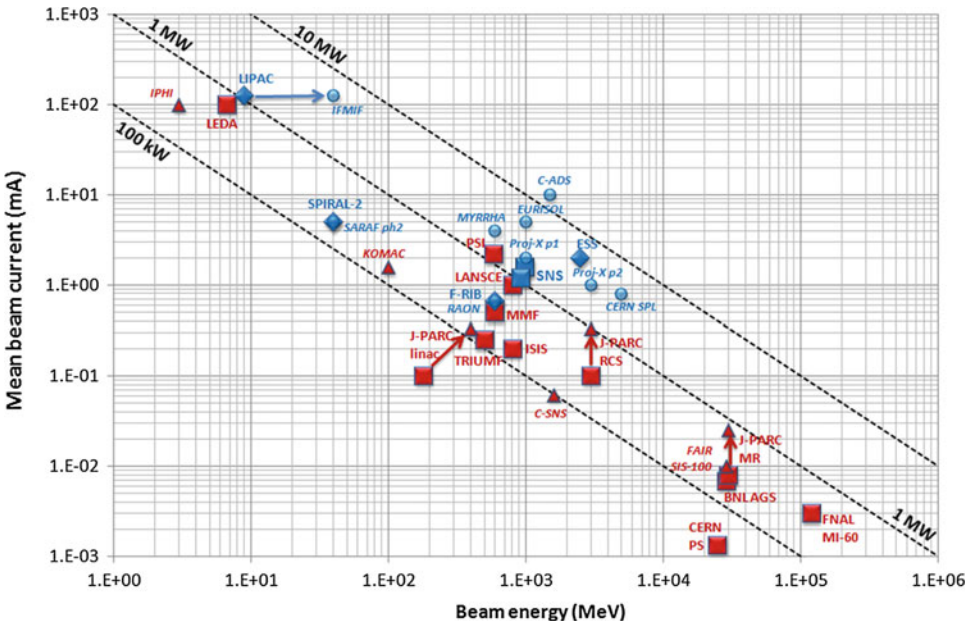

Fig. 2 Intensity versus energy plot for high-power accelerators

radius, which increases with particle energy. Driven by a radio-frequency (RF) that corresponds to a harmonic of the revolution frequency, the beam is, thus, macroscopically of CW nature as acceleration occurs simultaneously at all radii.

Clearly, this is a priori the most compact and cost-efficient layout and is further characterized by a good conversion efficiency of the RF to the beam power. At this stage, the main drawback is the absence of any strong vertical focusing, for example, through quadrupoles along the rather long (typically a few hundred meters) spiraling beam path, a drawback that provides a hard constraint on the possible intensities, limiting the beam to a few hundred μA.

For protons, as the energy rises above about 20 MeV, the simple frequency correspondence breaks down owing to the relativistic particle mass increase. Thus, the particles are no longer accelerated. To address the issue, high-energy cyclotrons are being designed so that the average magnetic field along the orbital path accordingly increases for a constant revolution frequency. In practice, this considerably increases the sophistication of the machine design and has become the domain of a small number of renowned experts. Without going into detail, we will just note that the "isochronous" cyclotron generally consists of a number of (typically spiral-shaped) sector magnets that may have a series of trim coils and pole gaps of radially-dependent width. The RF cavities can now be inserted between the cavities, along with devices for beam extraction at the outer orbit. This design also allows (some) vertical focusing owing to the alternating strong and weak magnetic field along the beam path.

The world's intensity record-breaking cyclotron is the PSI machine [12], delivering 590 MeV protons of a few mA, and well placed in the power performance plot of today's or projected machines (Fig. 2).

Further performance increase seems very difficult as increasing relativistic effects make the turn separation rapidly vanish. No construction attempt has ever been made, but there exists a paper [13] claiming that by pushing PSI technology to the extreme, one ultimately may reach 1 GeV and 10 mA.

Linear Accelerators

From their very principle of operation, linear accelerators (linacs) have a very high intrinsic performance capability in terms of beam energy and current.

A linac consists of a chain of very similar (or identical, above 2 GeV or so) accelerating cavities. Each cavity provides an acceleration of a few MeV. The cavities are driven by RF amplifiers at resonance. The final energy, not limited by a basic physics principle, is thus determined solely by the number of installed cavities. Of course, there are price and size considerations that act as practical limits. Yet these may be pushed to remarkable values, such as 500 GeV for the international linear collider (ILC) project [14].

Each linac cavity may be driven by its own RF system, with independent control of amplitude and phase for optimal settings. Thus, each cavity provides strong longitudinal focusing for each beam packet. In between the cavities, one can readily implement strong alternating-gradient (transverse) focusing, by means of quadrupoles. These two factors set a much higher charge per bunch limit than in cyclotrons and instantaneous beam currents in excess of 0.1 A have

long since been demonstrated (e.g., Los Alamos Neutron Science Center, CERN p-linac), see also Fig. 2, and one may note in passing that these values are actually defined by the (space charge) limits of the injectors at low energy rather than the main linac. A further advantage is the, in the literal sense straight-forward, extraction without beam losses at the end of the accelerator.

The "historical" linacs used normal-conducting (NC) copper accelerating cavities cooled by water with limited duty cycles. More recently, 100 % CW linacs have been designed and constructed (Fig. 2) that rely on liquid helium (LHe) cooled niobium superconducting (SC) structures. This eliminates the RF power losses, increasing the cavity-to-beam power efficiency to close to 100 % at the expense, however, of the required cryoplant. Another (and significant) advantage of the SC structures is their large aperture, which allows for very high transmission. Indeed, low beam losses are a key issue for accelerators of extreme beam power, in order avoid activation and to permit easy maintenance access.

On the other hand, the fact that the beam passes only once through each accelerating structure constitutes, at least at first glance, a real penalty as far as the required space and total cost are concerned (note that we speak of a machine that is of the order of 250 m long). Yet it is (only) this fact that permits the full implementation of the required reliability.

Reliability Implementation

As a general approach, maximizing reliability means applying a strategy of overdesign, redundancy, and fault-tolerance. This clearly requires a highly modular system where the individual components are operated substantially below their performance limits. While these conservative margins, a priori, provide operational reliability, they also provide fault tolerance: in the event of a faulty system (cavities, quadrupoles) the neighboring systems can be run momentarily at higher performance. This fault-tolerance through quick resetting (a few hundred milliseconds) of the operational parameters in the other ("redundant") systems is (for accelerators) unique to linacs. Extensive simulations [11] and also operational experience at the Spallation Neutron Source (SNS) linac [15] prove the concept.

In a cyclotron, the otherwise very efficient use of very few cavities, through which the beam passes several hundred times, intrinsically does not permit implementation of this fault-tolerance principle, in a situation where neither overdesign (for current and energy) nor redundancy (short of running a spare full cyclotron in parallel at full power) can be applied.

Present and Future High-Power Accelerator Projects

Figure 2 shows a compilation of a number of high-power accelerators in an intensity versus energy plot [16]. It is clear that all projects above the MW level have chosen superconducting linac technology. Accelerators, like the one planned for MYRRHA [11], are in mainstream development.

Incidentally, noting the remarkable position of the PSI cyclotron, showing compatibility with demonstration purposes, it is doubtful, at least to the perception of this author, if other "exotic" accelerator concepts could be pushed to the required performance. Indeed, the occasionally mentioned fixed-field alternating gradient (FFAG) accelerators and laser-plasma accelerators seem to suffer from limitations-by-concept for the former and lack of maturity for the latter. Although FFAG would implement (vertical) strong focusing to some extent and possibly could considerably push the duty cycle compared with synchrotrons, this hybrid machine could only compete with difficulty with the true CW character of a cyclotron. A high-power proton machine has never been built for this challenging concept, which also does not lend itself to the ready implementation of fault-tolerance. It will be interesting to observe for laser-plasma acceleration if the present limitations in duty cycle, power, and power conversion efficiency, which create a shortfall of many orders of magnitude for ADS applications, can be addressed by R&D in the long run.

Concluding Remarks

The outstanding PSI facility proves, on a daily basis, the possibility of a sophisticated cyclotron to deliver beams of about 600 MeV at the level of a few mA, and of driving its spallation source for many users. On the other hand, all other neutron spallation sources either currently operating, or under construction or projected, as well as high-power accelerators for other purposes in the considered energy range, have chosen SC-technology linacs.

The requirement for very high reliability for these new machines, pushed to the extreme for ADS applications, relies on the principles of overdesign, redundancy, and fault tolerance that, for the acceleration process, can only be applied through a modular design of independently phased cavities through which the beam makes a single pass.

One may, therefore, indeed conclude that the "historic" OECD-NEA assessment has kept its validity for the time being.

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Cyclotron Drivers for Accelerator-Driven Systems

M. Conjat, J. Mandrillon, and P. Mandrillon

Introduction

The requirements for accelerator-driven systems (ADS) present different technical solutions for the driver accelerator. This fact is reminiscent of the "Meson factory race" in the 1970s, where several rather different accelerator designs were proposed in the same energy domain (600–800 MeV) but with beam intensities in the 0.1–1 mA range to produce mesons.

As seen in Fig. 1, the dramatic scene observed by Ernst Michaelis at the CERN 600 MeV Synchrocyclotron, shows the powerful Los Alamos linear accelerator (linac), LAMPF, crossing the finishing line in 1972, followed by two large cyclotrons competitors in 1974; the Swiss SIN [1] (today the Paul Scherrer Institute, PSI) and the Canadian TRIUMF [2], both designs for 100 μA beam intensities, which was very challenging in the 70s.

These two cyclotron facilities are based on rather different concepts.

The PSI H^+ Two-Stage Cyclotron

The PSI has proven the soundness of the large separated magnet spiral sectors [3] concept with four powerful large single-gap radio frequency (RF) λ/2 cavities delivering up to 600 kV peak voltage at the running-in. This design established a two-stage cyclotron with a 72 MeV injector cyclotron. The ring cyclotron operates in the "single-turn operation mode" with a high extraction efficiency.

Since 1974, an outstanding intensity-improvement program has been carried out. In 1984, the compact isochronous Philips injector was replaced by a four-separated-sectors injector with an external 870 keV injection line. To reduce the number of turns in the booster ring cyclotron, new copper resonators were installed in 2008, allowing a higher energy gain per turn. The extracted beam intensity has been raised by a factor 20 and today the PSI cyclotron chain [3] is delivering a 2.2 mA, 590 MeV beam with 5×10^{-4} beam losses on the septum of the deflector of the booster (Fig. 2).

The TRIUMF H^- Single-Stage Cyclotron

The TRIUMF design proved the soundness of "single-stage acceleration up to 525 MeV" by exploiting the negative H^- ion acceleration with two simultaneous extracted beams at 100 % extraction efficiency.

The relativistic electromagnetic stripping effect of the H^- ions (the second electron is weakly bound at 0.754 eV) requires a low magnetic field. Therefore, the size of the machine is large (extraction radius = 6.9 m, total iron weight of the magnet = 2500 tons). As shown in Fig. 3, acceleration is achieved by a single, huge "dee" made of large resonators, resulting in a single "dee gap" with 90 kV peak voltage provided by a 1.8 MW RF amplifier chain. The injection is external and axial via a 300 keV injection line.

The stripping extraction works in the "overlapping-turns extraction mode". This design currently reaches 0.5 mA–525 MeV.

Based on the experience gained from these two outstanding pioneers and on the continuous upgrades of the PSI cyclotron facility, circular machines such as ring cyclotrons

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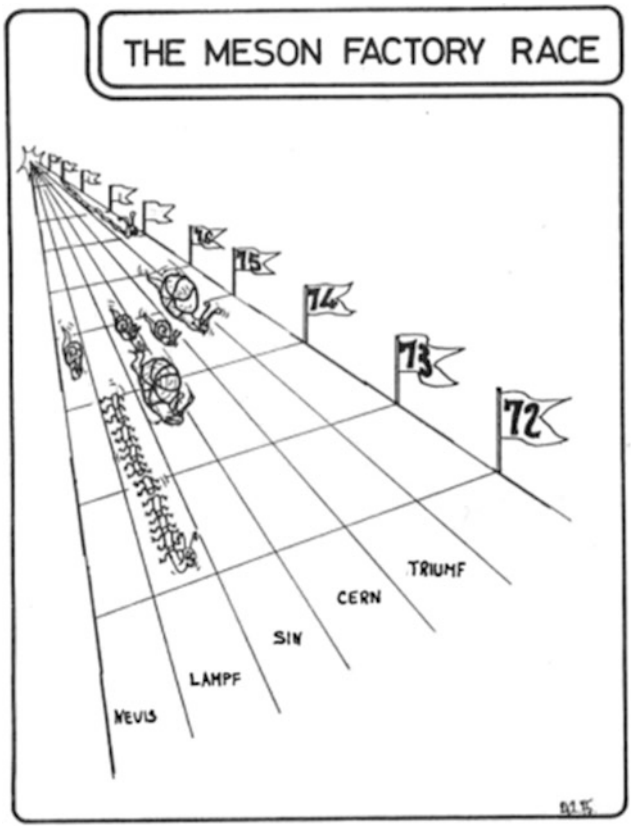

Fig. 1 The competitors of the Meson factory race

producing continuous wave (CW) megawatt beam power were proposed at the end of the 90s for the Energy Amplifier project proposed by Rubbia et al. [4]. This paper reviews various designs, from the early designs of the Energy Amplifier and TRADE, and presents new projects and perspectives for high-power cyclotron drivers.

Beam Dynamic Issues

The resonant concept along the multiturn long path in cyclotrons makes them a "natural" CW accelerator.

The circular geometry of cyclotrons brings the difficulty of the beam extraction process, the efficiency of which is of the utmost importance to avoid the activation of the whole accelerator.

Two modes of operation, illustrated by the examples of the above two pioneers, are possible.

Single-Turn Extraction

In this process, the radial separation, d, between the accelerated turns resulting from the energy gain per turn, E_g, is given by the following expression:

$$d = R\frac{\gamma}{\gamma+1}\frac{E_g}{T}\frac{1}{\nu_r^2}$$

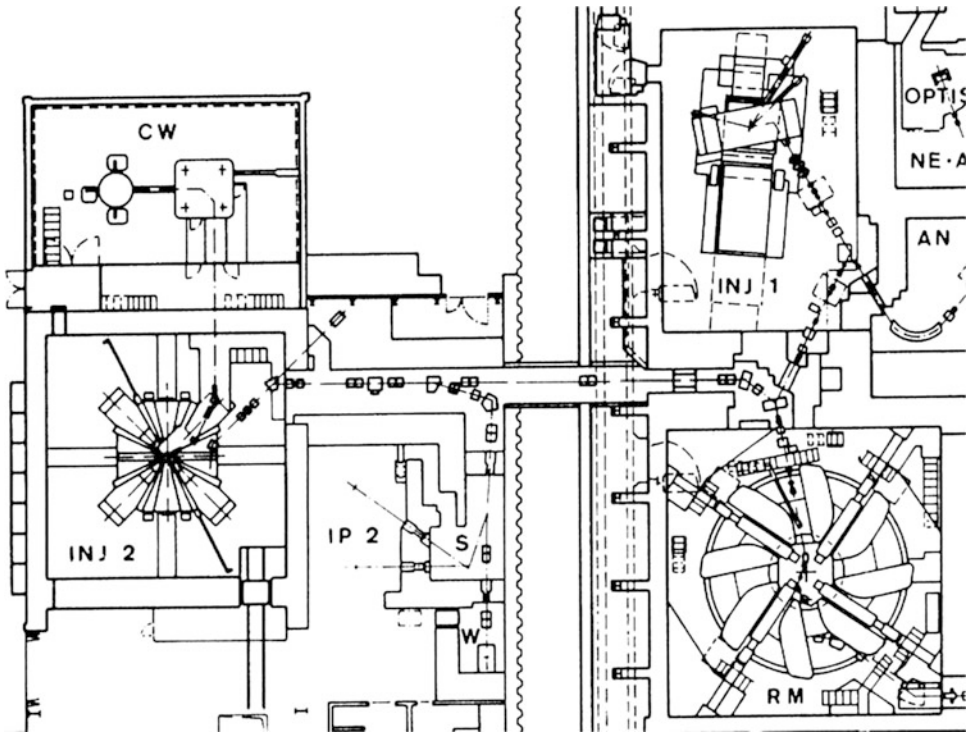

Fig. 2 Layout of the two-stage PSI cyclotron chain. INJ1 is the old compact isochronous injector cyclotron, which has been superseded by the four-separated-sectors INJ2 cyclotron. CW is the Cockroft-Walton 870 kV injection platform

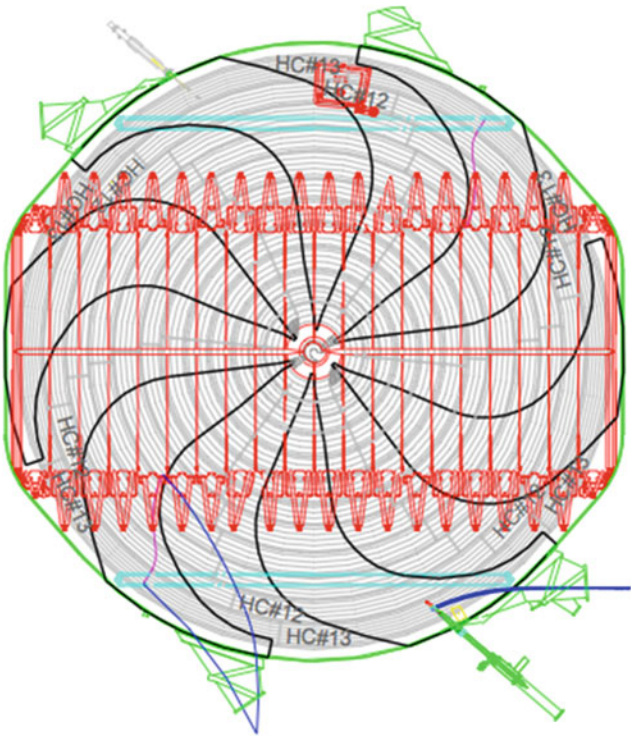

Fig. 3 The TRIUMF six-sectors single-stage cyclotron with the RF resonators in *red*

where R is the extraction radius, ν_r is the radial betatron tune number, and the maximal kinetic energy, T, is given by $T = E_0(\gamma - 1)$.

In cyclotrons longitudinal space charge forces dominate. The intensity limit calculations require complex numerical simulations, in which self-fields due to particles and external focusing fields should be accounted for. An insight into the intensity limit could be derived from simple models and is given by Joho's [5] practical formula:

$$I_{\max} = 1.4_{\mathrm{mA}} T_{\mathrm{MeV}} \beta_{\max} \Delta\varphi / 2\pi (100\,\mathrm{turns}/N)^3$$

where T is the final kinetic energy, $\beta_{\max} = v/c$ of the particle, $\Delta\varphi$ is the phase width of the beam that makes N turns in the cyclotron.

For the present performances [6] of the PSI machine as shown Fig. 4, with 188 turns in the ring and $\Delta\varphi = 8°$, we find a value of 2.18 mA, which is very close to the measured 2.2 mA.

The remarkable properties of the approximate $I_{\max}$ formula are that:

- $I_{\max}$ does not depend on the final radius or beam shape;
- $I_{\max}$ is inversely proportional to N^3, hence, requiring powerful accelerating cavities to provide high peak voltages.

The drawback of this concept is the need for an intermediate energy injector to reduce the number of turns in the booster ring. The matching of the different injection/extraction channels is highly demanding and could be a source of poor reliability for an industrial design.

Stripping or Overlapping-Turns Extraction

In this process at high energy, the extraction efficiency is very close to 100 % and it is no longer necessary to separate the turns.

The maximum intensity is, in this case, limited by the vertical space charge forces:

$$I_{\max} = \Delta z \nu_z^2 \omega_{\mathrm{RF}} \varepsilon_0 E_g \Delta\varphi / 2\pi$$

where Δz is the vertical beam size, ω_{RF} is the pulsation of the RF accelerating field, ν_z is the vertical focusing frequency, E_g is the energy gain per turn, and $\Delta\varphi$ the phase acceptance, which could be much larger than in the separated-turn extraction process; hence, reducing the longitudinal space charge forces. In TRIUMF, $\Delta\varphi$ is about 60° RF.

A TRIUMF-type single-stage six-sector cyclotron H^- design has been considered by Craddock [7], who found an accelerated beam intensity of 20 mA for 500 MeV, reduced to 16.7 mA at 600 MeV.

Recent improvements to the beam stability [8] have been carried out at TRIUMF, which could pave the way towards acceleration of higher beam intensities.

Two Examples of High-Power Cyclotron Designs

The Energy Amplifier Project

For driving the Energy Amplifier project proposed by C. Rubbia in 1995, a three-stage cyclotron driver was investigated in detail [9]. The goal was to deliver a 12 MW proton beam (12 mA, 1000 MeV). The extraction mode that was chosen for this design was the separated-turns mode. Hence, low number of turns in each stage was mandatory as well as high accelerating peak voltage in the two last stages.

The following cyclotron cascade was proposed:

- Two compact injector cyclotrons able to deliver a 6.25 mA, 15 MeV beam in a 30° RF phase width. One injector extracting H^- beam by a classical electromagnetic septum channel and the second injector extracting H^- beam by stripping to provide H^+;
- Funneling these two 15 MeV beams up to the injection line in an intermediate-stage separate sector cyclotron (ISSC);
- A four-sector ISSC accelerating the beam up to 120 MeV;

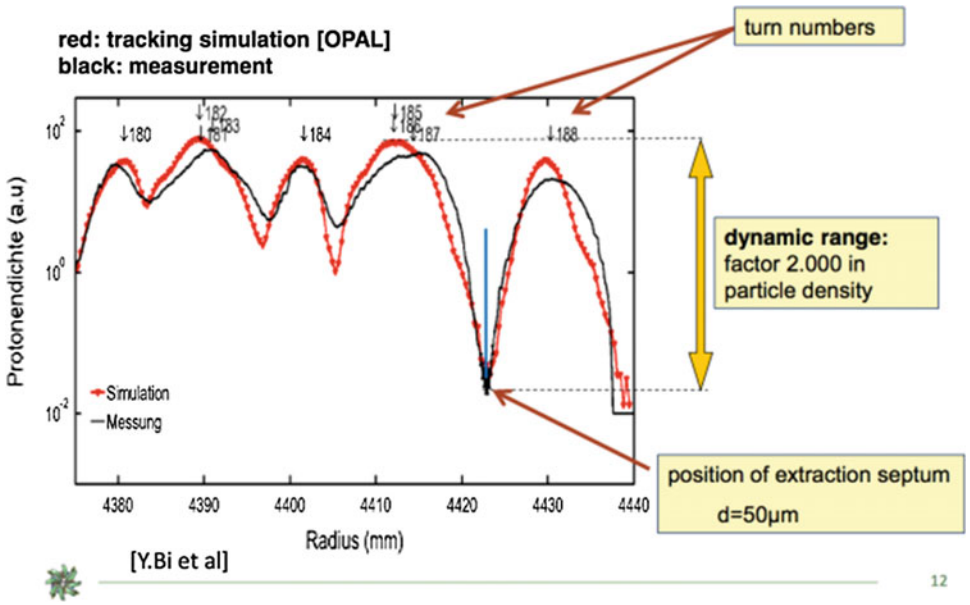

Fig. 4 Comparison between simulated and measured relative beam intensity in the turn separation extraction process [6] in the PSI ring cyclotron, showing the losses on the extraction septum

- A final-stage booster separated-sector ring cyclotron, which is made of 12 magnet sectors and six RF cavities, raising the energy up to 1000 MeV (Fig. 5).

Extensive beam dynamics studies, including different models for space charge calculations [9], were carried out in the cyclotron stages. Half-scale models of the large curved mono-gap cavities were also built to optimize and measure the needed high quality factor to reach a 1.1 MV peak voltage at the extraction (2.24 MW must be transmitted to each cavity; 400 KW for losses and 1.84 MW beam power).

The TRADE Project

The TRADE [10] experiment proposed by C. Rubbia consisted of driving a modified TRIGA reactor of the ENEA–La Casaccia Center with an external proton accelerator to a

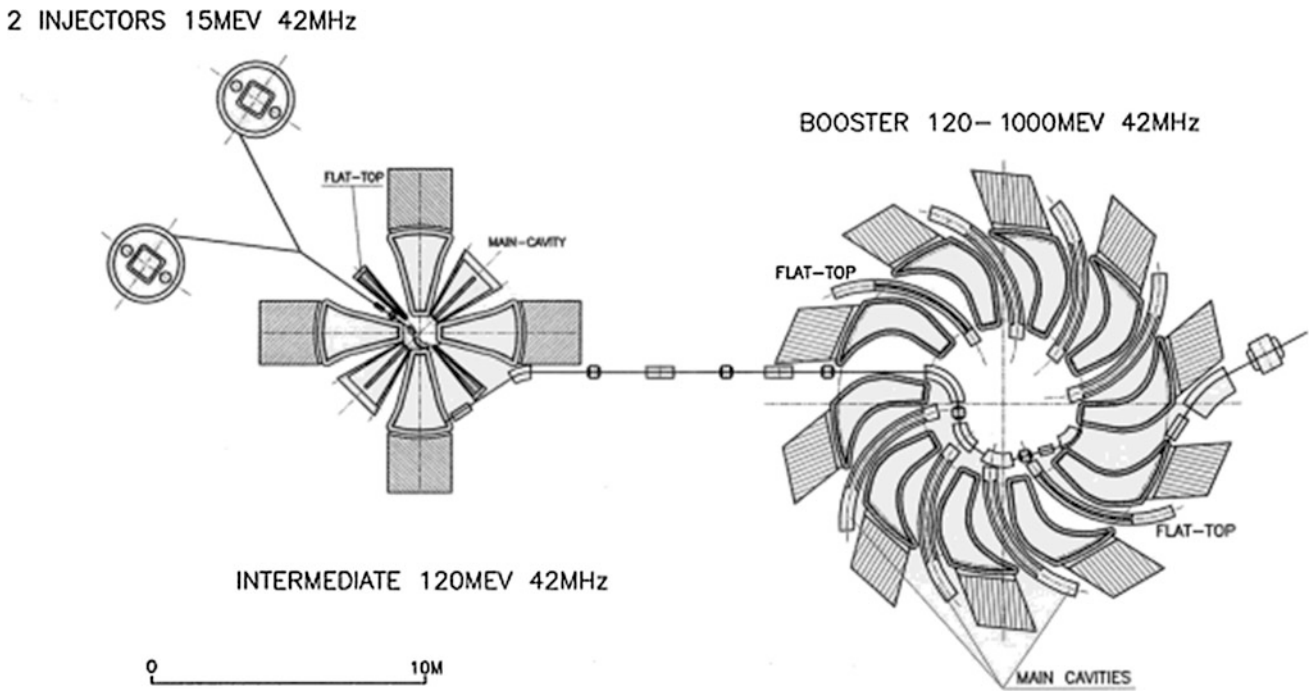

Fig. 5 The three-stage cyclotron for driving the Energy Amplifier

target to be installed in the central channel of the reactor scrammed to subcriticality.

For this ADS experiment, the AIMA Company proposes a 230 MeV H_2^+ superconducting compact isochronous cyclotron [11] that is able to deliver, by stripping extraction, a 115 MeV proton beam in the 2–3 mA intensity range. This overlapping-turns design leads to a single-stage, stand-alone cyclotron with very high extraction efficiency. The H_2^+ beam

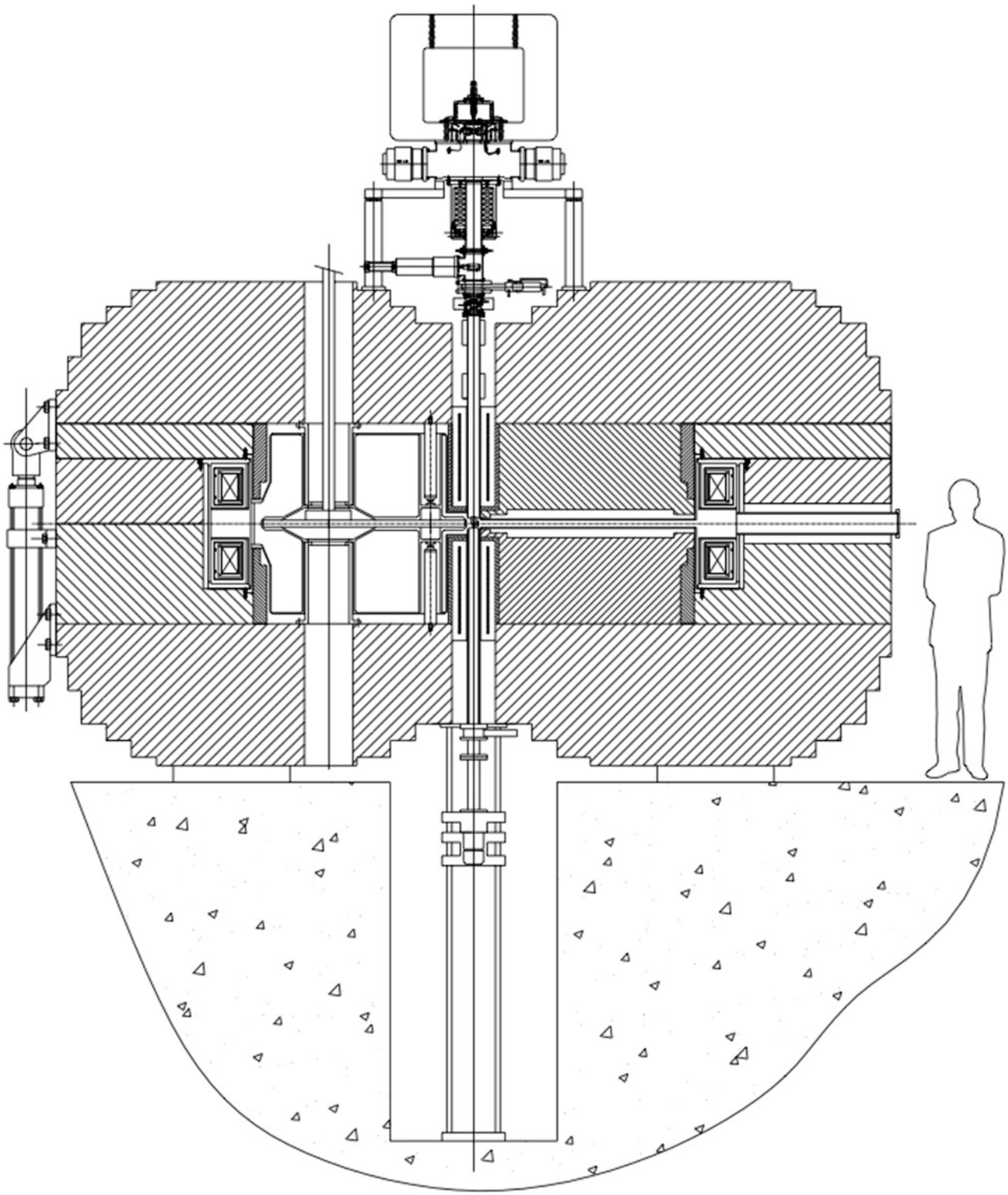

Fig. 6 Vertical cross section of the stand-alone H_2^+ superconducting cyclotron for driving the TRADE experiment

is axially injected in the vertical axis of the compact cyclotron. For a total of 790 KW RF power, four RF cavities with 160 kV peak voltage accelerate the beam at the extraction radius, where the H_2^+ beam is stripped into two protons. An optimal location of the stripper foil was found to be such that, on their inwards motion, the stripped proton trajectories remain sufficiently far from the cyclotron center (Fig. 6).

New Advanced High-Power Cyclotron Designs

The 800 MeV Superconducting Strong Focusing Cyclotron

This ambitious 800 MeV, 15 MW project is proposed by the Texas A&M University [12]. This project is characterized by three innovative key features:

- It is composed of a stack of three coupled cyclotrons, providing three beams of 5 MW in parallel. Each cyclotron drives two 2.5 MW spallation targets in an ADS core;
- It uses superconducting RF resonators with a high quality factor, about 10^{10} at 4.2 K;
- It uses quadrupole focusing channels to obtain high betatron tune numbers to provide strong transversal focusing.

The concept is a two-stage superconducting cyclotron; hence, requiring a separated-turn extraction process with a 100 MeV injector that is a stack of three coupled four-sector cyclotrons.

The booster ring is made of:

- Twelve flux-coupled stacks of dipole magnet sectors with low magnetic field (0.6 T). Therefore, the overall diameter of the machine is large, about 20 m;
- Ten superconducting 100 MHz RF cavities, providing a 20 MeV energy gain per turn resulting in 35 turns to reach the extraction energy. A 5 cm dynamic aperture is obtained on the extraction radius;
- The large energy gain per turn allows the possibility to insert strong focusing transport channels made of Panofsky quadrupoles, providing a strong gradient of the order of 4 T/m (Fig. 7).

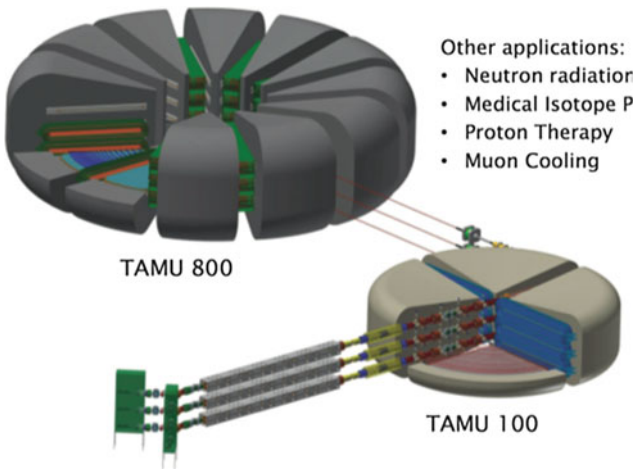

Fig. 7 The two-stage, strong focusing 800 MeV three-stack cyclotron

The DAEδALUS Project

To drive the DAEδALUS CP-violation neutrino experiment [13], which needs three neutrino sources, a complex of three accelerators based on high-power 800 MeV/amu H_2^+ cyclotrons able to deliver a peak intensity of 5 mA (i.e., providing a 1 mA average current and a 1.6 MW average beam power with 20 % duty cycle) is required by this experiment. These performances make them a potential competitor for cyclotron-based ADS as presented by Barlow [14].

Each accelerator consists of a two-stage cyclotron:

- A compact injector cyclotron, which accelerates H_2^+ beams up to 60 MeV/amu. The extraction is made by turn-separation via the two usual electrostatic deflectors. The same type of compact cyclotron could be used for the IsoDAR experiment [15].
- The superconducting ring cyclotron boosts the beam energy up to 800 MeV/amu. The extraction of the beam of protons is made by stripping of the H_2^+, which is dissociated in a thin foil. In the early option A, this stage was made of eight sectors excited by RIKEN-type [16] superconducting coils. Acceleration is given by four large mono-gap PSI-type resonators. Recent studies presented a more practical option B, with a six-sector design of the magnet, which allows it to host more easily the PSI-type resonators. The high magnetic rigidity of H_2^+ requires a maximum 4.72 T magnetic field in the sectors. As shown in the following figures, the extracted trajectories are a long path owing to the internal inwards motion of the stripped H^+ beams (Fig. 8).

The AIMA DEVELOPPEMENT (AD) Single-Stage High-Power Cyclotron

The goal of the high-power cyclotron proposed by the AD Company is to provide typically a 10 MW (12.5 mA, 800 MeV) proton beam for driving an industrial ADS. The proposed driver is a single-stage design, like the TRIUMF pioneer. The following main features [17] of two possible options were presented in November 2012 in Erice [18]:

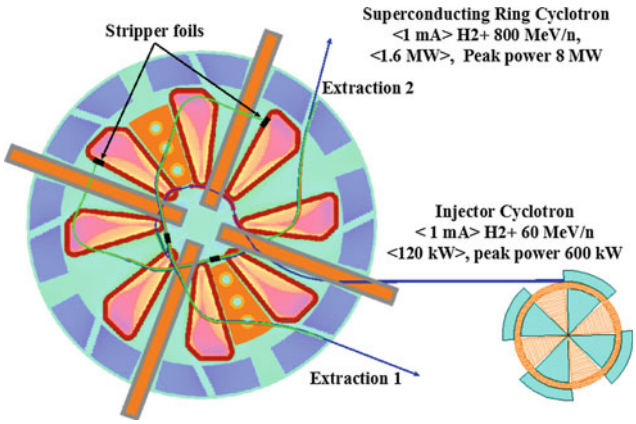

Fig. 8 Option A for the two-stage DAEδALUS superconducting H_2^+ 800 MeV/amu cyclotron

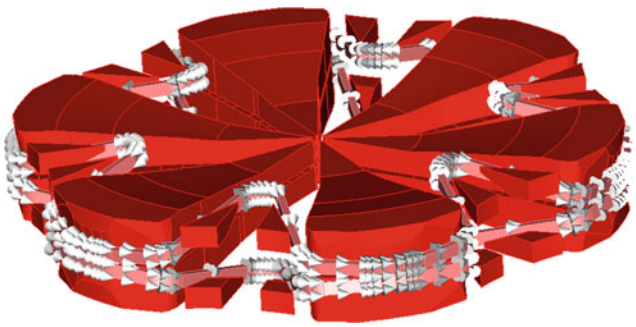

Fig. 9 3D view of the six-sector single-stage AIMA DEVELOPPEMENT cyclotron with reversed valley B-field

- Option A: acceleration of H_2^+ ions up to 1600 MeV kinetic energy, delivering 800 MeV protons by stripping;
- Option B: acceleration of protons up to 800 MeV and extraction by conventional channels.
- Each option is made of the following main components:
- A six-sector magnet excited by a single large superconducting coil, the particular shape of which produces a reversed magnetic field in the valleys (Fig. 9). The large magnetic field flutter provided by this magnet layout [18] avoids spiraling the sector edges to get the required vertical focusing. This particular geometry makes the extraction of protons by stripping of H_2^+ (option A) in the reversed field valleys much shorter and hence simpler than in the DAEδALUS booster, for example.
- Acceleration is provided by six delta cavities in the valleys. Double-gap RF cavities have been selected because their radial extension is smaller than that of single-gap cavities of the PSI-type. This type of cavity is more appropriate for a single-stage accelerator.
- Multibeam acceleration, up to three beams, is provided by three low-energy axial injection lines fed by three ion source platforms (Fig. 10).

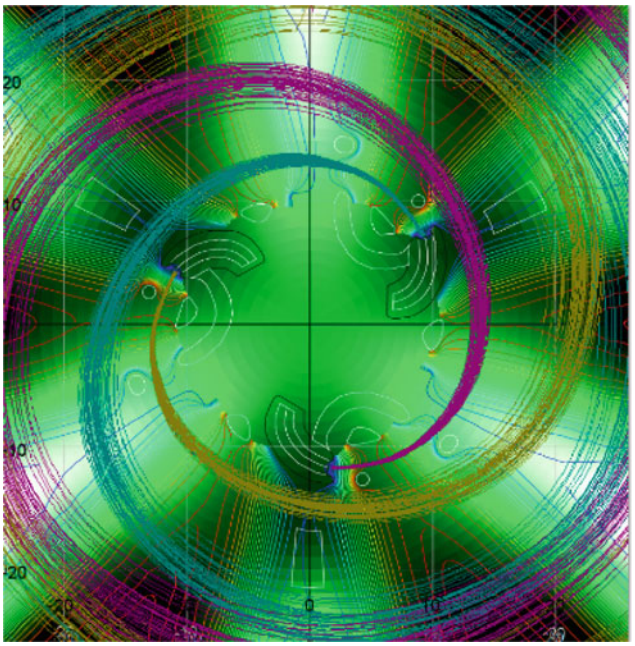

Fig. 10 Central region with the three simultaneously injected beams

Table 1 gives the characteristics of the most crucial component; the RF accelerating cavities.

Table 2 provides rough preliminary figures for the electric power needed to run a single-stage H_2^+ cyclotron feeding two beam lines.

Therefore, for a 10 MW beam power, the overall efficiency of the single-stage H_2^+ cyclotron is typically a 40 %.

Critical Issues for an Industrial ADS Cyclotron Driver

To reach the high level of reliability of an industrial cyclotron-based ADS driver that is able to typically deliver a 10 MW proton beam (e.g., 800 MeV, 12.5 mA), the following requirements of the subsystems are mandatory.

Ion Sources

A design with accelerated beams in parallel, for example, the AIMA DEVELOPPEMENT design, will provide possibilities to run the cyclotron with three sources and their associated short injection lines. This allows the required performances of each ion source to be relaxed somewhat and, therefore, reduces intensity fluctuations problems and better controls beam trips.

Table 1 Main characteristics of the H_2^+ single-stage cyclotron RF cavities

Type of cavity	$\lambda/2$, double-gap, tapered walls
Frequency	36.3 MHz
Cavity length	5.4 m
Peak voltage at injection	160 kV
Peak voltage at extraction	450 kV
Quality factor	6200
Losses/cavity	350 kW
Beam power/cavity	1650 kW
Total power/cavity	2000 kW
Total electric power/cavity (η = 70 %)	2800 kW

Table 2 Total electric power of a H_2^+ single-stage cyclotron

RF power (6 cavities)	17 MW
Superconducting magnet	1 MW
Injection (3 sources + 3 axial lines with bunching, foc. lenses, inflectors)	1 MW
Two external beam transport lines	4 MW
Spare power	2 MW
Total electric power	25 MW

RF Problems

Each large RF cavity will have to handle 1–2 MW of beam power, including the RF losses in the walls for non-superconducting cavities. Transmitting this power makes the coupling a non-trivial task. According to the PSI experts, one single RF window can handle up to 700–800 kW. Clearly, this means that three such windows and coupling loops are needed for each cavity. The RF generators will consist of a three-stage power amplifier. Tetrodes are to be used in the last two stages in the cathode-driven configuration in order to obtain the required stability of the whole system. Because of the presence of magnetic stray fields, it is not possible to locate the power amplifiers close to the cavities. Therefore, the power has to be transmitted through long RF feeders to the coupling loops. For safety purposes, directional couplers should be included.

For the overlapping-turns extraction option, in a worst-case scenario, the loss of a RF cavity could be compensated for by the other cavities as long as 70 % beam loading is acceptable in the other cavities. This would mean that the accelerated beam intensity should be decreased accordingly, but this of course might require some "off" time, which should be minimized. Nevertheless, such scenarios need to be further investigated.

Extraction Elements

This is certainly the most important concern for minimizing beam losses. According to the PSI experience in a multi-stage cyclotron, the different extraction channels with their septa are the most frequent causes of trouble and reduce the mean time between failures. Therefore, reducing the number of these channels by choosing a single-stage ring cyclotron is of the utmost importance.

Vacuum Problems

For cyclotrons accelerating H_2^+, a high vacuum quality is required. For large machines, the out-gassing rate of the RF cavities and of the vacuum chamber should be carefully controlled. CERN technical experience in this field on the needed passivation processes is certainly relevant.

High Global Yield of the Accelerator

For example, the power that is needed to run the cyclotron driver should be optimized. For the AD single-stage cyclotron, this yield will be close to 40 %.

A Cluster of Cyclotron-Based ADS for an Industrial Power Plant

A single-stage cyclotron-based ADS solution offers the attractive possibility of the parallel concept [19]. Figure 11 presents a typical cyclotron-based plant. It consists of three

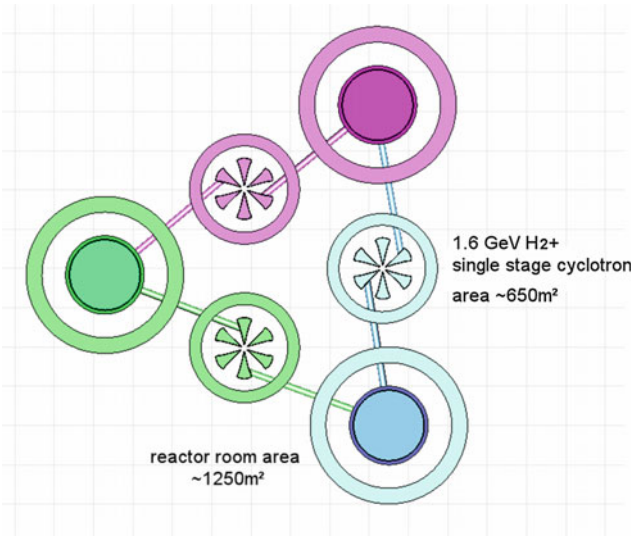

Fig. 11 A power plant made of three energy amplifiers fed by a cluster of three H_2^+ cyclotron drivers

modules with three H_2^+ cyclotrons in parallel. Each module consists of a 1400 MW_{th} unit, which could be fed by two single-stage cyclotrons in parallel. The relatively low cost of a single-stage cyclotron and its small footprint (about 500 m^2) leads to a compact layout. In the central part of the power plant, all the ancillaries equipment could be installed.

This parallel concept, as opposed to a series concept based on several linac stages, enables the overall reliability of the plant to be increased. Moreover, this layout will allow a cyclic maintenance of the plant by making two single-stage drivers permanently available.

For example, if we consider a $k = 0.98$ neutron multiplication coefficient and a 800 MeV maximal proton energy (intrinsic spallation gain $G_0 = 2.8$), the overall energy gain of an energy amplifier (EA) type ADS will be 140. Hence, for a 10 MW driving power, the thermal power for a single module is 1400 MW_{th}. Taking into account the electric power needed to feed the three cyclotrons (about 75 MW; Table 2), the electric power that could be delivered to the grid by such a cluster would be close to 1600 MW_e.

Conclusion

The state of the art of high-power cyclotrons is based on the outstanding performances achieved by the PSI facility, which is now running at close to 2 MW beam power, and it is worth observing that space charge studies [20] have shown a good agreement between measured beam profiles and simulations.

The analysis of the various studies of different designs carried out up to now have shown that no insurmountable obstacles are expected when extending the 500–600 MeV energy range of isochronous ring cyclotrons to the 800–900 MeV domain of the ADS drivers.

To enter the required 10 MW beam power while keeping the beam losses within the accelerator in the 0.01–0.05 % range requires investigation of new concepts. Avoiding a cyclotron chain, that is, an injector cyclotron with all the matching components between the stages, would be an asset. Additionally, the compactness of the geometry of a single-stage design should certainly result in a cost-effective solution for an industrial driver of an ADS. Preliminary rough cost estimates of the various components are close to 100 M€.

High reliability levels through redundancy are mandatory for a 10 MW driving beam power. The studies of various cyclotron designs in progress (Texas, AIMA, Daeδalus) reveal a common feature—the parallel concept aiming at several goals:

1. To multiply the final accelerated intensity while lowering the space charge problems:
   - The single-stage concept proposed by AD that employs injection of three beams in a common median plane of acceleration: factor 3;
   - The superconducting strong focusing cyclotron proposed by Texas A&M using acceleration in three separate median planes: factor 3;
   - Acceleration of H_2^+ ions (Daeδalus and AD, option B): factor 2.
2. Solving the problem of beam trips by a fast response:
   - The cluster (Fig. 11) of H_2^+ cyclotrons with two extraction lines for each cyclotron is an attractive economical solution, which allows smooth control of the fluctuations of beam intensities.
3. Easier cyclic maintenance program of the cyclotron complex:
   - This last advantage of the parallel concept is clearly of the utmost importance for an industrial ADS.

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Euratom MAX Project: The MYRRHA Accelerator eXperiment R&D Program

Jean-Luc Biarrotte, Frédéric Bouly, Luis Medeiros Romao, Dirk Vandeplassche, and Didier Uriot

Abstract

The goal of the MYRRHA project is to demonstrate the technical feasibility of transmutation in a 100 MW_{th} accelerator-driven system (ADS) by building a new flexible irradiation complex in Mol (Belgium). The MYRRHA facility requires a 600 MeV accelerator, delivering a maximum proton flux of 4 mA in continuous operation, with an additional requirement for exceptional reliability. This paper will briefly describe the present status of this ADS accelerator design, and will then focus on fault tolerance aspects and their impact on the beam operation, and the ongoing R&D activities.

Introduction

MYRRHA ("Multipurpose Hybrid Research reactor for High-tech Applications") is a new flexible fast spectrum research reactor that is planned to be operational around 2023 in SCK•CEN, Mol, Belgium [1]. Composed of a proton accelerator, a spallation target, and a 100 MW_{th} core cooled by liquid lead–bismuth, it is especially designed to demonstrate the feasibility of the ADS concept in view of high-level waste transmutation. To feed its subcritical core with an external neutron source, the MYRRHA facility requires a powerful proton accelerator (600 MeV, 4 mA) operating in continuous wave mode (CW) and, above all, featuring a very limited number of unforeseen beam interruptions. In addition, a specific beam time structure is required to enable the monitoring of the reactor subcriticality (more details are given in [2]).

The conceptual design of such an ADS-type proton accelerator was initiated during previous EURATOM Framework Programs (PDS-XADS and EUROTRANS projects). It is a linac (linear accelerator) based solution that brings excellent electric efficiency thanks to the use of superconductivity and high potential for reliability by the use of several redundancy schemes.

R&D on ADS-type accelerators is presently being pursued in the frame of the MAX project [3], supported by EURATOM FP7. This project aims at delivering an updated and consolidated reference layout of the MYRRHA linac with sufficient details and an adequate level of confidence to initiate in 2015 its engineering design and subsequent construction phase. To reach this goal, advanced beam simulation activities are being undertaken and a detailed design of the major accelerating components is being carried out, building on several prototyping activities. Strong focus is also being put on all the aspects that pertain to the reliability and availability of this accelerator, with the development of a detailed reliability model of the MYRRHA accelerator and with dedicated R&D to experimentally prove, in particular, the feasibility of the innovative "fault-tolerance" redundancy scheme.

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Overview of the MYRRHA Accelerator

The energy of the proton beam driving a Pb or Pb–Bi ADS reactor must typically be around 1 GeV. This value is the result of a compromise dictated by different criteria. The proton energy must be high enough to maximize the efficiency of the spallation reaction (and reach a typical yield of 25–30 neutrons per incident proton and GeV), but also to limit the energy deposited in the target and its window, thereby, facilitating its design and usage constraints. On the other hand, cost optimization of the accelerator promotes low energy. Therefore, a reference energy of 600 MeV has been chosen for the MYRRHA 100 MW_{th} demonstrator. For the record, this compromise has been set to 800 MeV for the industrial prototype 400 MW_{th} version called EFIT [4].

Once the proton energy is fixed, the beam current necessary to ensure control of the reactor is then mainly determined by the thermal power and the level of subcriticality of the core. In the case of MYRRHA (k_{eff} = 0.95), the required nominal average current is 2.5 mA—with up to about 4 mA being foreseen for burnup compensation through the cycle—whereas for EFIT (k_{eff} = 0.97) it would be about 20 mA, corresponding to a mean beam power of 16 MW. With the current state of technology, the only possibility to produce such a high beam power continuously (and reliably) is the use of a linear accelerator. It is partly for this reason that MYRRHA, as a demonstrator, has also opted for this option at the expense of the cyclotron option, following at the same time the recommendations of the 2002 OECD/Nuclear Energy Agency dedicated workshop [5].

In addition to the very high level of mean beam power to be produced by the MYRRHA accelerator (2.4 MW), the main challenge to address for this machine is the level of reliability required, as the number of beam interruptions should be limited to extremely low values: the current maximum limit is set to ten beam interruptions per three-month operating cycle, with only interruptions of more than 3 s being counted, leading to a global accelerator MTBF (mean time between failures) of about 250 h. This draconian reliability specification, settled during the EUROTRANS project, is motivated by the fact that beam interruptions longer than a few seconds could lead, if repeated frequently, to unacceptably high thermal stresses on the highly irradiated materials of the target window, the fuel claddings, and more generally on all the reactor structures. In addition, such long beam interruptions will probably be systematically associated with reactor shutdowns, which could also significantly affect the availability of the system as the considered restart procedures could typically last about 20 h [6]. It is, however, to be underlined that other studies in Japan and the United States [7] are quoting much less severe reliability specifications for similar ADS machines, highlighting that many uncertainties and unknowns still remain on this theme. In any case, it is clear that the reliability target for the MYRRHA ADS accelerator will need to be significantly higher than the actual reliability recorded worldwide on comparable accelerators in operation today, such as the SNS, which exhibits a MTBF of a few hours presently at best. This gap to be filled is illustrated in Fig. 1 [8], which definitively shows that reliability is indeed the main challenge for the MYRRHA accelerator.

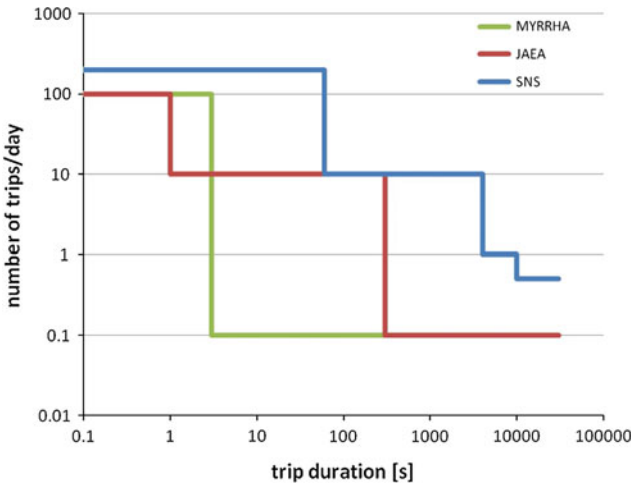

Fig. 1 Beam trip frequencies as a function of their duration: recorded in the Spallation Neutron Source (SNS), allowed by the Japanese ADS (Japan Atomic Energy Agency), accepted for MYRRHA

The conceptual design of the MYRRHA machine has been on-going for several years [9–13] and it is becoming quite clear that in order to reach the reliability goal, the linac scheme should consist—apart from the final beam transport lines—of two clearly distinct sections (see Fig. 2): a low energy part, composed of two redundant compact injectors with fast switching capabilities, and a highly modular and acceptant 17–600 MeV superconducting linac, based on independently controlled accelerating cavities yielding a strong tolerance to faults.

Tolerance to Faults

Injector Beam Reconfiguration in Fault Cases

The 17 MeV MYRRHA injector line [14] is designed to provide optimal acceleration efficiency with a minimized number of components. It is composed of a 30 kV electron cyclotron resonance (ECR) proton source, with its 2 m long low energy beam transport (LEBT), a 4 m long, 176 MHz

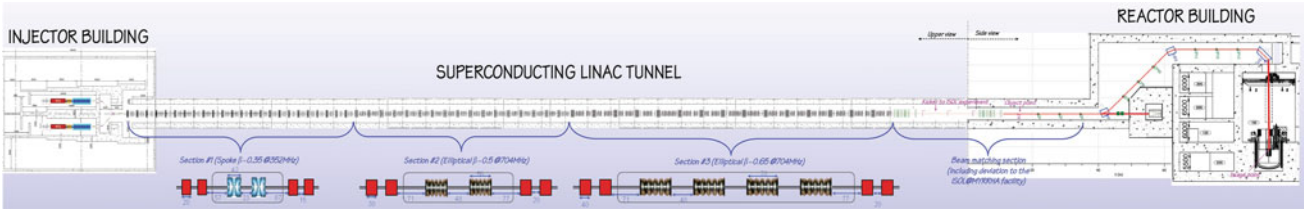

Fig. 2 Schematic overview of the MYRRHA linac (to scale; the superconducting linac tunnel is ~300 m long)

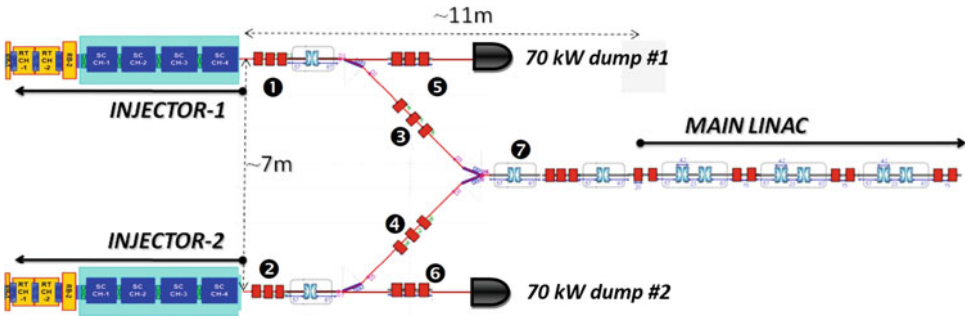

Fig. 3 General layout of the MYRRHA medium energy beam transport line

four-rod radio-frequency quadrupole (RFQ) [15] accelerating the beam to 1.5 MeV and operating with a very conservative inter-vane voltage (30 kV), followed by a booster—still under fine optimization within the MAX project—made with several room-temperature and then superconducting multicell cross-bar H-type (CH) cavities [16].

To increase the reliability, the philosophy is to double the whole 17 MeV linac, providing a hot stand-by spare injector able to quickly resume beam operation in case of any failure in the main one [9]. The fault-recovery procedure is based on the use of a switching dipole magnet with a laminated steel yoke connecting the two injectors through a 'double-branch' medium energy beam transport (MEBT) line, as shown in Fig. 3.

In a fault case, the injector beam reconfiguration should last not more than 3 s. The mains steps of the reference scenario for such a retuning are the following:

- In the initial configuration, one of the injectors (e.g., Injector #1) is providing beam to the main linac. The other parallel injector (#2) is also fully operational, but the produced beam is sent to a dedicated beam dump and continuously monitored.
- A serious fault is detected during operation and the beam is **immediately** and automatically stopped at the source exit in both injectors by the machine protection system (MPS), by means of the LEBT chopper (as a first step).
- The origin of the fault is analyzed and diagnosed by the control system.
- If the fault is indeed localized in Injector #1, the necessary injector retuning procedure is started, and a new beam path is set to be able to feed the main linac using Injector #2; in particular, the polarity of the power supply feeding the MEBT switching magnet is changed and the Injector #2 45° dipole is switched on.
- Once a steady-state condition is reached in the retuned magnets, the beam is resumed in Injector #2, at first with very short pulses to check that the transport is okay up to the target; the duty cycle is then ramped within a second or so ("fast commissioning mode") to recover nominal beam operation.
- Once beam operation is resumed, maintenance is then started on the failed Injector #1, the casemate of which is supposed to be accessible during operation of Injector #2. If the failure can be fixed, Injector #1 is then retuned, the beam is re-commissioned locally on the dedicated beam dump. Injector #1 then stays in "stand-by-mode", ready to relive Injector #2 in case of failure.

It is clearly expected that the numbers of this retuning procedure will have to be as low as possible, to reach a MTBF of 250 h. To optimize the performances of the injector, specific R&D topics within the MAX project are dedicated to the normal and superconducting [16] CH cavities as well as the RFQ [15]. It is planned to install a

Table 1 Main parameters of the MYRRHA main linac

Section #	#1	#2	#3
E_{input} (MeV)	17.0	80.8	184.2
E_{output} (MeV)	80.8	184.2	600.0
Cavity technology	Spoke	Elliptical	Elliptical
E_{Cav} frequency. (MHz)	352.2	704.4	704.4
Cavity optimal β	0.375	0.510	0.705
Number of cells/cavity	2	5	5
Focusing type	NC quadrupole doublets		
Number of cavities/cryometer	2	2	4
Total number of cavities	48	34	60
Nominal E_{acc} (MV/m)	6.4	8.2	11.0
Synch. phase (deg)	−40 to −18	−36 to −15	
4 mA beam load/cavity (kW)	1.5–8	2–17	14–32
Nominal quadrupole gradients (T/m)	5.1–7.7	4.8–7.0	5.1–6.6
Section length (m)	73.0	63.9	100.8

prototype of the first part of the injector (LEBT + RFQ) at UCL (Université Catholique de Louvain-la-Neuve) [17].

The LEBT construction is currently under progress and will be first tested at LPSC (Laboratoire de Physique Subatomique et de Cosmologie de Grenoble). This experiment will also enable the study of the "space charge compensation" phenomena, which will be used to maximize the beam transmission to the RFQ. This will also be a test bench for improvements to beam diagnostics technologies.

It is indeed clear that the MYRRHA fault compensation scheme is based on reliable and accurate beam monitoring. Non-destructive beam diagnostics will be extremely useful to settle the retuning procedures in the injector and in the main superconducting linac [13].

Superconducting Linac Beam Reconfiguration in Fault Cases

Main Design Aspects

The architecture of the 17–600 MeV MYRRHA main superconducting (SC) linac is summarized in Table 1 [13]. It is composed of an array of independently powered superconducting cavities with high energy acceptance and moderate energy gain per cavity (low number of cells and very conservative accelerating gradients), the goal being to increase the tuning flexibility as much as possible and to provide sufficient margins for the implementation of the fault-tolerance scheme. Three distinct cavity families are used to cover the full energy range: the first section uses 352.2 MHz spoke two-gap cavities (β_{opt} = 0.37), whereas the two following sections use 704.4 MHz elliptical five-cells cavities (β_{opt} = 0.51 and 0.70). Such a choice is based on the results of a longitudinal optimization using the GenLinWin simulation code [18]; this analysis actually clearly shows that three sections is a straightforward choice for such a 17–600 MeV SC linac, and that playing around with cavities' beta values and the number of cells does not change the picture much. Nevertheless, it is to be underlined that using European Spallation Source (ESS)-type β = 0.5 double-spoke cavities [19] could be an interesting back-up option for section #2.

The design of the elliptical cavities has been performed through previous dedicated R&D programs, including prototyping and RF tests [20, 21]. The design of the MYRRHA spoke cavity [22] has been recently achieved within MAX and prototyping should begin very soon. The operating accelerating gradients of the MYRRHA cavities have been chosen to be on the conservative side, taking, in particular, as a reference the actual average operating point of the SNS β = 0.61 cavities in 2008 [23]. The chosen rules for the operation of the MYRRHA superconducting cavities are the following: the RF fields at the inner surface of the SC cavities is always kept under 35 MV/m peak electric field and 60 mT peak magnetic field; the nominal "derated" operation points are then obtained by removing 30 %, to be used as a margin for fault compensations. These rules lead to nominal accelerating fields of 6.4, 8.2, and 11.0 MV/m in the three different sections, with a required maximum E_{acc} capability of 8.3, 10.7, and 14.3 MV/m, respectively.

Fault Compensation Scheme

The present reference scenario for a fault-recovery procedure in the MYRRHA main linac is based on the use of a local compensation method, in which only adjacent elements are

used to recover nominal beam operation. This procedure should not last more than 3 s and is defined as follows.

- In the initial configuration, the main linac is operational, with all elements operating with nominal (derated) parameters. The typical needed derating level has been evaluated to be about 30 % for cavities (accelerating field), 40 % for amplifiers (RF power), and 10 % for quadrupole power supplies.
- A serious fault is detected during operation (abnormal beam loss, for example) and the beam is immediately and automatically stopped at the source exit in the operating injector by the machine protection system, by means of the LEBT chopper.
- The origin of the fault is analyzed: if it is successfully diagnosed and can be compensated, the fault recovery procedure is initiated (otherwise the beam needs to be stopped permanently). The proposed basic preliminary rules to be fulfilled to make the compensation possible are the following:
  - Fault implying a cavity: the four nearest neighboring cavities operating derated (i.e., not already used for compensation) are used for compensation; the maximum allowed number of consecutive failed cavities is two (in sections #1 or #2) or four (in section #3);
  - Fault implying a quadrupole: the whole doublet is switched off and the four neighboring doublets are used for compensation; the maximum allowed number of consecutive failed doublets is one.
- The recovery procedure is then processed as follows (an example of a cavity fault in is given in [24] and more specific details on the linac beam dynamics in [13]).
  - The failed cavity RF loop is immediately disabled and the cavity quickly detuned by typically more than 100 bandwidths to avoid the beam loading effect when the beam is resumed (time budget: less than 2 s).
  - In parallel, new (voltage/phase) set-point values for compensating cavities are picked in the control system database; these values should have been determined beforehand from past beam experience if the fault configuration has already been met, or from a predictive calculation using a dedicated beam dynamics simulation code if not. The new set-points are then applied in the corresponding low-level RF systems.
- Once a steady-state condition is reached in the retuned cavities (or magnets), the beam is resumed in the linac at first with very short pulses to check that the transport is okay up to the target; the duty cycle is then ramped within a second or so ("fast commissioning mode") to recover nominal beam operation.
- Once beam operation is resumed, maintenance is then started if possible, that is, if the faulty element is located outside the tunnel. If the repair is clearly successful, the opposite procedure could be envisaged to come back to the initial configuration.

This retuning strategy has been successfully assessed in several fault test scenarios [13, 24–26], including the case of the lower energy cavities and the case of multiple simultaneous faults (one cryomodule off in each section). The main conclusion of these fault-recovery scenario analyses is that the fault recovery scheme is a priori feasible everywhere in the MYRRHA main linac to compensate for the loss of a single cavity or even a full cryomodule. This statement should be confirmed by the upcoming advanced beam dynamics studies to be performed in the MAX project, where these compensation schemes will be also assessed through full start-to-end (source to target) simulations including random errors.

To practically implement such compensation schemes, a first detailed recovery procedure has been defined. Several steps of this procedure appear to not be straightforward and will require further studies. The switching time of less than 3 s will clearly be a critical issue, with probably huge consequences on the required capabilities of the machine control system (efficient and fast fault diagnostic, fast automated beam restart, and associated consequences). Also, an efficient predictive beam simulation code will need to be developed and benchmarked during the machine commissioning phase so as to be able to efficiently predict the optimal retuning set points in every fault configuration.

On the RF cavity side, dedicated low-level RF digital systems and fast and reliable cold tuning systems also need to be developed together with suited regulation loops. A R&D program is on-going in MAX at IPN (L'Institut de physique nucléaire) Orsay [27] on these aspects, especially on the tuner design, which specifically needs to provide a large detuning range for the duration of the recovery procedure (i.e., a few seconds) to minimize the perturbation from the unused cavity [13, 28].

Conclusion

The successful and reliable production of the MYRRHA high power and stable beam is a challenge that clearly requires a huge R&D investment beforehand. The present activities performed within the MAX project and at SCK•CEN are mainly dedicated to the general design of the accelerator and to the development of a few main primary components. Huge additional effort will be required in the coming years, not only to push these on-going R&D activities towards an engineering design phase (construction of a

full injector test stand, cryomodules prototyping, etc.), but also to initiate activities on the additional topics related to beam operation. This concerns, in particular: better understanding of beam physics at low energy for transients management, fine optimization of the strategies and procedures for faults compensation, development of a predictive beam simulation code ("virtual machine"), design of a smart control system including efficient "fast fault diagnostic" and "second-class" automated beam restart capabilities, design of a suited machine protection system, development of a set of reliable beam instrumentation diagnostics, and, in particular, a robust near-target imaging system, among others.

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Accelerators for Accelerator-Driven Subcritical Reactors

Roger Barlow

Abstract

Thorium-fuelled accelerator-driven subcritical reactor (ADSR) systems require high-power proton beams, beyond the capacity of existing machines. This paper reviews the requirements for energy, current, and reliability, and how they might be met by cyclic accelerators, looking particularly at the DAEδALUS MultiMegawatt Cyclotron, and a non-scaling fixed-field alternating gradient (nsFFAG) design.

Accelerator Requirements

Beam Energy

For the production of spallation neutrons, protons are the obvious choice of beam particle. Although electrons are easier to accelerate, the production cross sections are much lower. There is no compelling advantage for deuterons or other heavier ions: they may produce a few more neutrons at a given energy per nucleon, but that energy is harder to achieve owing to the lower charge-to-mass ratio.

The number of neutrons produced as a function of proton energy depends on the target design, but broadly speaking it is well established that the production rate plateaus around 1 GeV. Figure 1 shows this quantity for a typical target: a lead cylinder of 30 cm length and 30 cm radius, as simulated by MCNPX [1].

This makes 1 GeV the obvious choice of energy of a proton accelerator for a spallation source, including an accelerator-driven subcritical reactor (ADSR), under the assumption that in the design it is easier to increase the current by some factor rather than increase the energy by the same amount. The exact choice is a matter of balancing this with costs and practicalities. Thus, MYRRHA, a prototype system [2], which is strongly limited by cost, has chosen to run at 600 MeV, whereas the European Spallation Source (ESS) [3], which aims to run at high neutron flux and is limited by space charge, has chosen 2 GeV to benefit from the lower current.

Limits to Reactivity: The Protactinium Problem

The number of neutrons required to produce a given power from a reactor depends on the reactivity, k_{eff}, through a factor $(1 - k_{\mathrm{eff}})$. The closer to criticality that one is prepared to allow, the fewer neutrons will be required and the easier the accelerator will be to design and build. Values chosen in different designs cover the range between 0.95 and 0.995. However, the choice is limited by the *protactinium problem*.

During running of a thorium ADSR, thorium is converted into uranium via protactinium:

$$^{232}\mathrm{Th} \rightarrow {}^{233}\mathrm{Pa} \rightarrow {}^{233}\mathrm{U} \rightarrow \text{fission products}$$

and a dynamic equilibrium is reached in which ^{233}Pa and ^{233}U are formed and destroyed at the same rate. If the accelerator is switched off, then the first and third steps stop as there are no neutrons to drive them. But the second step, being a decay, continues, with a half-life of 27 days. So when reactor operation is stopped the concentration of ^{233}U will start to rise, over a period of weeks, and the criticality will increase. This increase depends on the specific reactor design, but is typically around 1 %. Thus, an ADSR with k_{eff} above $\sim$0.99 will

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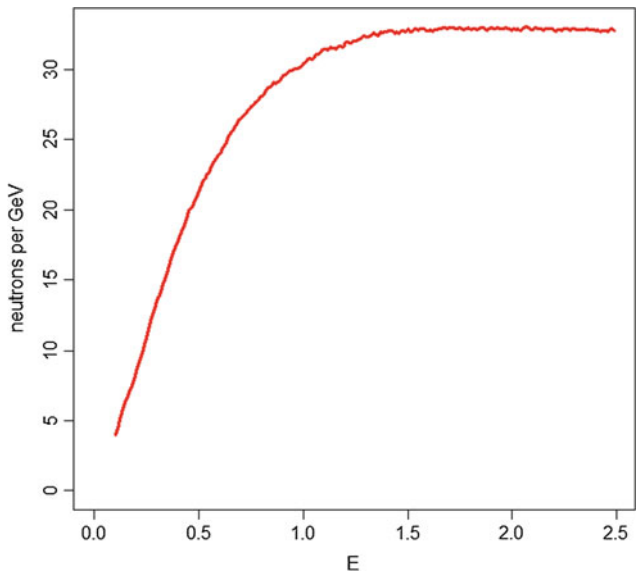

Fig. 1 Number of spallation neutrons per GeV of beam energy, as a function of that energy, produced from a 30 cm cylindrical lead target

require control rods, which must be inserted as part of the shutdown procedure. Such a design is not 'deterministically safe': there is a small, but non-zero, probability that accident, error, or sabotage may result in the rods being absent, leading to a criticality incident. By contrast, an ADSR with k_{eff} low enough that it stays below 1 even when all the ^{233}Pa has converted to ^{233}U just cannot go critical. We therefore take a value of 0.98 as an upper limit for ADSR design.

Beam Current and Machine Power

With these values, in approximate terms, each proton produces 30 spallation neutrons, and this number is enhanced by a factor $[1/(1 - k_{\text{eff}})]$, through the chain reaction; some 20 % of these neutrons induce fissions, and each fission yields 200 MeV. A 10 mA beam thus produces MW; enough for a mid-range power plant.

The typical accelerator needed for an ADSR is, thus, a 1 GeV, 10 mA (10 MW) proton machine. This power is somewhat beyond the range of today's existing and planned machines, as shown in Table 1, but building such a machine is not unthinkable.

Reliability

Although the power requirements are not so far above the present-day values listed, requirements for reliability far exceed present-day accelerator performance. These requirements arise from two causes. First, from commercial reasons, as long trips cause the reactor to cool and, thus, reduce/cease power generation, incurring financial penalties, which may be severe. Second, from an engineering perspective, as short trips will cause thermal changes and, thus, mechanical stress. One summary, taken from reference [4], is shown in Table 2.

These limits are orders of magnitude beyond those achieved by today's accelerators. It was stated during a previous session at this conference that the performance of the Large Electron-Positron (LEP) and Large Hadron Collider (LHC) accelerators at CERN showed that the desired reliability could be achieved. Such an assertion provides false comfort, and it cannot be taken at face value. These accelerators were and are magnificent achievements, but their reliability, though adequate for the needs of particle physics research, is nowhere near that required for an ADSR. The experience of users, and an examination of the control room logs, reveal many short and long breaks in delivery, for a wide variety of causes.Techniques for reliable engineering are well known from other fields, and include:

- Simple design using proven techniques;
- A holistic analysis of the system, so the interaction between components is understood;
- Redundancy in systems;
- Under-rating of components;
- Graceful failure;
- Automatic pick-up by other systems when one fails;
- Planned maintenance, testing, and replacing of components before they wear out.

These all cost money, and it is understandable that accelerators have not, up till now, been designed with

Table 1 Current and proposed proton accelerators

Machine	Energy (GeV)	Current (mA)	Power (MW)
PSI	0.59	2.0	1.2
ESS	2.0	2.5	5.0
CSNS-II	1.6	0.125	0.2
SNS	1.0	1.4	1.4
MYRRHA	0.6	3.5	2.1
ISIS	0.8	0.2	0.16

Table 2 Limits on trips/year for various lengths of trip (from reference [4])

Trip time	Limit (Engineering)	Limit (Commercial)
<1 s	25,000	–
1–10 s	2500	–
10 s–5 min	2500	250
>5 min	50	3

reliability as a prime criterion. This is something we have yet to learn.

The periods of maintenance pose a problem in that they entail downtime for the reactor. They will be especially necessary in the hostile environment of the accelerator, and will involve time delays while activated elements with short half-lives decay to levels where access for maintenance is possible. Although planned downtime is not as problematic as unplanned stoppages, it still involves a financial penalty. It is, therefore, attractive to postulate a system, extending the concept of 'redundancy', in which there are three accelerators to each reactor. Normally, two might be operational while the third is undergoing maintenance or on standby, and if one of the two has problems, the other could operate for a short period at an increased current to make good the deficit.

If the accelerators are linear accelerators (linacs) then such triple redundancy is prohibitively expensive. But if a cyclic accelerator could be used, then the cost, which is driven by the footprint, and the number of radio-frequency (RF) cavities, would be considerably reduced and the idea can be considered. So this gives a further incentive to the development of a cyclotron or fixed-field alternating gradient (FFAG) solution to the ADSR requirements, and one of each will be outlined in the following sections.

The DAEδALUS MultiMegawatt Cyclotron

DAEδALUS, the Decay At Rest Experiment for δ_{CP} At a Laboratory for Underground Science, is a proposed neutrino oscillation experiment using three $\bar{\upsilon}_{\mu}$ sources and one $\bar{\upsilon}_{e}$ detector [5]. In measuring the oscillations of neutrinos at different distances, the detector is more expensive than the source, so the scheme proposes three 800 MeV proton accelerators (each operating with a 20 % duty cycle) at distances of 1.5, 8, and 20 km, as shown in Fig. 2. The near, medium, and far accelerators run with powers of 1.0, 1.6, and 4.8 MW—values that put such machines in the region suitable for ADSRs.

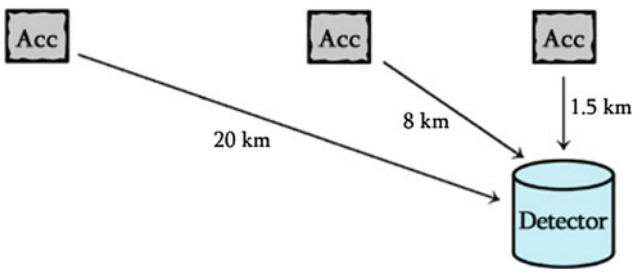

Fig. 2 Schematic design of the DAEδALUS experiment. The accelerators are on the surface and the detector is underground

The Main Cyclotron

An accelerator that meets the criteria has been designed [6]: the DAEδALUS Superconducting Ring Cyclotron (DSRC). It provides 4 mA of 800 MeV protons by acceleration of H_2^+ ions to 1.6 GeV (Fig. 3).

Extraction from the cyclotron has to be done by stripping as the orbits get very close. H_2^+ is chosen for this purpose as its binding energy of 2.75 eV is considerably greater than the 0.75 eV of the more familiar H^-. This avoids problems of spontaneous stripping in the strong magnetic fields, and also in the RF. The greater mass reduces effects of space charge by a factor of 2, quantifying the effect using the generalised perveance, K. The disadvantages of H_2^+ are the need for a higher vacuum (10^{-8} torr rather than the more usual 10^{-7}), and the greater magnetic fields required.

A thin (2 mg/cm^2) pyrolitic graphite foil, a few cm^2 in size is used for the extraction. A pulse width below 1 ms ensures the foil temperature does not exceed 2500 K: based on the experience at Spallation Neutron Source (SNS), foil lifetimes of several months are anticipated. The electrons produced are stopped on a copper shield. Any neutral hydrogen produced can be extracted safely.

The higher magnetic field required means that superconducting magnets must be used. There are six of these, with current densities of 3400 A/cm^2. (An earlier design [7] had eight magnets, but this did not leave enough room for the RF cavities).

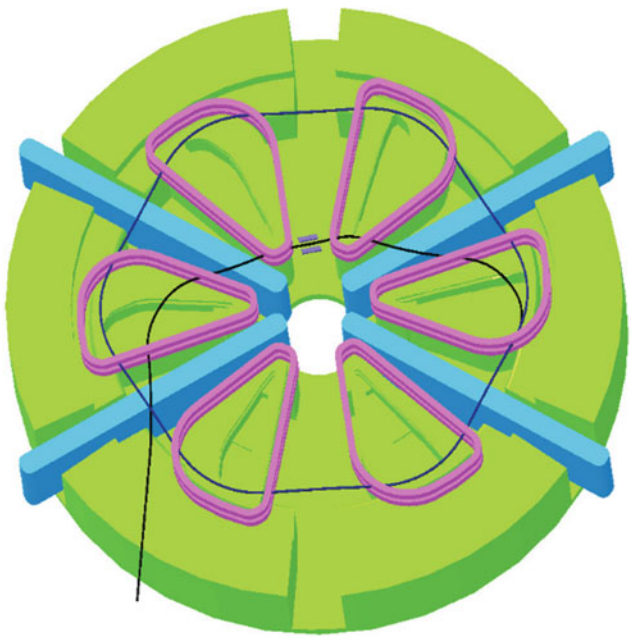

Fig. 3 The DSRC, showing the final orbit and extraction path. The H_2^+ ion dissociates into two protons, which bend twice as much in the magnetic field. Taken from reference [6]

The cryostat is made of iron and stainless steel (steel plates connect opposite segments) and is subject to large forces. The design, though challenging, is similar to the existing RIKEN Superconducting Ring Cyclotron (SRC); hence, the parameters are in general credible. A preliminary engineering study has been performed, using OPERA and ANSYS. Modelling has been done for the conductor, the high-temperature superconductor, and copper current leads for the magnet. The structural design of magnet cold mass, the cryostat and warm-to-cold supports, the cryogenic design of the magnet cooling system, and the magnet power supply have all been considered.

The RF design uses four single-gap copper cavities, 3 m high and 8.8 m long, operating at 49.2 MHz (the 6th harmonic), with a Q value of 37,500. These give acceleration from 2.0 to 4.0 MV per turn. The size, frequency, and power are similar to those of the Paul Scherrer Institute (PSI) cyclotron; hence, this design is also credible (It may be possible to add two double-gap cavities: this is under development.).

The lattice analysis is satisfactory. The isochronicity of 0.05 % is adequate, and the focussing is sufficient in both planes. The Walkinshaw resonance is crossed rapidly, and should not lead to beam losses. Simulations including the effects of space charge now are being done using the OPAL code [8] for the new six-sector model. No surprises are expected as the earlier eight-sector version behaved well.

The DSRC is fed by a separate injector cyclotron. The ion source has to provide 50 mA of H_2^+. 20 mA has already been achieved, and work is ongoing to improve this number. Experimental tests with the source, and the matching of the beams from it, are currently under way at BEST cyclotrons, in Vancouver.

A spiral inflector is used to inject the ions into the Injector Cyclotron at 70 keV (35 keV/amu). This is fairly conventional; it is 6.2 m in diameter and 2.7 m high, using four conventional 1 T sector magnets to accelerate 5 mA of H_2^+ up to 60 MeV/amu. Four 49.2 MHz double-gap RF cavities accelerate the bunches in 107 turns. The bunches can be cleanly extracted by two conventional electrostatic deflectors, as the orbit separation at extraction is large (2 cm). Simulations show that this machine is space-charge dominated, but 5 mA is achievable. The injector on its own could provide an interesting neutrino source (IsoDAR) and possibilities are being strongly pursued [9]. This would provide valuable experience of high currents and low (70 MeV) energies.

So the requirements for neutrino sources are motivating the development of a high current cyclotron suitable for ADSRs. This could be another instance, as with superconducting cable and the world wide web, where the demands of particle physics push technology forward, blazing the trail for the benefit of the applications that follow.

The Non-scaling Fixed-Field Alternating Gradient (nsFFAG)

The FFAG, Past and Present

A fixed-field alternating gradient (FFAG) accelerator appears somewhat similar to a sector cyclotron, but its magnetic fields increase or decrease strongly with radius. The sign of the gradient differs between magnets, though the increase outweighs the decrease. As the particles accelerate, their orbits expand and they experience an increasing mean field, with an alternating gradient, as they would in a synchrotron, even though the magnets themselves are DC. Such a machine is not naturally isochronous, like the classical cyclotron or the relativistic synchrotron, but with a suitable field configuration, the orbit period may be almost independent of energy, enabling the machine to run continuously at a constant RF frequency. The FFAG can, thus, combine the best features of a cyclotron (simple DC magnets and high currents), and a synchrotron (strong focussing and a smaller beam aperture).

The FFAG was invented [10] and, it is fair to say, abandoned in the 1950s. It could not compete with the synchrotron as a machine at the energy frontier. However, the concept was revived in the 1990s [11], when its other merits became appreciated, especially its large acceptance and its capacity for fast acceleration limited only by the RF rather than by the time for magnet ramping. Prototype proton machines at Kyoto University Research Reactor Institute (KURRI) have achieved 150 MeV [12].

The nsFFAG

In a conventional, or ‘scaling’ FFAG, a particle sees no change in the beam optics during the acceleration cycle: the bending and focussing fields increase at the same rate, just as they do in a synchrotron, where the dipole and quadrupole currents increase in step. This was felt to be necessary as changing optics entails changing tune, and if the tune were not held constant, it would at some point go through an integer value, and resonances would destroy the beam. To achieve this scaling, the field dependence has to have the

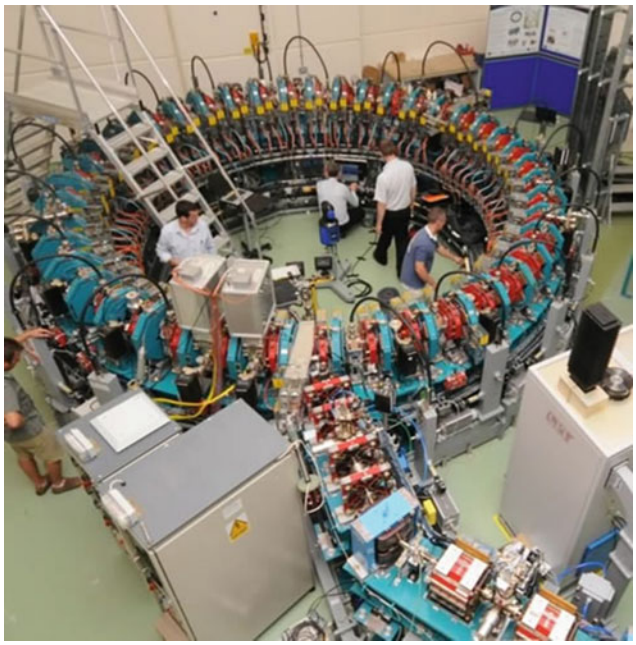

Fig. 4 EMMA under construction

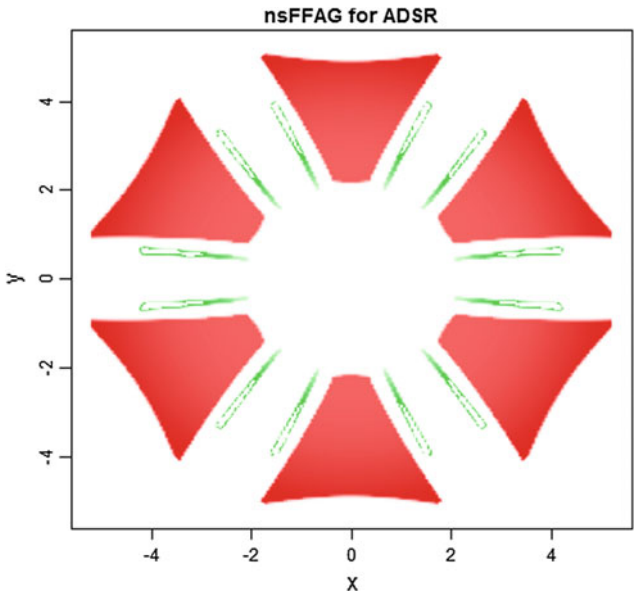

Fig. 5 Field map for a 1 GeV nsFFAG

form $B(R) \approx R^k$, where the constant k is called the Field Index. Even for large k, this means that the variation is slow, requiring a wide beam pipe and large-aperture magnets, adding considerably to the expense and complexity.

It was suggested [13] that the scaling requirement could be relaxed if the acceleration were fast enough. Although acceleration in a synchrotron is generally considered as a quasi-static process, where the energy change per orbit is small, for an FFAG this need not be true. Particle bunches may move through an integer tune value so rapidly that the cumulative effects of a resonance on betatron oscillations do not have time to build up. Without this restriction, the beam pipe can be made smaller and the magnet simpler. However, a test was required, as the lattice imperfections that give rise to the potentially disruptive resonant effects cannot be reliably simulated. The EMMA (Electron Machine with Many Applications) machine (Fig. 4), an 18 MeV electron nsFFAG, was designed, built, and commissioned at the Daresbury Laboratory [14], and its successful operation [15] validated the nsFFAG principle—or at least went some way towards doing so, as a proton nsFFAG is yet to be built.

An NsFFAG for ADSR

A potential field map for a 1 GeV proton nsFFAG is shown in Fig. 5, [16]. It is some 5 m in radius, a very modest size compared with a linac. There are six large bending magnets, the positive fields of which are shown in red in the figure, and twelve small counter-bends, the negative fields of which are shown in green. The designs of the magnets to produce these fields has not yet been performed; however, there is no reason to believe they are impossible. Figure 6 shows the field profiles through (a) the main magnets and (b) the counter-bends. The fields are large (up to 2 T for the bends and 5 T for the counter bends, showing that superconducting magnets will be required) and the field gradients are considerable.

From the field maps, one can find the reference orbits, shown in Fig. 7, for energies of 100, 150, 200, 300, 400, 500, 600, 900, and 1000 MeV. Injection, extraction, and acceleration are not yet considered, but there is sufficient space in the long straights for the insertion of kickers, cavities, etc., as required.

From the reference orbits, one obtains the isochronicity, shown in Fig. 8. This is fairly flat, though not at the parts-per-mille level obtained for isochronous cyclotrons. Whether this flatness is sufficient to enable operation at a fixed frequency requires further investigation: if this is not the case, then another iteration will be required on the field-map design.

The betatron tunes turn out to vary with beam energy, as expected, though not by very much and it may be possible to avoid major resonances.

The acceptance is very generous, especially at low and high energies. Particles deviating by as much as 10 cm/50 mrad from the reference orbit still survive for many turns.

These first investigations are promising; more detailed studies are needed and are in progress. If successful, they can lead to a full design for an nsFFAG system, which would have many advantages in bringing the ADSR concept closer to commercial reality.

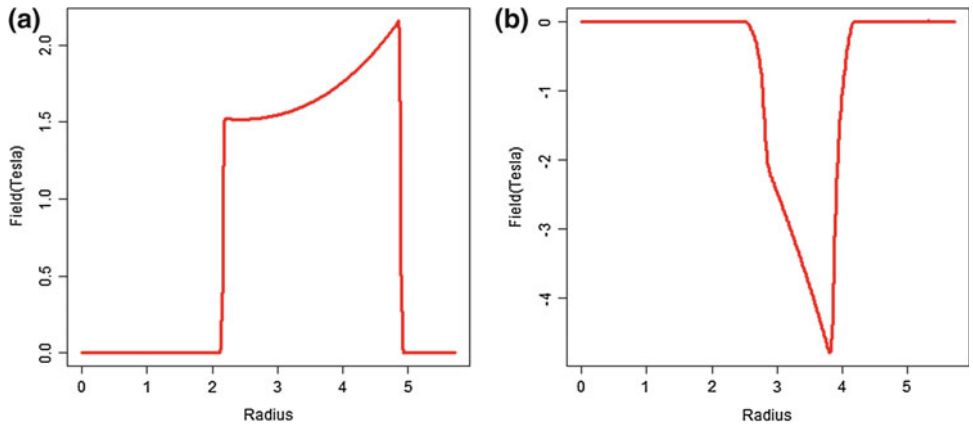

Fig. 6 Sections showing the field profiles through **a** the main magnets and **b** the counter-bends

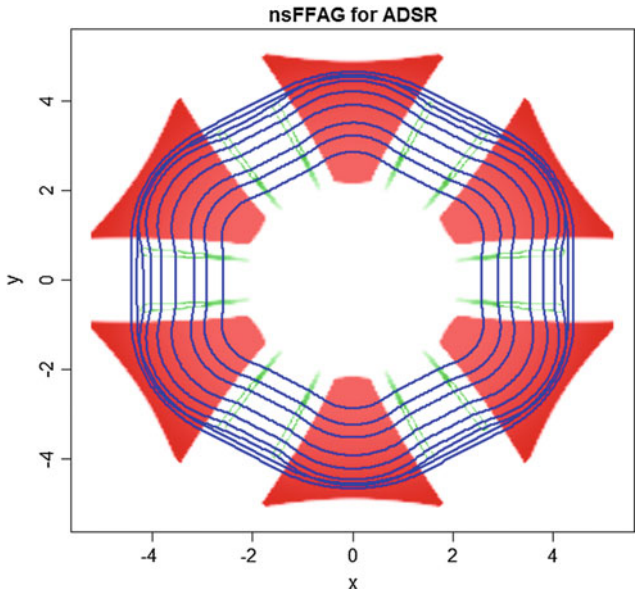

Fig. 7 Reference orbits in the ADSR nsFFAG

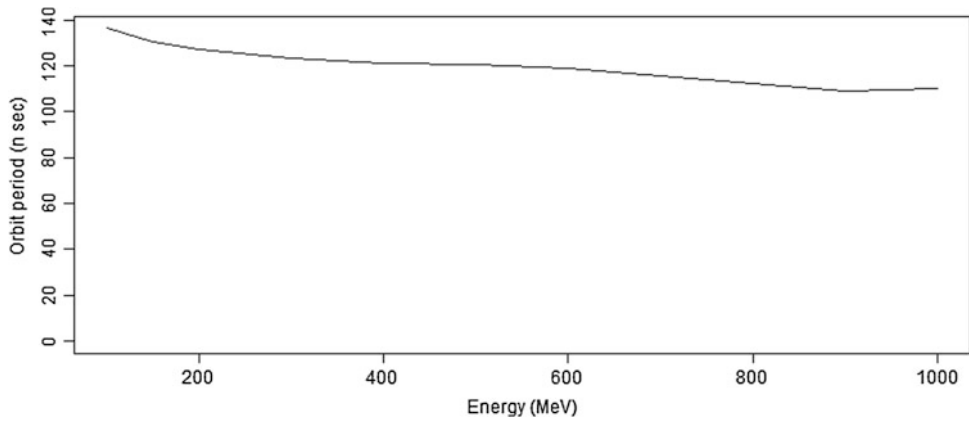

Fig. 8 Orbital period as a function of beam energy

Conclusion

ADSR systems will require proton accelerators operating at ~1 GeV and several MW, and with unprecedentedly high reliability. These requirements can be met with a linear accelerator, and for a prototype projects such as MYRRHA, this is the safe technological choice, but if one looks beyond the early stage to future commercial deployment, the large size and cost of a linac presents obstacles that may be overcome by using a cyclic machine. Designs for such a machine exist at a detailed level for the cyclotron and as a concept for the nsFFAG.

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Spallation Target Developments

B. Riemer, M. Wohlmuther, H. Takada, and N. Takashi

Abstract

Spallation targets withstanding beam powers of several megawatts are required for significant transmutation of waste in accelerator-driven systems (ADS). Such target systems are being designed and operated in current spallation neutron source facilities. This paper presents a brief overview of the development history of spallation targets used at several facilities. In particular, the spallation target development programs of the Swiss Spallation Neutron Source, SINQ, and of the Spallation Neutron Source, SNS, in Oak Ridge are reviewed.

Introduction

The production of neutrons through a spallation reaction of accelerated incoming particles on a target material with high atomic mass is now a mature technology. After decades of development, the megawatt range has been reached in the latest facilities. Spallation targets are broadly characterized by the power absorbed from the accelerator beam, which is itself a function of the beam current, particle type, and end energy.

As the power of the facilities has increased, the breadth of their application should rapidly extend beyond fundamental physics areas such as superconductivity research to increasingly applied science areas such as industrial tomography, radiotherapy, or indeed energy production as in accelerator-driven systems (ADS).

The latest results in the field of spallation target development give some indications as to the best development path that may be followed in the years to come, to expand their field of application to the greatest possible number of users.

M. Wohlmuther presented this paper at the Thorium Energy Conference 2013 (ThEC13)

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Spallation Target Development Roadmap

In the drive to extend the power of spallation targets, much has already been achieved, as illustrated in Fig. 1, which shows the capabilities of various facilities currently in operation around the world. Not listed are facilities being built in China (CSNS) and Sweden (ESS), which are in an advanced stage of development and are due to come on-line within the next few years.

In recent years, the trend towards ever-growing energy deposition rates has led to a departure from solid targets towards liquid metal targets as in the Spallation Neutron Source (SNS), Japan Proton Accelerator Research Complex (J-PARC), and partly at the Swiss Spallation Neutron Source (SINQ) at the Paul Scherrer Institute (PSI) [1]. This trend is not universal as a soon-to-be-built facility, the ESS, is currently examining a solid target variant, consisting of a rotating solid wheel. For this reason, a distinction should be drawn between:

- Static solid targets, in which a circulating fluid (water, helium, etc.) actively cools the solid;
- Liquid targets, where a liquid absorbs the beam deposition and is recirculated towards a heat exchanger, which removes the heat;
- Moving solid targets, in which a solid is actively cooled by a circulating fluid and is constantly removed from the beam deposition area; either by rotation such as in the

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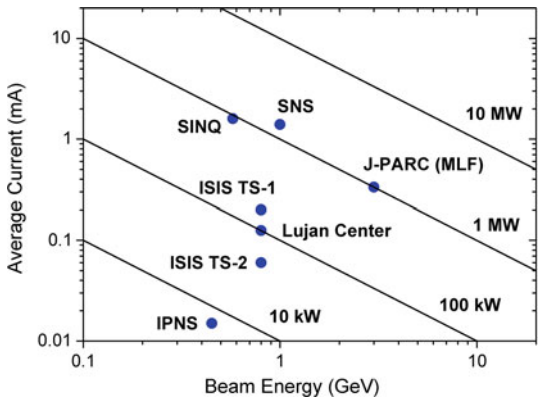


Fig. 1 Spallation neutron sources currently in operation

wheel concept, or through granular flow as in other concepts being pursued, for instance, in China.

The SINQ facility at the Paul Scherer Institute in Switzerland is a prime example of the shift between solid and liquid target technologies and gives some useful insights into their relative merit. The facility is somewhat unique in having a pro-ton beam entering a vertical target from below [1], a configuration originally chosen for a naturally circulating liquid metal target. The beam structure is not pulsed but continuous.

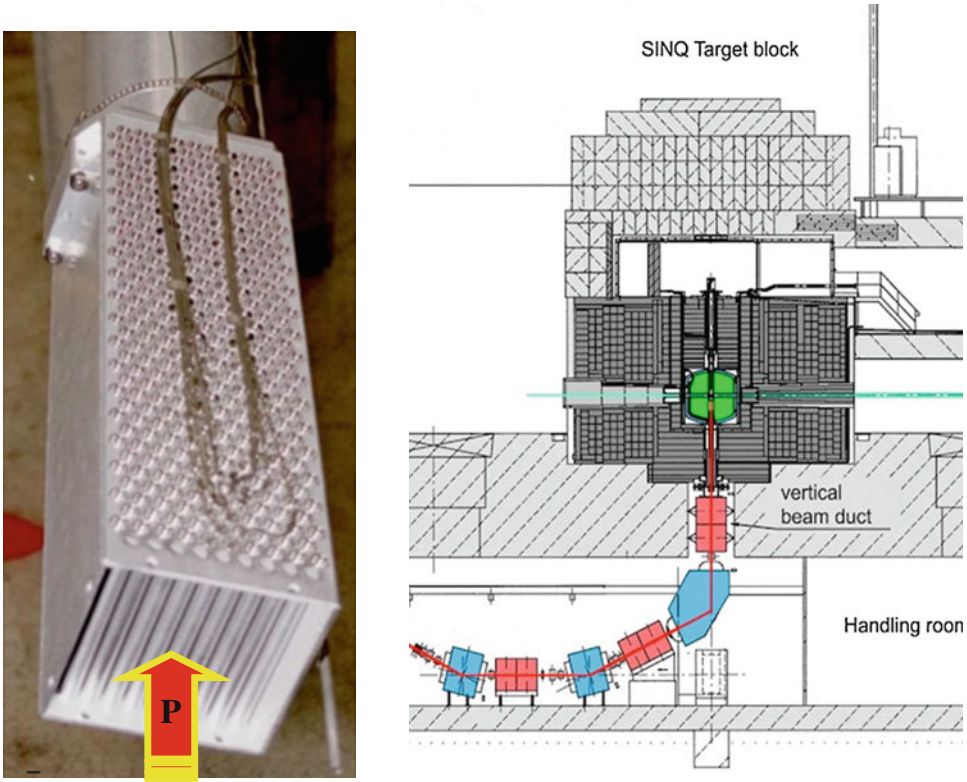
Spallation Target Development at SINQ

The MEGAPIE development, the world's first liquid metal spallation target was a landmark event in the history of SINQ [2]. It was, however, both preceded and followed by the development of solid targets, which have greatly benefited from the parallel development of both technologies.

Leading up to the installation of MEGAPIE in SINQ in 2006, the Mark-III target (Fig. 2) was in operation. Developed from previous versions in the late 1990 s, it featured a series of closely packed steel-clad lead rods, which were actively cooled by heavy water circulating around the tubes. The square box arrangement allowed installation of materials for the SINQ Target Irradiation Program (STIP) program in the tubes, a research effort still on-going and focused on studying materials under high doses (displacements per atom or dpa).

The MEGAPIE target, which was irradiated in late 2006 for a period of 4 months, is the subject of a dedicated paper at this conference [3], and its success in furnishing an 80 % increase in the neutron field density inspired a series of changes to the SINQ Mk. IV target that followed. By keeping with solid target technology, a simplification of operational constraints was sought, but some of the changes that were a success to MEGAPIE were incorporated, notably:

Fig. 2 Mk.-III SINQ spallation target (*top*) and target in SINQ facility (*bottom*)



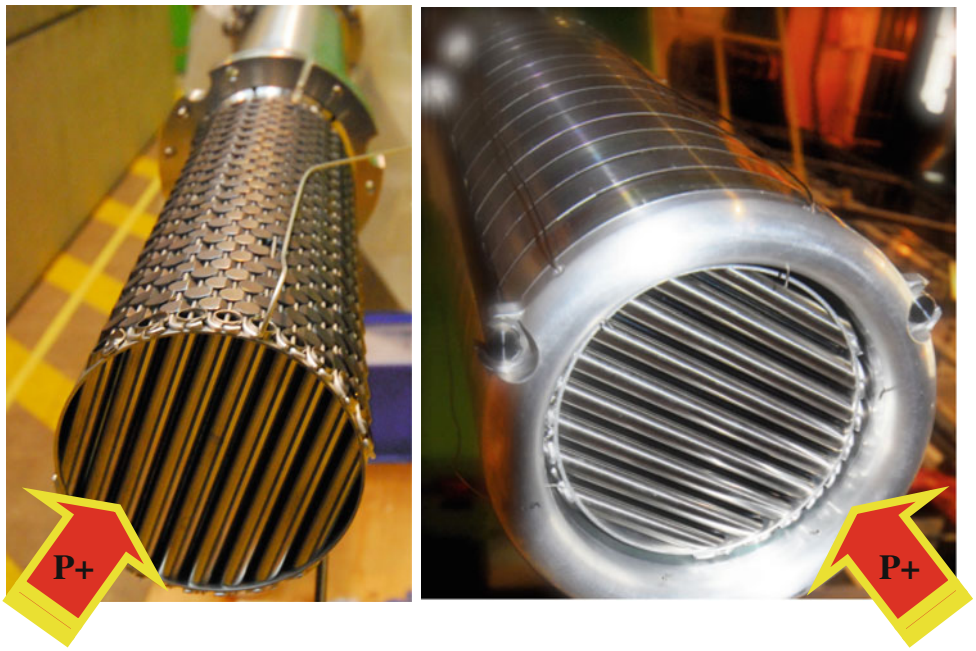


Fig. 3 Mk.-IV SINQ spallation target lead tubes (*top*) and guide tube (*bottom*)

- A streamlined guide tube to reduce pressure losses;
- Compact arrangement of the rods to diminish beam losses to the D_2O;
- Safety shroud from MEGAPIE with integrated leak detection capability.

The geometry of the Mk. IV target, shown in Fig. 3, thus allowed an increase of 60 % in the neutron flux density compared with the pre-MEGAPIE model. Although this remains less that the 80 % achieved in MEGAPIE, the relative ease of operating a solid target overweighed the small difference in performance. The lifetime of the target is 2 years and is primarily limited by material embrittlement and fatigue caused by numerous beam trips/interrupts.

The latest performance figures for the SINQ Mk. IV solid target are given in Table 1 and compared with those of SNS.

Table 1 Characteristics of two high-power neutron spallation targets

1. SINQ Mk. IV solid target	
Proton beam energy	600 MeV, 1.6 mA/0.96 MW
Beam profile	σ_x = 2.14 cm, σ_y = 2.96 cm
Peak current density	40 μA/cm^2
Peak power deposition	820 W/cm^3
Coolant	D_2O @ 8 Bar
Coolant inlet temperature	30 °C
Coolant outlet temperature	70 °C
Coolant flow speed	~1 m/s
2. SNS target 6	
Proton Beam energy	1.2 MW
Beam profile	σ_x = 10 cm, σ_y = 3.5 cm
Peak current density	~10 μA/cm^2
Peak power deposition	480 W/cm^3
Coolant	Hg @ 3 Bar
Coolant inlet temperature	60 °C
Coolant outlet temperature	90 °C
Coolant flow speed	3.5 m/s

Fig. 4 SNS spallation target

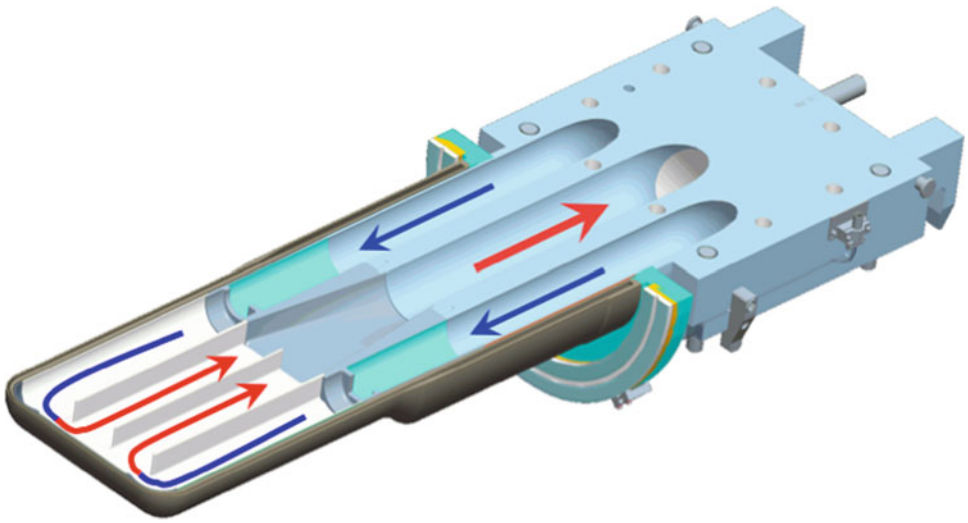


Fig. 5 SNS beam window inner surface sample extracted after irradiation

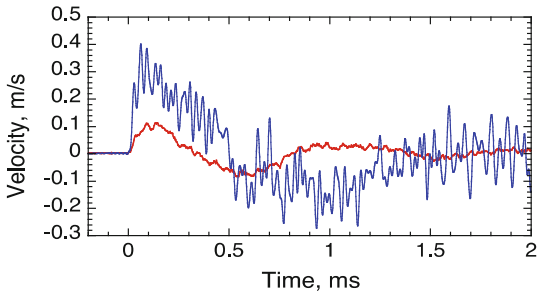


Fig. 6 Laser Doppler vibrometer measurements at J-PARC on the target beam window with a bubble curtain (*red line*) and without (*blue line*)

Spallation Target Development at SNS

Oak Ridge National Laboratory (ORNL) in the USA is now operating one of the most powerful neutrons spallation sources, based on liquid metal technology, in this case, mercury. Mercury lends itself well to absorbing large energy depositions at low temperatures, but suffers from the inconvenience of a low boiling point relative to other liquid metals such as lead–bismuth eutectic, for instance.

The SNS target is essentially a flat conduit funneling mercury at high speed towards a central spallation zone form where the heat is evacuated by the liquid metal to a secondary heat exchanger further downstream, outside the irradiation area. The beam penetrates the forward section of the target through a very thin beam window, which is exposed to a variety of stress intensities (Fig. 4).

Early on, it became evident that the beam window was being eroded far more rapidly than had been anticipated. The damage caused by cavitation shows up quite clearly in the sample extracted from the beam window, the inner surface of which is illustrated in Fig. 5. It was believed that the likely cause would be a pressure wave.

The postulated pressure wave is itself the result of the beam pulse, which causes sudden thermal expansion in the liquid metal in the beam deposition area. This pressure wave then propagates through the liquid medium until it impacts the inner surface of the target. The peak damage occurs where the local velocity of the liquid metal coolant on the inner surface is sufficiently low, such that the pressure wave is able to impinge on the surface.

The interaction between the impinging pressure wave and the structure was much debated, but it is likely that high-frequency vibration of the structure ensues, which causes cavitation at the interface.

The severity of the problem at SNS caused numerous early shutdowns to re-place the entire target. Ultimately, a solution could be found, which led to a collaboration with J-PARC that was experiencing similar problem on its Japan Spallation Neutron Source (JSNS) target.

Spallation Target Collaboration with J-PARC

Given that the cause of the problem was cavitation at the surface, some mechanism had to be found that could interfere with the impingement of the cavitation bubble. As cavitation is essentially the formation of a void, it seemed reasonable to attempt to fill up this void by compressing the fluid locally in the region. A solution to the problem could be found by injecting a curtain of small helium bubbles, which, by expanding in the fluid on the inner surface of the beam window, essentially suppress cavitation before it can impinge on the inner surface and cause cavitation damage.

The proof of the effectiveness of this remediation technique is shown in Fig. 6, which indicates how the bubbles decrease the high-frequency structural vibration as measured by a laser Doppler vibrometer. High-frequency vibration is generally known to be an indication of cavitating surfaces.

Conclusion

Recent experiences gathered in the development of high-power spallation tar-gets show that it is an extremely dynamic and even, at times, quite competitive field of technology. The drive to produce ever-higher power levels requires taking a fresh look at the fundamental technology choices that have been made in the past with a clear focus on the needs of the user, which in turn demands a reliable supply of neutrons for experiments in the expanding field of neutron science. Hence, the choice need not be lead to any particular concept, but rather to a blend of technologies, always taking into account latest development from the neutron science community at an international level.

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MEGAPIE: The World's First High-Power Liquid Metal Spallation Neutron Source

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Abstract

A key experiment in the accelerator-driven systems (ADS) roadmap, the MEGAwatt PIlot Experiment (MEGAPIE) (1 MW) was initiated in 1999 in order to design and build a liquid lead–bismuth spallation target, then to operate it at the Swiss spallation neutron facility, SINQ, at the Paul Scherrer Institute (PSI). The target has been designed, manufactured at ATEA (France), then set-up, fitted with all the ancillary systems, on an integral test stand at PSI for off-beam tests dedicated to thermo-hydraulic and operability tests, carried out during the last months of 2005. The system was then placed with its ancillary system in the SINQ facility, for final operation under irradiation, which was carried out from August 14th to December 21st, 2006. The irradiation allowed very important results to be obtained, including those showing the very good operational behavior of the target. Moreover, a lot of results in the thermal hydraulic and neutronics fields, and gas analysis contributed to validating the design studies and the assumed theoretical models. Decommissioning of the target has been carried out and PSI teams have prepared samples (760) for post-irradiation examination (PIE). The samples have been distributed to the following partner laboratories: Commissariat à l'énergie atomique et aux énergies alternatives (CEA), Karlsruhe Institute of Technology (KIT), Paul Scherrer Institute (PSI), SCK·CEN, and the US Los Alamos National Laboratory (LANL). The Centre national de la recherche scientifique (CNRS) and ENEA will also contribute to the PIE to be carried out in the SCK-Mol and PSI hot laboratories, respectively. The goal is to study the changes to the structural material properties caused by the harsh environment of high temperatures, contact with flowing liquid metal (lead–bismuth eutectic, LBE), and proton irradiation. After presenting an overview of the project history, we describe the target decommissioning, PIE sample

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preparation and distribution, and highlight the first results that have been obtained. The successful behavior of the target and the coming results about irradiated materials will provide to ADS community with unique operational feedback, paving the way to develop high-power spallation targets as neutron sources and for future ADS-based options for the transmutation of nuclear wastes.

Introduction

Partitioning and transmutation (P&T) techniques could contribute towards reducing the radioactive inventory of nuclear waste and its associated radiotoxicity. Subcritical accelerator-driven systems (ADS) are potential candidates for dedicated transmutation systems, and thus their development is a relevant research and development topic in Europe. A key experiment in the ADS roadmap, the MEGAwatt PIlot Experiment (MEGAPIE) (1 MW) was initiated in 1999 in order to design and build a liquid lead–bismuth spallation target, then to operate it at the Swiss spallation neutron facility, SINQ, at the Paul Scherrer Institute (PSI) [1]. The MEGAPIE project is supported by an international group of research institutions: PSI (Switzerland), Commissariat à l'énergie atomique et aux énergies alternatives (CEA, France), Karlsruhe Institute of Technology (KIT, Germany), Centre national de la recherche scientifique (CNRS, France), ENEA (Italy), SCK·CEN (Belgium), US Department of Energy (DOE, USA), Japan Atomic Energy Agency (JAEA, Japan), and Korea Atomic Energy Research Institute (KAERI, Korea). The European Union Commission has contributed financially to some phases through EU research Framework Program contracts (FP4 to FP6) for the design and currently post-irradiation experiments (PIE).

Three main constraints were faced for the design:

- Design of a completely different concept of target in the same geometry of the current spallation targets used at PSI;
- Develop and integrate two main prototypical systems: a specific heat removal system and an electromagnetic pump system for the hot heavy liquid metal in a very limited volume;
- Design of a 9Cr martensitic steel (T91) beam window able to reach the assigned life duration [2]. Lead–bismuth eutectic (Pb 44.5 %–Bi 55.5 %) has been selected, owing to its attractive neutronic and physical properties, including its heat transfer coefficient, low melting point (125 °C). However, Bi induces the production of activation products such as polonium.

Description of the Target

The target has been designed by CNRS, CEA, IPUL, and PSI (Fig. 1), [2, 11]; the components have been manufactured in France by the ATEA Company and sub-contractors and in Latvia (EM pumps), then it was assembled in France. The ancillary systems have been designed and manufactured in Italy and Switzerland (Ansaldo, Criotec, PSI) [3].

Thermal energy deposited in the LBE in the bottom part of the target (650 kW) is removed by forced upward circulation by the main in-line electromagnetic (EM) pump through a 12-pin heat exchanger. The heat is evacuated from the target heat exchanger (THX) by intermediate diathermic oil and an intermediate water-cooling loop to the PSI cooling system. The cooled LBE then flows down in the outer annulus (4 L/s). The beam entrance window, which is welded to the lower liquid metal container (LLMC) and includes the beam window, both of which were manufactured with T91 ferritic/martensitic steel, is specially cooled by a cold LBE jet extracted at the THX outlet and pumped by a second EM pump (0.35 L/s) through a small diameter pipe down to the beam window. The main flow guide tube, equipped with thermocouples, separates the hot LBE up-flow from the cold down-flow in the outer annulus. Attached to the top of the tube is the electromagnetic pump system, designed by IPUL (Institute of Physics in Latvia), consisting of a concentrically arranged by-pass pump and the in-line main pump on top of it. Both pumps are equipped with electromagnetic flow meters, and are surrounded by the heat exchanger, designed by CEA. A central rod is inserted inside the main flow guide tube, carrying a 22 kW heater and neutron detectors, provided by CEA. The lower liquid metal container, the flange of the guide tube, and the heat exchanger constitute the boundary for the LBE, called the hot part. The second boundary is formed by three components, which are

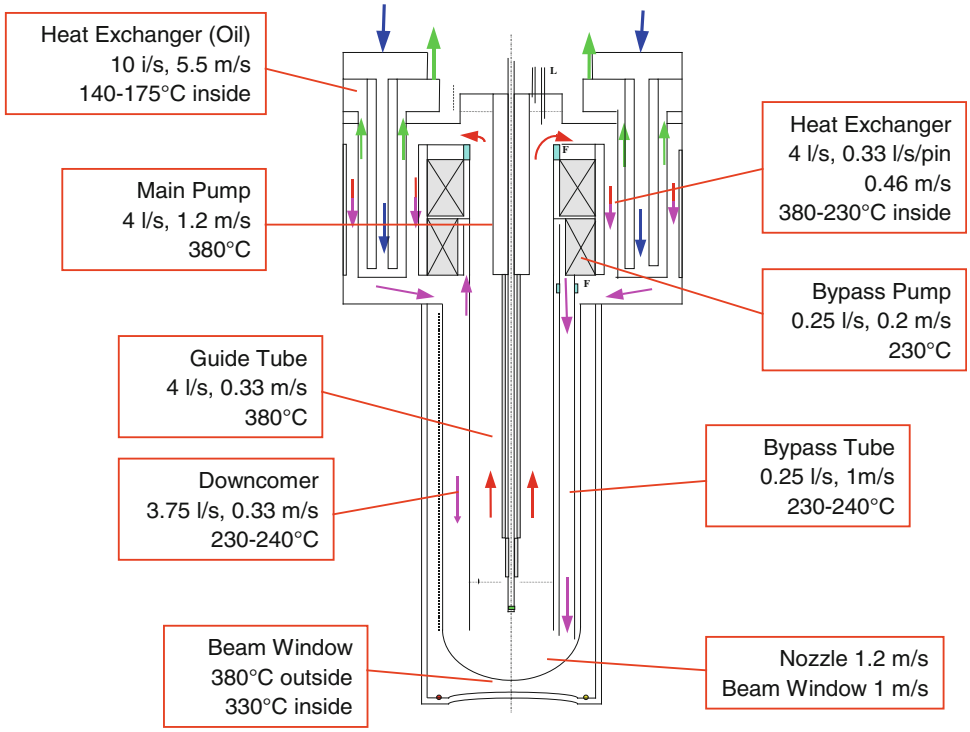

Fig. 1 MEGAPIE target

separated from the inner part by a gas space filled with 0.5 bar He. The target contains tungsten to shield the head area from the intense radiation of the LBE and the noble gases and volatiles collected in the gas expansion tank.

The Road to Irradiation of the Target

Many of the studies that were carried out by the project partners addressed specific critical issues in the fields of neutronics, materials, thermal hydraulics, mass and heat transfer, structure mechanics, and liquid metal technology, by employing analytical, numerical, and experimental approaches. Moreover, it was necessary to perform safety and reliability assessments in order to demonstrate the integrity and operability of the target and, thus, to develop the licensing process. To reach this goal, the design had to mainly consider the structural integrity of the target under normal operating conditions, transient situations, and hypothetical accidents, as well as the capability to evacuate the deposited heat with the heat exchanger and the electromagnetic pump system. The target was manufactured in Nantes (France) then shipped to PSI in May 2005 [3]. To demonstrate its characteristics and safe operability prior to irradiation in 2006, the target was first installed, fitted with all the ancillary systems, which had already been commissioned, and then it was tested out-of-beam.

The integral tests consisted of the following main tests [6]:

- Filling of the target with lead–bismuth eutectic;
- Checking the operability of the main components of the target (heat exchanger, electro-magnetic pumps, etc.);
- Checking and calibration of the instrumentation (specifically the flow-meters);
- Carrying out the thermo-hydraulic tests with a heater to simulate heat deposition;
- Performance of transients for qualification of heat removal and control systems.

At the end of the integral tests, the central rod of the target was cleaned, then the neutron flux detector provided by CEA was inserted. The electrical cabling and other connections were installed in the target head. The LBE leak detector was then installed, prior to the final welding of the lower target enclosure, with a qualified procedure. The target was then installed in SINQ and connected to the ancillary systems, including the fill and drain, heat removal system, cover gas system, isolation gas system, etc.

To fulfill the requirement of 1 mSv criterion for public safety in case of incidental release, some measures for the

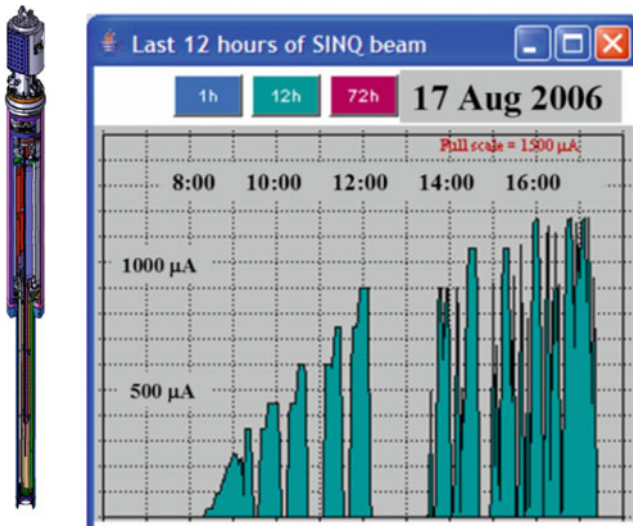

Fig. 2 Third phase of the start-up procedure

reduction of the source term were decided and carried out, including:

- Better sealing of the buildings over and below the target (chamber housing the target head and ancillaries: TKE and STK), when installed at SINQ;
- Implementation of an inertization system using nitrogen injection to prevent inflammation by the thermal oil under the most extreme conditions;
- Connecting the TKE with the cooling plant to reduce the possible activity concentration in air;
- Upgrades of the ventilation system (earthquake resistant stand-alone exhaust equipment) and the filter systems (with both activated carbon and particle filters).

Then, the target was ready to be operated following the three main operational modes [3]: isolation case, hot standby case, and beam operation case.

The first beam was received on August 14th, 2006. The start-up phase was successfully accomplished on August 15th (Fig. 2). Steady-state MEGAPIE operation for normal users was started on August 21st around 8:30 a.m. and continued until the normal annual winter shut-down, which started on December 22nd, 2006.

During the entire MEGAPIE irradiation experiment, the target served as the source for the neutron scattering program at PSI with respect to the required availability of neutrons: full power at >95 % of the scheduled operation time with a continuous (51 MHz) 590 MeV proton beam hitting the target and expected to reach routinely an average current around 1350 µA, corresponding to a beam power ~0.80 MW. Numerous operating parameters were monitored, including pressure, fluid flow rates and temperatures, neutron fluxes, etc. The target behavior was considered very satisfactory both during stable operation and transients due to beam trips. The temperature distributions and transients were as expected, very close to predictions. The electromagnetic pumps (EMP) operated stably and reliably, without any indication of degradation so far. Conversely, the performance of the electromagnetic flow meters (EMF), particularly in terms of accuracy, were low: difficulties were mainly induced by the proximity to the EMPs (leakage of magnetic flux) and by transients due to immersion in LBE (>10 °C/s). For the main MEGAPIE ancillary systems directly connected to the target, that is, the heat removal system (HRS), the cover gas system (CGS), and the insulation gas system (IGS), the operating feedback was generally positive [3]. The experience with the safety devices implemented to monitor the proton flux from SINQ was also very positive: the VIMOS camera was replaced and worked satisfactorily; the stripe LBE leak detector gave satisfactory results, but the TC LBE leak detector was considered to be the most sensitive beam diagnostic. Experience from the post-test analysis of MEGAPIE was analyzed carefully by the teams involved in the project for the various following items: thermal hydraulics, code verification, lower target thermal hydraulics, target heat exchanger performance, temperature control performance, neutronic and nuclear measurements, gas production. The main results were reported in [4, 5, 7, 8].

Decommissioning Phase

After irradiation, the target remained in the operating position until mid-February, 2007, when the decay heat had decreased to about 300 W. Controlled freezing of the LBE was necessary owing to expansion of solid LBE after re-crystallization: the expansion can be mitigated if the cooling rate is kept as low as 0.02 °C/min from the solidification point to 60 °C. A specific procedure for freezing the LBE in the lower target enclosure was carried out. Then, the cooling circuits and gas volumes were emptied, rinsed, and dried. The target was disconnected and sealed with blind flanges, stored for several months, transferred to the ZWILAG hot laboratories by using a steel container (Fig. 3), then it was cut into 19 slices with a band saw (provided by Behringer). About 8 wt% of the target was transported to the PSI hot laboratories as samples for post-irradiation examinations. The remaining target pieces (92 wt%) were conditioned in a steel cylinder in a KC-T12 concrete container (TC2), for storage and disposal.

Visual inspection of the T91 window was performed and this showed some materials deposited in the central area of the window; however, after removing the materials, no cracks or other damage were observed. Scanning electron microscopy (SEM) investigations indicated that the deposited materials have a pore structure and consist of carbon and

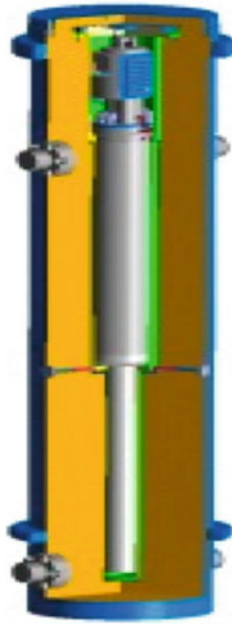

Fig. 3 Container for target transportation

oxygen with little silicon for the black material and Si and O with a little C for the white material. Moreover, ultrasonic measurements showed no detectable change in the thickness of the T91 window, which implies no evident (dissolution) corrosion effects. This observation validates the choice that was made initially to not control the oxygen activity in the LBE.

Post-irradiation Examinations

The objectives of the post-irradiation examinations are to understand:

- microstructural, mechanical, and chemical changes in the structural materials in the target induced by irradiation and LBE corrosion;
- the production, distribution, and release of the spallation and corrosion products in the LBE.

The PIE is carried out by an organized effort of the eight partners of the MEGAPIE initiative: CEA, CNRS, ENEA, FZK, JAEA, LANL-DOE, PSI, and SCK.

In the MEGAPIE PIE program, mechanical tests and microstructure analyses will be performed on different components, particularly on those in the lower part of the target with high irradiation doses. After segmenting the target in ZWILAG, small specimens for the PIE had be cut from the pieces with an electrical discharge machining (EDM) machine and a diamond disc saw in PSI's hot cells (Fig. 4).

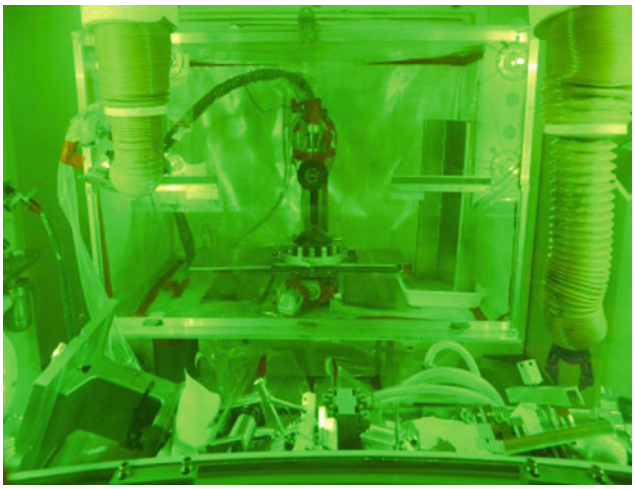

Fig. 4 EDM cutting machine

Suitably prepared specimens of the target are foreseen to undergo mechanical tests and microstructure analyses, including by TEM (transition electron microscopy), OM (optical microscopy), SP (small punch), tensile and bending specimens. The OM specimens will be also used for other surface analyses, such as SEM, EPMA (electron probe micro-analysis), etc. (Fig. 5).

As requested by some partners, specimens should be contamination free. This induced the necessity to clean the structures to remove the LBE; the cleaning was performed in a hot cell after melting the LBE and segmenting the larger pieces into smaller ones. A suitable method to remove LBE was to sweep the LBE away with tissues after heating the specimens in oil to about 150 °C. To dissolve the remaining LBE on the surface of the samples, two different solutions were used: the first was an equimolar mixture of acetic acid, hydrogen peroxide, and ethanol; the second was 7 M HNO_3. After tests, it was decided to use nitric acid for the final

Fig. 5 Samples produced by EDM machine

cleaning of LBE owing to higher cleaning kinetics, less gas and heat production, and less effluents.

Concerning the transport of the PIE samples to the partner laboratories (CEA, JAEA, KIT, LANL, SCK-Mol), investigations on the transportability have been performed based on calculations with detailed nuclide inventories for all samples. Transports have been performed as "UN2915, Radioactive Material, TYPE A" transports. All the transports were completed by the end of April, 2013.

The irradiation parameters, such as irradiation dose (dpa), helium and hydrogen production, proton and neutron fluences, irradiation temperature, etc., had to be determined for all the specimens under investigation. Except for the irradiation temperature, the remaining parameters can be evaluated from the final (accumulated) proton fluence distribution in the target by neutronic calculations. The irradiation temperature relies mainly on the instant proton beam current density, which is known to change significantly but, unfortunately, could not be measured during the operation of the target. To evaluate the distribution the proton fluence accumulated after about four months of irradiation, gamma-mapping on the beam window of the aluminum safety hull of the MEGAPIE target was conducted. The gamma-mapping was performed on an area of 160 × 160 mm^2 in steps of 4 mm. At each measuring position, a full gamma spectrum was obtained. ^{22}Na was the only radioactive isotope detected well, which was mainly produced by the incident high-energy protons. To deduce the proton fluence distribution from the measured data, the ^{22}Na counts had to be corrected because of differences in actual measuring volume and the distance at different positions on the window. The distribution of proton fluence was obtained and was available for neutronic calculations of irradiation parameters. The results show that the maximum proton fluence of the target is 1.93×10^{25} p/m^2 (Fig. 6).

Thermal hydraulics calculations have been carried out with a computational fluid dynamics (CFD) model to evaluate the temperature distribution in the lower part of the target, and compared with temperatures measured by thermocouples. Some temperatures were measured at about 15 °C higher than predicted. Some wrongly attributed positions of the thermocouples could explain some of the calculated/experimental discrepancies. A significant difference of temperature was calculated between the inner side and outer side; up to 25–30 °C at the flow guide edge (Fig. 7).

There were also instabilities, particularly due the effect of the by-pass pipe, and seen thanks to experiments with infrared cameras during the integral tests. Large eddy scale

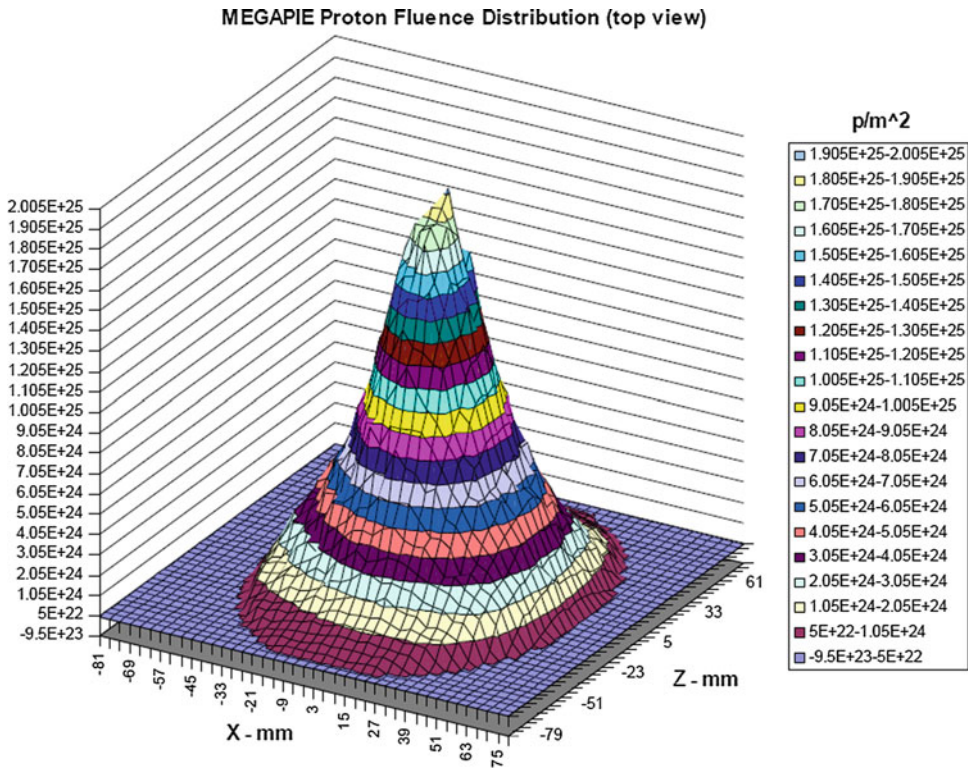

Fig. 6 3D view of the proton fluence distribution

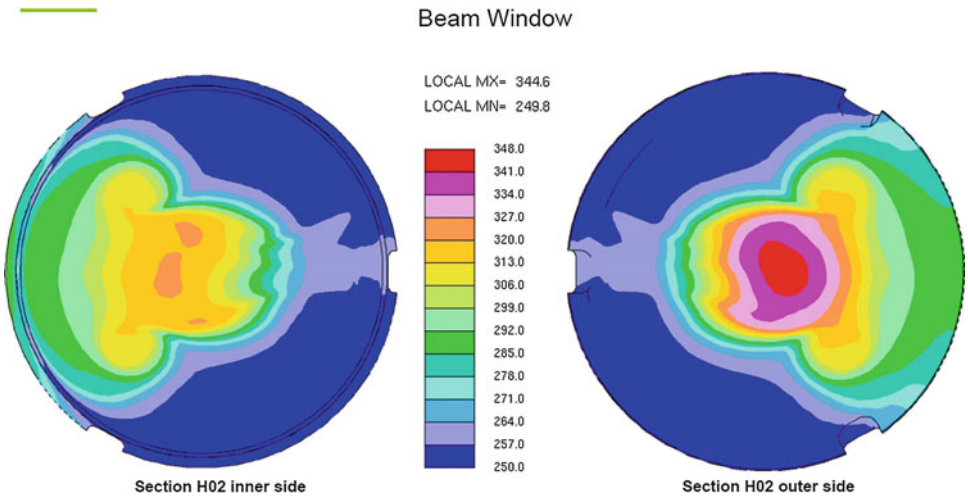

Fig. 7 Computed temperatures of the window

simulations (LES) were carried out close to the window (using TRIO-U-VEF parallelized code) to analyze these instabilities, and, consequently to assess the level of temperature and velocity fluctuations near the window to give realistic data for thermomechanical studies aiming to demonstrate the integrity of the T91 window (Fig. 8). These data also show the difficulty in attributing an average temperature of LBE (T_{av}) close to the window. Nevertheless, T_{av}, should be recalculated using the heat deposition data from neutronic calculations.

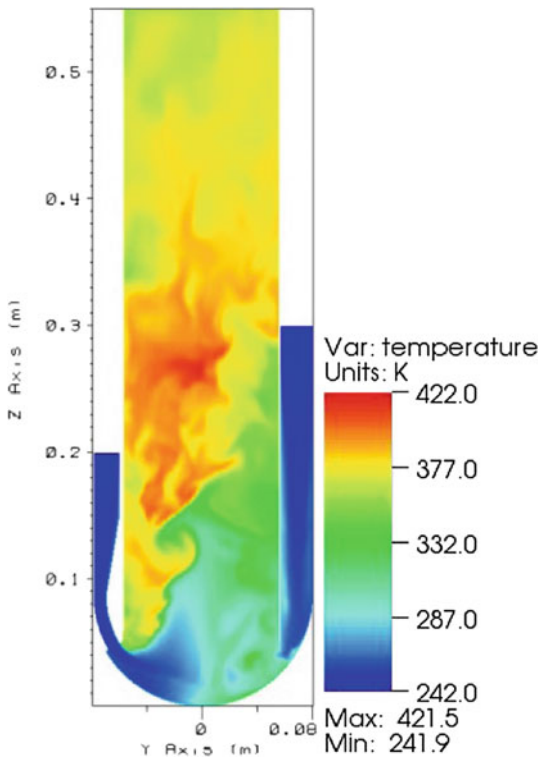

Fig. 8 Large eddy scale simulations (TRIO-U-VEF code)

It was decided to perform chemical and radiochemical analyses on the spallation and corrosion products in the LBE and deposited at the Ag absorber and cold-trap (control gas system). Samples were taken from all slices before the LBE was melted out in order to prepare the samples for mechanical testing. The size of the LBE samples that were taken was determined by the total activity that can be handled in the laboratories. The distribution of the activity within the target material was not found to be homogeneous because of different phenomena, such as deposition, transport during solidification, and diffusion. For an estimation of the maximum activity, we have relied on the activation calculations. The maximum activity that can be present in the LBE can be calculated by assuming homogeneous distribution. This assumption was surely conservative, as the deposition of radioactive material led to a decrease of the LBE activity. Such phenomena have been observed in model experiments as well as in a liquid LBE target irradiated at CERN ISOLDE. When the slices arrive at the hot laboratory, the composition at the surface will probably have been influenced by smearing of material during the sawing process and/or contamination with sawdust. Therefore, at each individual sample position, a pre-drilling/cutting is necessary to remove the contaminated surface.

The following results from chemical characterizations have been obtained:

- γ-spectrometry measurements of all specimens (as is, without prior chemical separations) have been completed. Results of ^{207}Bi, ^{194}Hg/Au, and ^{173}Lu distribution in the MEGAPIE target are reported in [9];

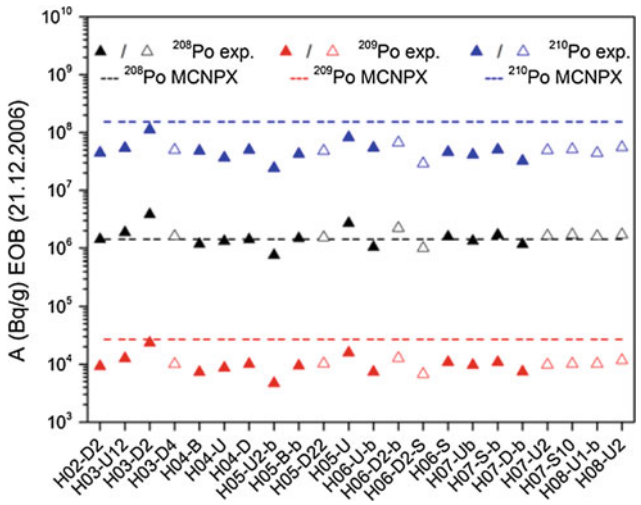

Fig. 9 Example of measured/modeled activity (Po)

- The following radionuclides have been identified by γ-spectrometry: ^{60}Co, ^{101}Rh, ^{102}Rh, ^{108m}Ag, ^{110m}Ag, ^{133}Ba, ^{172}Hf/Lu, ^{173}Lu, ^{194}Hg/Au, ^{195}Au, and ^{207}Bi;
- For some of these nuclides, the activities can be easily evaluated from γ-spectrometry results (e.g., ^{207}Bi, ^{194}Hg/Au), whereas other nuclides can only be determined after chemical separation (e.g., ^{108m}Ag, ^{110m}Ag, ^{195}Au, ^{129}I, ^{36}Cl and α-emitting $^{208-210}$Po) [9, 10];
- Bulk LBE contains only noble metals that have a significant solubility in LBE;
- Radionuclides of elements that have only low solubility in LBE or are sensitive to oxidation are only detected in samples taken at the LBE/steel interface and the LBE/cover gas interface;
- Accumulations on the walls increased dose rates and locally increased decay heat;
- ^{129}I is lower than expected; $^{208-210}$Po is homogeneously distributed [10];
- The α activity is well-predicted with FLUKA and MCNPX codes (Fig. 9).

In Fig. 9, filled triangles show bulk LBE samples and non-filled triangle represent samples that were taken from the LBE/steel and LBE/cover gas interfaces.

Complementary to these characterizations, PSI performed studies dedicated to the separation of spallation products from the LBE by using alkaline extraction, which was found to be suitable for Po extraction in nuclear systems [12]. For this purpose, irradiated LBE samples from CERN ISOLDE have been used for model experiments. For the extraction of Po isotopes, maintaining reducing conditions and avoiding any ingress of moisture guarantee their fast and reliable separation. Such purification would decrease radiological concerns during maintenance and shutdown of the facility. Nevertheless, several crucial problems remain to be solved before such a separation method may be licensed on an industrial scale, namely, the high corrosivity of molten hydroxides as well as their hygroscopic properties.

Conclusion

The target was designed, manufactured, and tested in integral tests, before irradiation was carried out at the end of 2006. During irradiation, neutron and thermo-hydraulic measurements were performed, allowing deep interpretation of the experiment and validation of the models used during the design phase. The decommissioning, post-irradiation examinations, and waste management phases were defined properly. The phases dedicated to cutting, sampling, cleaning, waste management, samples preparation, and shipping were performed by PSI teams: each of these phases constitutes a huge amount of work, which has allowed PIE of structural material irradiated in relevant conditions to be performed. Moreover, chemical analysis already performed on spallation and corrosion products in the LBE are very relevant for further applications of LBE as a spallation media and, more generally, as a coolant. The already performed steps—conceptual and engineering design, manufacturing and assembly, safety and reliability assessment, thermo-hydraulic off-beam tests, irradiation, decommissioning, waste management, and post-irradiation examinations—bring to the ADS community unique and relevant design and operational feedback, which will be a decisive contribution to the development of accelerator-driven systems options for the transmutation of minor actinides.

Acknowledgments The authors warmly thank all contributors to the MEGAPIE project and more particularly the PSI team for its constant dedication and huge efforts during the decommissioning and sampling phase.

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Target Design for a Thorium Accelerator-Driven Molten Salt Reactor (AD-MSR)

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Abstract

Th–FLiBe fluid target design is proposed for a 1 GeV, 1 mA proton accelerator-driven molten salt reactor (AD-MSR). Simulations are carried out by FLUKA. The core of the system is an assembly with EURISOL target geometry. Results are presented for the geometrical distribution of neutron fluencies, the energy produced and fission density distribution, as well as neutron spectra in fuel, graphite moderator, reflector and concrete shielding. It is forecast to produce 10 MW_{th} power, neutron fluence of 10^{22} n/cm^2/GeV/mA produced by the spallation source and induced fission in FLiBe fuel.

Introduction

This work is based on a nuclear power development philosophy put forth by Kazuo Furukawa and collaborators in Japan [1], a philosophy that assumes use of the thorium fuel cycle and that relies on the properties of two basic nuclear processes. The first is the fact that nuclear fission is an "energy-rich, neutron-poor process". Indeed, fissions of uranium or plutonium produce energy of about 200 MeV per fission, but only of the order of 2–3 neutrons per fission. The second property on which this philosophy is based, is the fact that the spallation reaction is a "neutron-rich, energy-poor reaction": an energetic proton impinging on a target containing heavy, high atomic number atoms (such as Hg, Pb, or Bi) produces by a spallation reaction, 10–40 neutrons, depending mainly on the beam energy. This is about an order of magnitude more neutrons than fission of fissile material (^{235}U, ^{239}Pu). Consideration of the previous facts then leads to two principles that thorium nuclear power development should be based on. The first principle is that reactors, being efficient POWER producing devices, should be simple, reliable, and commercially attractive. Ideally, reactors should be capable of consuming fissile fuel for 30–40 or more years of continuous energy production without refueling. However, current reactors are not designed for breeding fissile materials. The second principle is that the fissile material should be bred from fertile material using neutrons produced by a specialized spallation reaction device: the thorium-based accelerator-driven molten salt reactor (Th-AD-MSR). We suggest a roadmap for development of nuclear energy generation based on the thorium fuel cycle and give a simple model in which we use the computational and predictive power of FLUKA [2, 3] and the experience of the EURISOL neutron source design development [4, 5] to predict the operational characteristics of an AD-MSR.

The MSR Roadmap

The five-stage "Roadmap" for development of thorium fuel cycle-based nuclear power is based on the Thorium Molten Salt Nuclear Energy Synergetics (THORIMS-NES) [6, 7] Japanese proposal.

1st Stage

The first stage is the construction of a Mini-FUJI reactor; a simplified replica of the 1969 Molten Salt Reactor Experiment

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(MSRE), which was built and operated at Oak Ridge National Laboratory (ORNL) in the period 1965–1969. The purpose of this stage is to recover the basic technology and know-how of molten salt reactors existing at ORNL 47 years ago. Although this experiment and following design exercise of the molten salt breeder reactor (MSBR), carried out at ORNL, were fully documented and are currently available [8], the personal experience and know-how has been lost. The timetable for this stage of development, is set at 5 years and is based on technical considerations at a location where all the resources of a first world scientific community are available and there are no financial limitations. It also does not consider development hindrances such as license delays or social or political factors that may prevent the technical progress of the project. This development timetable is surprisingly short due to the nature of the available information on the MSRE: the entire file of the ORNL experience on the development status of the molten salt reactor, with detailed chronological progress reports and engineering details of the work, is freely available on the Web ($\sim$3.5 GB) [6].

2nd Stage

The second stage is to build a denatured MSR (DMSR) or FUJI MSR [9]. This would be a 300 MW_{th} power demonstration reactor, which would be a scaled-up version of the device developed in the first stage. It would incorporate the engineering experience gained and supply chain built in the first stage. A DMSR deviates somewhat from the thorium fuel cycle, but it has the advantage of using fissile material that is "conventionally" accepted (20 % enriched uranium): an arbitrary, imposed by political restrictions derived from nuclear weapons proliferation considerations. This demonstration power reactor would incorporate thorium in addition to the required amount of uranium to be a near-breeder and could be operational in 10 years from the start of the program.

3rd Stage

The third stage consists of the worldwide deployment of thorium MSRs. The fissile fuel supply for these reactors is to be procured by nuclear weapons dismantling/spent fuel reprocessing until the end of the currently available fissile materials: ^{233}U, ^{235}U, and ^{239}Pu. This timescale is estimated at 30–35 years from the start.

4th Stage

The fourth stage consists of the introduction of the accelerator molten salt breeder (AMSB) into operation. Its primary function would be to produce ^{233}U fissile fuel for a future worldwide fleet of thorium MSRs. The timescale is estimated at 35+ years from start of the program. It is in this fourth stage that our modeling presented below finds its applicability.

5th Stage

The fifth stage consists of the establishment of regional centers (several according to demand) [10] where different sensitive operations will take place to support the future thorium nuclear era: light water reactor (LWR) dry spent fuel reprocessing, chemical processing plants, and plurifuctional AMSBs would be integrated mainly to burn long-lived actinides (TRUs) from LWR spent fuel, to produce fissile ^{233}U for the thorium-fueled reactors and to prepare Th–FLiBe fuel. Timescale estimates put stage five at 35+ years from the start, extending to the year 2100 onward.

The Accelerator-Driven Molten Salt Reactor Concept

It is worthwhile first to give a general scheme of the nuclear power plant concept that has been on the project desk for decades. The system is often referred as the Rubbiatron or the energy multiplier device CERN/AT/93-47 preprint [11]. The concept of a molten salt target was put forth by Japanese groups as a solution to the materials degradation problems anticipated with a solid-target accelerator-driven system as described in reference [1]. These devices operate as subcritical assemblies and the chain reaction is sustained by an in-core spallation-type neutron source. Early results on spallation reaction target design have been considered [12]. They conclude that the breeding process taking place in the AD-MSR scheme conveniently operates close to the critical value ($k \approx 0.95$).

Moving from the left to right in the schematic view presented in Fig. 1, we observe an ion source that produces a proton beam, which enters with 100 keV kinetic energy into the so-called ¨driver¨ accelerator and that exits as a 1 GeV high-energy H^+ beam to impact on the neutron source in the reactor core.

This is a target station where neutrons are generated and the thorium fuel undergoes irradiation. The target, by design, is located in a large tank lined with graphite in which the fuel at high temperature circulates in a closed loop from the core to the heat exchanger. The target station hosts the neutron source and the molten salt, which contains the fuel. In this process, ^{235}U is employed as an external fissile fuel to initiate and contribute to maintaining the chain reaction. The molten salt contains thorium as ThF_4, which acts as a fertile

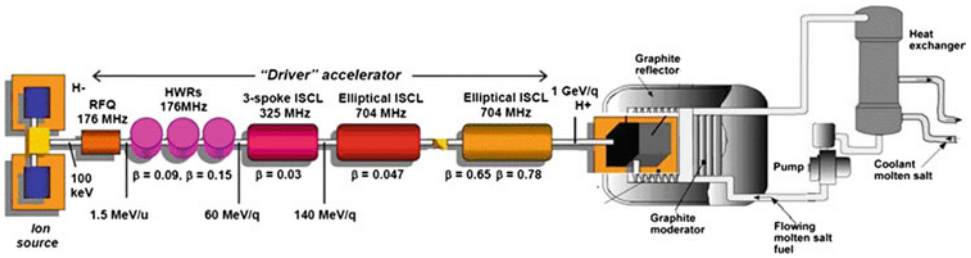

Fig. 1 Diagram of a near-critical MSR in which the accelerator supplies a beam of high-energy protons to the spallation target inside the core to maintain the chain reaction

material for the production of fissile ^{233}U. The ^{238}U contained in the fuel also contributes as a fertile material as fissile ^{239}Pu is produced.

Characteristics of the Neutron Spallation Source

Figure 2 shows the AD-MSR core with the central spallation target assembly. The spallation target area is a modification of the EURISOL neutron source, which has liquid mercury as the target material. The model of the neutron source was modified by substituting liquid mercury with the molten fuel salt, FLiBe. The surrounding uranium target of the EURISOL source was also substituted with a graphite moderator and liquid fuel. The geometry is cylindrical with the proton beam along the axis impinging near the axial center on an inner area filled with FLiBe fuel. This area is surrounded by a cylindrical inner graphite moderator. The majority of the fuel is contained in an outer FLliBe zone, which in turn is surrounded by a graphite reflector. This geometry, with an outer concrete container, allows the building of a simple model capable of supplying basic operating information for the device, without entering into undue detail for a preliminary study. The composition of the molten salt used in this study is shown in Table 1. Detailed properties have been reported by Cantor [13]. The fissile material in the molten salt is contained in UF_4 of natural composition, which is dissolved in LiF, BeF_2, and ThF_4.

FLUKA Model

Table 2 shows some of the parameters used as inputs to the program. The origin of the coordinates is in the mid-point of the core. The *z*-axis is defined along the proton beam direction and the *y*-axis along the vertical direction. The table contains some of the model numerical values.

FLUKA Results

Neutron Fluence, Power, and Fission Density

We present a graphical summary of the results of the FLUKA model given for a proton beam of 1 GeV and a current of 1 mA (neutrons/cm^2/mA). Figure 3 shows the neutron fluence distribution and the distribution of the power density in the core of the reactor. Energy produced by fission in the molten salt is shown as heat, which is conducted into the surrounded graphite reflector and concrete. The total power produced is estimated at 10 MW. Figure 4 shows the fission

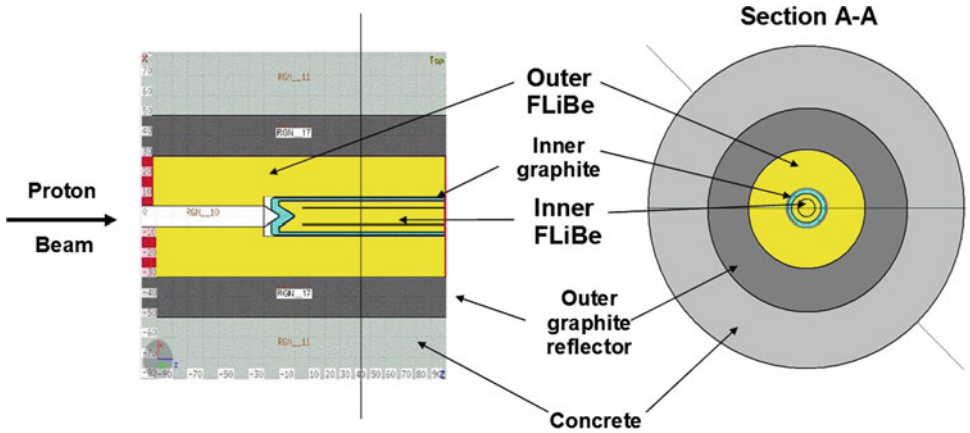

Fig. 2 Geometry of the model core of an AD-MSR. *Left* The longitudinal section, showing the area where the proton beam hits the inner FLiBe fuel. *Right* Cross sectional view

Table 1 Composition of the FLiBe molten salt fuel used for the modeling in this work

Compound	Amount (mol%)
LiF	70
BeF_2	20
ThF_4	8
U (^{238}U and ^{235}U)	2.0

Table 2 Beam parameters for simulations[a]

Beam particle	Protons
Energy and current	1 GeV, 1 mA
FWHM of a Gaussian profile in *x*-direction	−3.53223
FWHM of a Gaussian profile in *y*-direction	−3.53223
Beam profile	Gaussian
Total FLiBe volume	5.42×10^5 cm^3
Total energy in FLiBe	6.77×10^3 kW/mA
Total energy density in FLiBe	5.76×10^{-2} kW/cm^3/mA

[a]FWHM = full width at half maximum

distribution in the fuel as a result of spallation and fission neutrons. These figures (Figs. 3 and 4) indicate a certain asymmetry of the neutron fluence, power density, and fission density in the forward direction of the proton beam.

Nuclear Reactions

The FLUKA model yields the results of nuclear reactions that take place in the reactor by drawing an isotope table diagram of nucleon number (A) as a function of proton number (Z) for the core zones where fission take place, that is, the inner and outer FLiBe zones. Isotopes are produced by both fission and capture reactions in these zones; hence, light and heavy elements other than fission products are expected to be contained in the fuel.

Neutron Energy Distribution

In this section, we show the neutron energy distribution in the form of spectra for the neutrons in the various zones of the AD-MSR as predicted by the FLUKA model. Figure 5 shows the results for the zones containing molten salt fuel. Figure 6 shows the neutron spectra for the two graphite-containing zones: the inner graphite moderator and outer graphite reflector. Finally, Fig. 7 shows the neutron spectra at the outer concrete wall.

Discussion and Conclusions

The model presented is limited to some neutronic calculations. It does not consider heat transfer or fluid flow. In principle, the fluid would circulate at a moderate speed of 0.5 m/s, which is sufficient for heat removal and transfer from the core region to a heat-transfer region with part of the flow diverted to a chemical processing plant with several functions, including removal of the fissile material produced (^{233}U and ^{239}Pu), removal of radioactive gaseous and solid fission products, and reconditioning of the molten salt by addition of fertile thorium and light actinides for burning and reduction of the overall lifetime of nuclear

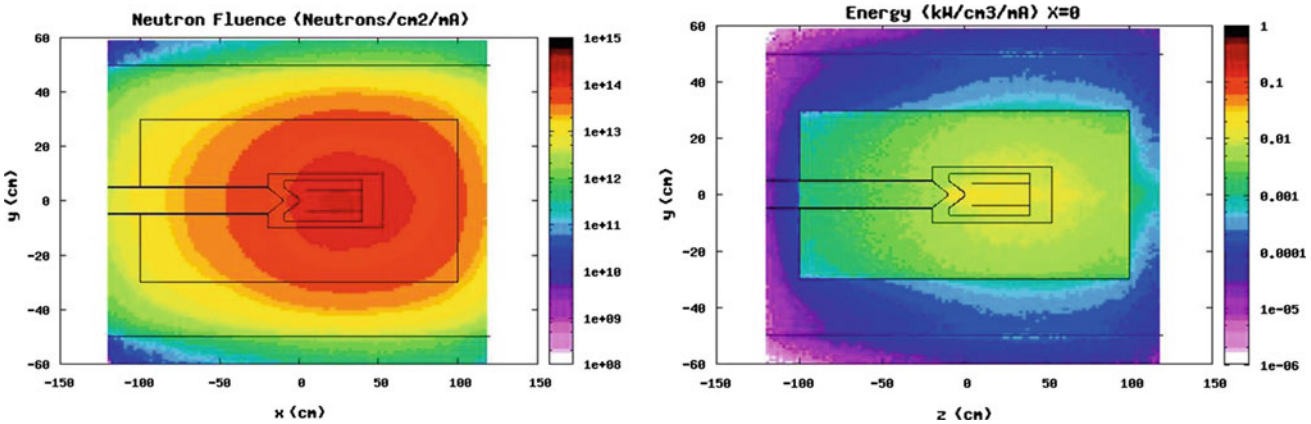

Fig. 3 *Left* Neutron fluence distribution in the core (neutrons/cm^2/mA). Results are shown for a proton beam of 1 GeV and a current of 1 mA. *Right* Calculated power density (kW/cm^3/mA) distribution in the core of the reactor

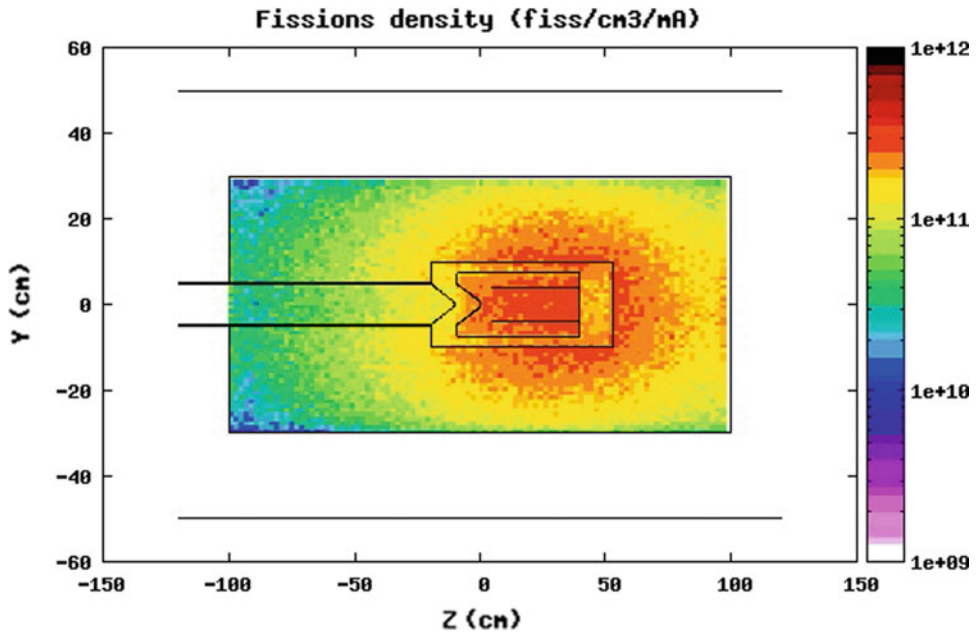

Fig. 4 Fission density distribution in the core of the AD-MSR (fissions/cm^3/mA)

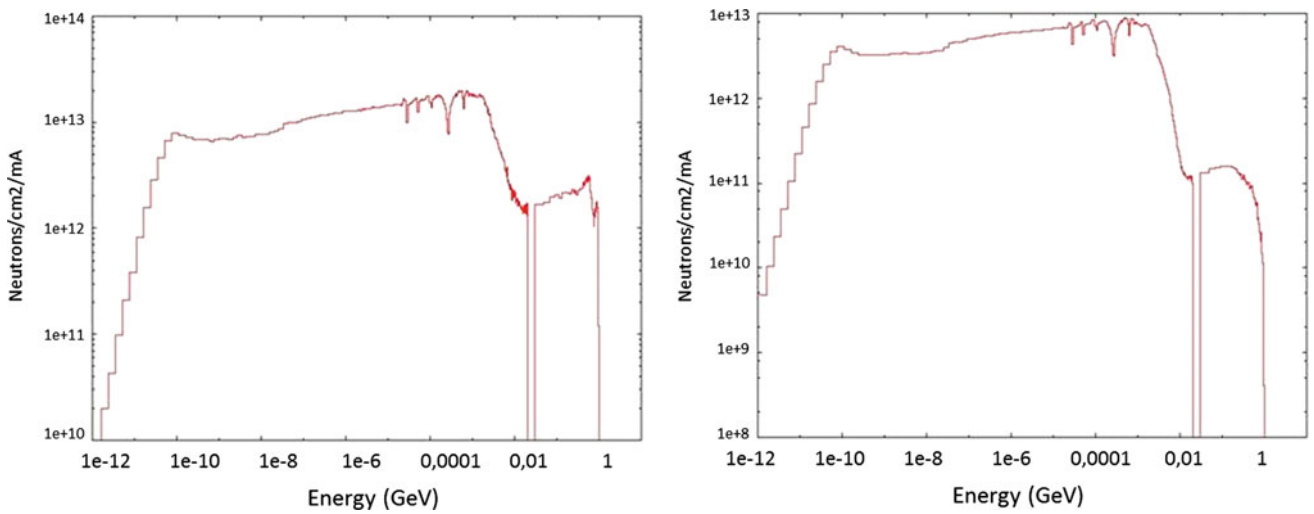

Fig. 5 Neutron fluence spectra for the molten salt, FLiBe, (neutrons/mA) for the inner zone (*left*) and for the outer zone (*right*)

waste. Today's accelerators are demonstrably capable, of achieving proton beams of around 1 MW (PSI, SNS), suggesting the feasibility with today's technology to build a device as is modeled here. In view of the results, we come to the conclusion that it is indeed possible to use the EURISOL neutron source for the modeling of a neutron source for an AD-MSR. The preliminary results indicate that further refinement in the size scaling, fissile concentration, and a more realistic geometry can provide a model of a reactor that is a power device, a fuel breeder, and an actinide waste burner.

These results support the CERN "Opinion of the Scientific and Technical Committee" (STC) on a nuclear energy amplifier.

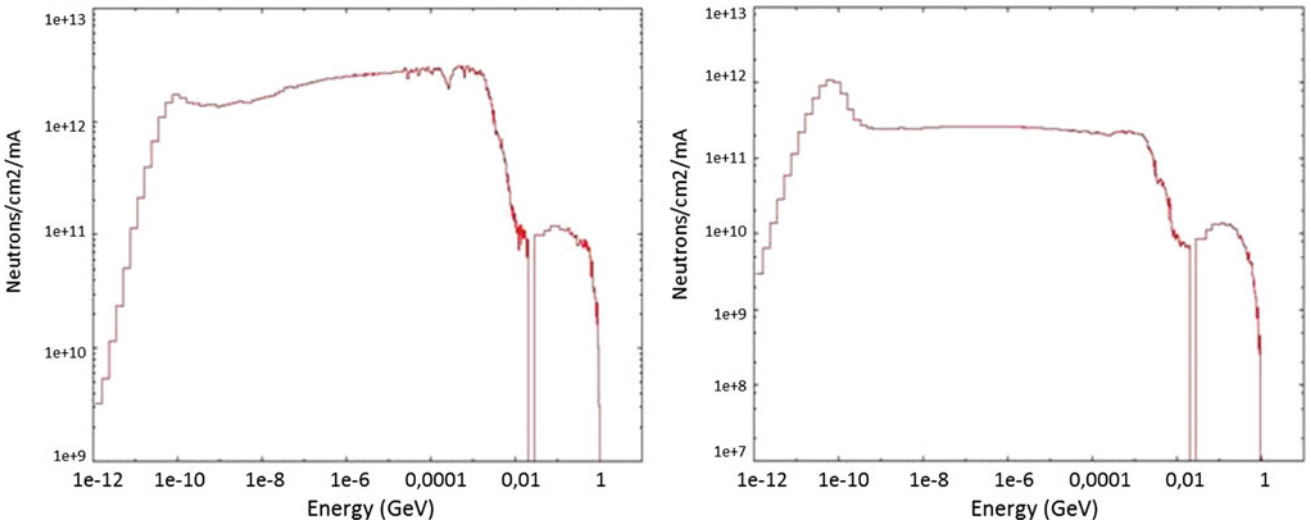

Fig. 6 Neutron fluence spectra for the graphite (neutrons/cm^2/mA) for the internal graphite moderator (*left*) and the outer graphite reflector (*right*)

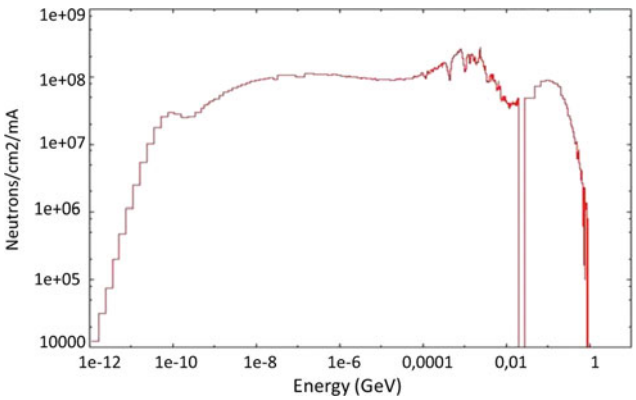

Fig. 7 Neutron fluence spectra (neutrons/cm^2/mA) at the concrete wall

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Virginia Nuclear Energy Frontier Research Center

Ganapati Myneni

Abstract

Virginia General Assembly recently established the Virginia Nuclear Energy Consortium Authority (VNECA) effective July 1, 2013. A non-profit Virginia Nuclear Energy Consortium (VNEC) will be created by VNECA in the coming months. One of the major planned goals for the VNEC is to foster strategic partnerships between Virginia educational institutions, federal laboratories, nuclear industry and non-profit organizations to rapidly expand the nuclear energy business sector in the commonwealth. Several of the VNECA stakeholder institutions have already formed a Virginia ADS Consortium with a vision to create a state-of-the-art Science and Technology Center (STC) for the application of high-power accelerators for the advancement of innovative multidisciplinary science and to educate future generation of scientists and engineers. In this presentation we will explore the objectives of the Virginia ADS Consortium and its plans to implement them.

A group of institutions consisting of several Virginia universities, corporations, national labs and a not-for-profit organization have established the Virginia Accelerator-Driven Systems (ADS) Consortium to pursue R&D in nuclear waste transmutation, development of new nuclear power cycles, new approaches of isotope production for medical, life, and physical sciences applications; and to train future generations of scientists and engineers. The membership of the Consortium includes Virginia Commonwealth University (VCU), University of Virginia (UVa), Virginia Tech (VT), Old Dominion University (ODU), Longwood University, Muons's Inc., Casting Analysis Corporation, Dominion, South Dakota School of Mines and Technology, Oak Ridge National Laboratory (ORNL), non-profit International Symposium On Hydrogen In Matter (ISOHIM), and Jefferson Lab. During the legislative session that ended February 24, 2013, the Virginia General Assembly established the Virginia Nuclear Energy Authority (VNEA) and Virginia Nuclear Energy Consortium (VNEC), and the Virginia ADS Consortium institutions are involved in both of them.

The long-term strategic vision of VA ADS Consortium is the establishment of Virginia ADS R&D Center in order to train future generations of scientists and engineers in the fields of materials science, nuclear and biomedical engineering, molecular imaging, nanotechnology and nanomedicine. As a first step towards the goal, we are jointly planning to establish a nuclear medical isotope development facility (NMIDF) based on 50 MeV continuous-wave (CW) electron linear accelerator (linac) technology developed at Jefferson Lab, and photonuclear isotope production techniques developed by consortia members. It is proposed that the required building with the necessary infrastructure will be provided by several not-for-profit centers either at the tech center near Jefferson Lab or on a site provided by York County. The rest of the NMIDF is to be funded by VA State and Corporations on an equal partnership basis. UVa, East Virginia Medical School (EVMS), Duke University, and VCU's Center for Molecular Imaging (SOM), and the School of Engineering have expressed strong interest in the initial development of new approaches to provide a "step change" in the production of copper radioisotopes, in particular ^{67}Cu for multi-modality biomedical imaging and

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multi-therapeutic applications. The strategy is extendable and amenable to the production of other radioisotopes spanning a variety of applications for scientific, medical, and industrial applications.

The NMIDF will not only help us in the development of critically needed medical isotopes but will also provide huge opportunities in materials research, including theranostic (therapeutic and diagnostic) nanoparticles and complementary research in novel approaches in non-invasive image-guided technologies in surgical, radiation, and drug therapeutics. In addition, the NMIDF will be a springboard for launching the VA ADS R&D Center. The ADS R&D Center will lay the foundation for the transmutation of nuclear waste, help in the development of new nuclear energy fuel cycles, and train future generations of scientists and engineers.

Thorium-Loaded Accelerator-Driven System Experiments at the Kyoto University Research Reactor Institute

Cheol Ho Pyeon

Abstract

Experimental studies on the thorium-loaded accelerator-driven system (ADS) have been conducted at the Kyoto University Critical Assembly. Mockup experiments are carried out in both the critical and subcritical states to investigate the profile of the neutron flux for thorium capture and fission reactions. Thorium plate irradiation experiments for thorium capture and fission reactions demonstrate fission reactions in the critical state, and the calculated versus experimental C/E values of the reaction rates show accuracy within a relative difference of about 30 %. In the ADS experiments with an external neutron source (14 MeV neutrons and 100 MeV protons), subcritical experiments were carried out in the thorium-loaded cores to investigate the profile of neutron flux for capture reactions by the measurement of ^{115}In(n, γ)^{116m}In reactions. The results reveal the difference between reaction rate distributions attributed to varying not only the neutron spectrum of the core but also the external neutron source at the target location. A comparison between the measured and calculated reaction rate distributions reflects the accuracy of reaction rate analyses for the thorium-loaded ADS experiments with an external neutron source. Additionally, kinetic experiments were carried out to deduce the delayed neutron decay constants and subcriticality by the pulsed-neutron method.

Introduction

The concept of an energy amplifier with the accelerator-driven system (ADS) proposed in the 1990s [1–3] was comprised of thorium (^{232}Th) fuel and a high-gain and -power accelerator. It was based on an essential feature, capture reactions, of ^{232}Th breeding to uranium-233 (^{233}U) as shown below, whereas ^{232}Th demonstrated another important feature of a threshold reaction of approximately 2 MeV neutrons based on the fission ^{232}Th(n, f) process:

$$^{232}\mathrm{Th}(\mathrm{n},\gamma)\,^{233}\mathrm{Th}\xrightarrow[22.3\mathrm{m}]{\beta^-}\,^{233}\mathrm{Pa}\xrightarrow[26.967\mathrm{d}]{\beta^-}\,^{233}\mathrm{U}$$

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At the Kyoto University Critical Assembly (KUCA), spallation neutrons generated by high-energy proton beams were successfully injected into the thorium-loaded system, in addition to the first injection [4] of high-energy protons. By combining the fixed-field alternating gradient (FFAG) accelerator [5] with the thorium-loaded system at KUCA, a series of the ADS experiments [6, 7] could be carried out under conditions whereby the spallation neutrons were generated at a tungsten target by 100 MeV protons at an intensity of 30 pA. Regarding the two major reactions, capture and fission, of ^{232}Th, special attention was paid to the thorium fission reactions in the thorium-loaded ADS experiments.

Among the series of ADS experiments at KUCA, the mockup experiments [8] of thorium-loaded ADS carried out by varying the neutron spectrum and the external neutron source were aimed at investigating the influence of different neutron profiles on thorium capture reactions and the prompt and delayed neutron behavior in the subcritical system.

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The results provided the important effects of the neutron spectrum and the external neutron source on both the static and kinetic parameters: the effect of the neutron spectrum was investigated by varying the moderator material in the fuel region, and that of external neutron source by separately injecting 14 MeV neutrons and 100 MeV protons into the thorium-loaded core, varying the moderator. Before the subcritical experiments, a thorium plate irradiation experiment was carried out in the KUCA core to analyze the thorium capture and fission reactions in the critical system as a reference for the subcritical system, although the feasibility of ^{232}Th capture and ^{233}U fission reactions could only be examined in the subcritical state. The objectives of this study were to investigate the neutronic characteristics of the thorium-loaded ADS with an external neutron source through static and kinetic experiments, and to evaluate the accuracy of the Monte Carlo analyses through calculations with the MCNPX [9] code with ENDF/B-VII.0 [10], JENDL/HE-2007 [11], and JENDL/D-99 [12] libraries. The thorium fission reaction, thorium plate irradiation, and the ADS static and kinetic experiments at KUCA are presented here, along with the results and discussion of the experiments and calculations and the conclusion of the study.

Experimental Settings

Thorium Fission Reaction

The thorium-loaded systems (thorium–graphite and thorium; Fig. 1) in the ADS experiments were composed of a 2″ × 2″ square and 1/8″-thick fuel plate, and a 2″ × 2″ square and 1/8″- and 1/2″-thick fuel and graphite plates, respectively. By adding graphite to the thorium system, the effects of neutron leakage by a large size core and neutron spectrum softening by the graphite could be found in the thorium–graphite system. Thus, the change in neutron multiplication caused by the neutron leakage and spectrum softening was expected to be revealed in a comparison of the two thorium systems.

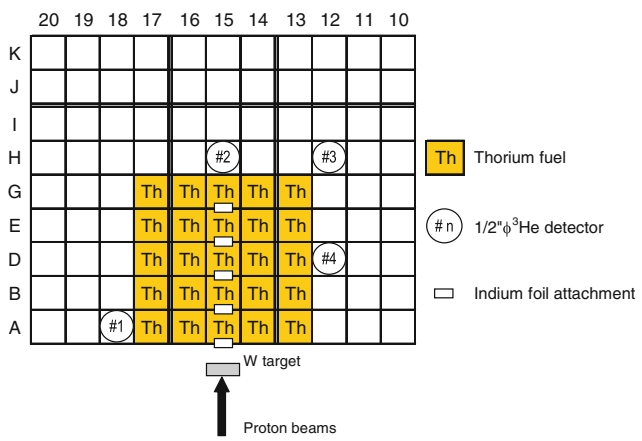

Fig. 1 Core configuration in thorium fission reaction experiments

KUCA comprises two solid polyethylene-moderated thermal cores, designated A and B, and one water-moderated thermal core, designated C. The A-core is mainly used for experiments of ADS basic research. The three cores are operated at a low mW power in the normal operating state; the maximum power is 100 W. The constitution and the configuration of the cores can be altered easily, and the coupling with the conventional Cockcroft–Walton-type accelerator and with the FFAG accelerator has allowed experiments to be conducted separately with the use of 14 MeV neutrons from deuteron–tritium fusion reactions and 100 MeV protons with a heavy metal target, respectively.

Thorium Plate Irradiation

The thorium metal plate (2″ × 2″ × 1/8″) irradiation experiment was carried out in the A-core, which is equipped with polyethylene reflector rods and three different fuel assemblies: normal, partial, and special fuel (highly enriched uranium and thorium, HEU–Th; Fig. 2) assemblies. The thorium plate was set in the fuel assembly at the center position to obtain irradiation information on the thorium capture and fission reactions in the critical state.

Thorium-Loaded ADS Benchmarks

In the ADS with 14 MeV neutrons and 100 MeV protons, the fuel rod was composed of a thorium metal plate and a polyethylene (PE), graphite (Gr) or beryllium (Be) moderator arrangement. Other components were selected from HEU and natural uranium (NU; 2″ × 2″ × 1/8″) plates. The cores were comprised of Th–PE (Fig. 3), Th–Gr, Th–Be, Th–HEU–PE, or NU–PE, and 14 MeV neutrons and spallation neutrons were generated outside the core after the injection of deuteron and proton beams onto the tritium and tungsten targets, respectively. The thorium-loaded ADS experiments were conducted to investigate, in particular, the relative influence of different neutron profiles on capture reactions of ^{232}Th and ^{238}U: the reaction of ^{238}U was taken as reference data for evaluating the validity of ^{232}Th capture cross sections.

The main parameters of the 14 MeV neutrons were as follows: 160 keV high voltage, 0.6 mA intensity, 100 Hz pulsed frequency, 10 μs pulsed width, and 25 mm diameter spot size. The level of the neutron yield was around 1.0×10^6 s^{-1}. The proton beam parameters were: 100 MeV

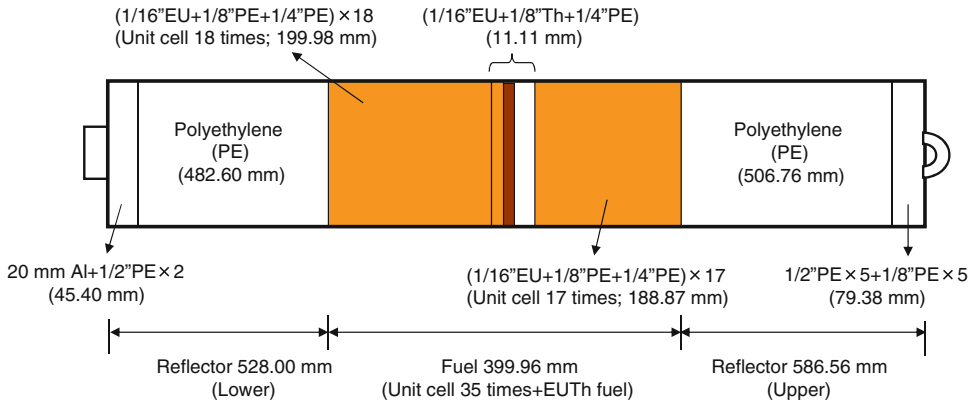

Fig. 2 Fuel assembly employed in the thorium plate irradiation experiments

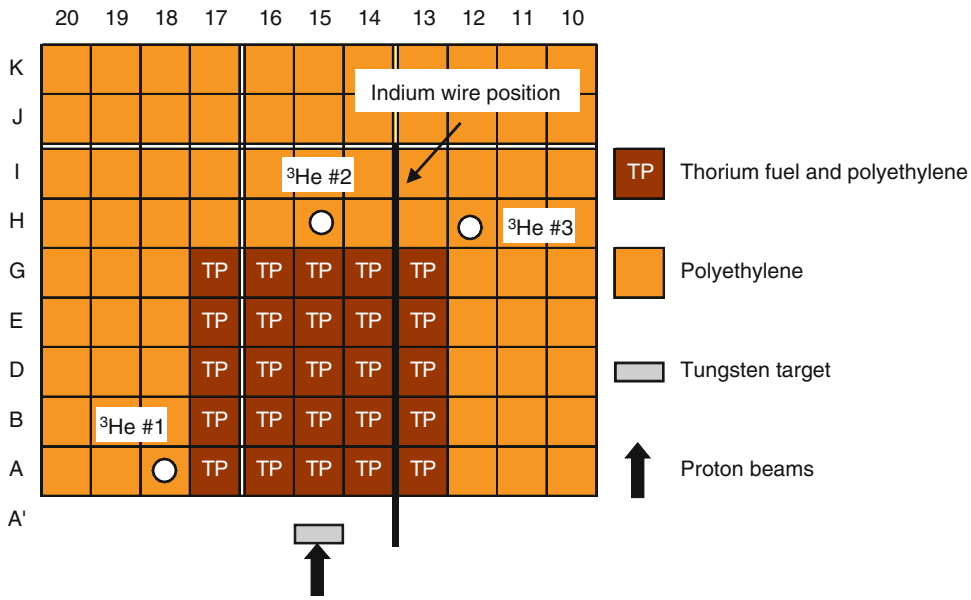

Fig. 3 Top view of thorium-loaded ADS core with 100 MeV protons

energy, 0.3 nA intensity, 20 Hz pulsed frequency, 100 ns pulsed width, and 40 mm diameter spot size at the tungsten target (50 mm diameter and 9 mm thick). The level of the neutron yield generated at the target was around 1.0×10^7 s^{-1} from injection of 100 MeV protons onto the tungsten target.

The prompt and delayed neutron behaviors were monitored by placing three ^{3}He detectors (20 mm diameter and 300 mm long) at three locations. Throughout the time evolution of the prompt and delayed neutrons, the prompt neutron decay constant was deduced by the least-square fitting method to an exponential function over the optimal duration. Subcriticality was deduced by the extrapolated area ratio method [13] on the basis of the prompt and delayed neutron behaviors. For 100 MeV protons, neutron detectors (^{3}He detectors; #1, #2, and #3) were set at three locations.

Results and Discussion

Thorium Fission Reactions

The high-energy neutron flux distribution was estimated by measurement of the ^{115}In(n, n′)^{115m}In (threshold energy of 0.32 MeV neutrons) reaction rate distribution with the use of the foil activation method of indium foils ($50 \times 50 \times 1$ mm^3). The foils were set at an axial center position (beam injection position) of five fuel rods (position 15, A, B, D, E, G in Fig. 1) to obtain information on high-energy neutrons in the system. All foils were normalized by the ^{115}In(n, n′)^{115m}In reactions excited in another indium foil ($20 \times 20 \times 2$ mm^3) setting at the tungsten target position to obtain the source neutron generation. The effect of neutron spectrum softening by graphite was comparable to the measured reaction rates

Table 1 Comparison between measured reaction rates in the Th and Th–Gr systems

Position	Reaction rate (cm^{-3} s^{-1})		Difference (%)
	Th system	Th–Gr system	
(15, A)	$(5.32 \pm 0.07) \times 10^{-2}$	$(6.06 \pm 0.13) \times 10^{-2}$	13.9
(15, B)	$(1.92 \pm 0.03) \times 10^{-2}$	$(2.16 \pm 0.06) \times 10^{-2}$	12.5
(15, D)	$(7.39 \pm 0.15) \times 10^{-3}$	$(8.51 \pm 0.32) \times 10^{-3}$	15.2
(15, E)	$(3.15 \pm 0.08) \times 10^{-3}$	$(4.38 \pm 0.21) \times 10^{-3}$	39.0
(15, G)	$(1.39 \pm 0.04) \times 10^{-3}$	$(2.17 \pm 0.13) \times 10^{-3}$	56.1

Table 2 Measured thorium capture and fission reaction rates, and C/E values

Reaction	Nuclide	Measured reaction rate (cm^{-3} s^{-1})	C/E
Capture	^{233}Pa	$(5.82 \pm 0.01) \times 10^{6}$	2.18 ± 0.04
Fission	^{91}Sr	$(1.42 \pm 0.04) \times 10^{5}$	1.39 ± 0.05
	^{92}Sr	$(1.46 \pm 0.05) \times 10^{5}$	1.36 ± 0.06
	^{97}Zr	$(1.47 \pm 0.02) \times 10^{5}$	1.35 ± 0.03
	^{135}I	$(1.50 \pm 0.04) \times 10^{5}$	1.32 ± 0.04
	^{142}La	$(1.56 \pm 0.05) \times 10^{5}$	1.27 ± 0.05

(Table 1) of the thorium (Th) and thorium–graphite (Th–Gr) systems. From the relative difference between measured reaction rates in the two systems, the effect of neutron spectrum softening was clearly revealed to be over 50 % at the maximum in the core, according to stepping away from the neutron source.

Thorium Plate Irradiation

The thorium plate irradiation experiment was carried out in the A-core in the critical state to acquire information on the thorium capture and fission reaction rates. The experimental results from the core are shown in Table 2, and the numerical analyses for the experiments were conducted with the use of the Monte Carlo code MCNPX and the nuclear data library ENDF/B-VII.0. Although the C/E values of the measured and the calculated reaction rates revealed a discrepancy in the capture reactions, the reaction rates of the fission reactions were found to be in agreement at around 30 %. Both measured and calculated thorium capture and fission reaction rates were normalized by the ^{197}Au(n, γ)^{198}Au reaction rates of gold foil.

Thorium-Loaded ADS Benchmarks

Static Experiments

The absolute values of the measured reaction rates (Fig. 4) revealed differently the variation of reaction rates attributed to varying the neutron spectrum (Fig. 5; MCNPX calculations) at location (15, W) in Fig. 3 when spallation neutrons

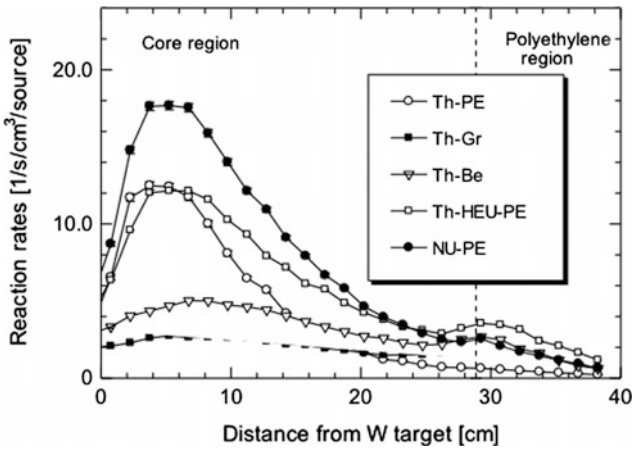

Fig. 4 Measured reaction rates of indium wire obtained from the thorium-loaded ADS experiments with 100 MeV protons

generated by 100 MeV protons were injected into the core. The moderating effect of the high-energy neutrons in some cores (Th–PE, Th–HEU–PE, and NU–PE) was observed around the boundary between the core and polyethylene regions. The ^{115}In(n, γ)^{116m}In reaction rates in the NU–PE core were higher than in other cores, demonstrating that the reaction rates of ^{238}U in the NU–PE core are larger than those of ^{232}Th in the thorium cores with the use of 100 MeV protons. Additionally, the effect of the neutron spectrum on the reaction rates was observed with 100 MeV protons by comparing the measured results of reaction rates shown in Fig. 4. Thus, the expected physical effect was indeed observed due to the neutron spectrum change obtained by varying the moderator materials in the fuel assembly.

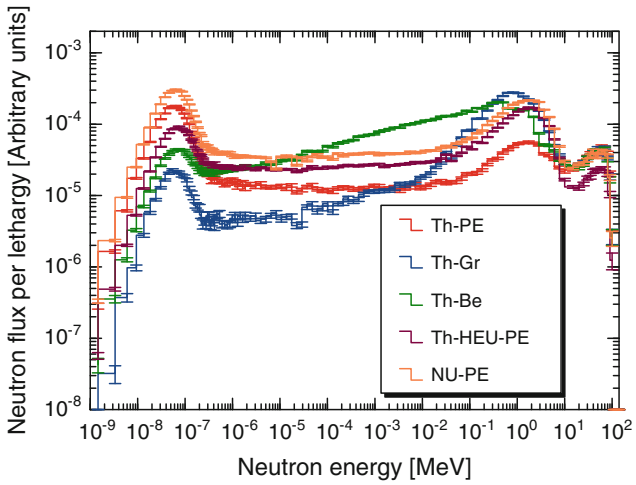

Fig. 5 Calculated neutron spectra obtained from the thorium-loaded ADS experiments with 100 MeV protons

The numerical calculations of reaction rate distributions were performed with the use of the MCNPX code coupled with ENDF/B-VII.0 (transport) and JENDL/D-99 (reaction rates). The calculated ^{115}In(n, γ)^{116m}In reaction rates approximately reconstructed the experimental ones. Also, comparison between the experiments and the calculations demonstrated good agreement among the Th–Gr, Th–Be, Th–HEU–PE, and NU–PE cores.

For the injection of 100 MeV protons into the tungsten target, as shown in Fig. 5, the neutron spectrum (MCNPX calculations) was varied in the core at location (15, B) in Fig. 4. The bombardment of 100 MeV protons at the target and neutron generation at high energy were modeled with the combined use of MCNPX and JENDL/HE-2007 or ENDF/B-VII.0, respectively.

Kinetic Experiments

The time evolution of prompt and delayed neutron behavior was examined through the injection of an external neutron source. In the Th–HEU–PE core, the prompt neutron decay constant (determined by the least-square fitting method; Table 3) at ^{3}He detector #1 was different from those at the other detectors, regardless of the type of external neutron source and the position of neutron detection. This value is considered to be overestimated particularly at detector #1, which was located near the external neutron source. The two different neutron sources provided different delayed neutron backgrounds. Subcriticality in dollar units was deduced by the extrapolated area ratio method with the use of prompt and delayed neutron components, and experimentally evaluated according to the type of external neutron source and the location of neutron detection. As is well known, these results revealed subcriticality dependence on both the type of external neutron source and the location of neutron detection, although the value of subcriticality was theoretically unchanged, regardless of the external neutron source and the location of the detector. Consequently, the experimental results showed that the subcriticality in pcm units for 14 MeV neutrons was different from that for 100 MeV protons; remarkably, the discrepancy was also observed in the experiments and calculations.

Table 3 Measured results of prompt neutron decay constant (α [s^{-1}]) in the Th–HEU–PE core

Source	^{3}He #1	^{3}He #2	^{3}He #3
14 MeV neutrons	5735 ± 5	5155 ± 4	5161 ± 4
100 MeV protons	5788 ± 5	5338 ± 5	5229 ± 5

Conclusion

A thorium-loaded ADS study was conducted as observed by the reaction rates determined through static experiments. The thorium fission reactions were obtained from calculations of the reaction rates, although the intensity and the neutron yield by 100 MeV protons were low at 30 pA and 1×10^{7} s^{-1}.

The thorium plate irradiation experiment was conducted in the critical state. Thorium fission reactions were observed in the critical system, and the C/E value of the reaction rates demonstrated a relative difference of about 30 %, whereas that of the thorium capture reaction rate showed a discrepancy.

Mockup experiments of thorium-loaded ADS with 14 MeV neutrons and 100 MeV protons revealed a difference in reaction rate distributions, which was attributed to varying not only the neutron spectrum but also the external source. The comparison between thorium capture reactions with 14 MeV neutrons and 100 MeV protons showed the need for further investigations into the influence of external neutron sources on thorium capture reactions. Kinetic experiments were carried out to deduce the prompt neutron decay constants and subcriticality by the least-squares fitting method and the extrapolated area ratio method, respectively.

Acknowledgments This work was supported by the KUR Research Program for Scientific Basis of Nuclear Safety from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan. The authors are grateful to all the technical staff of KUCA for their assistance during the experiments.

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A Status and Prospect of Thorium-Based ADS in Korea

Jong-Seo Chai

Abstract

This paper describes the current status of research and development (R&D) of accelerator-driven systems (ADS) and nuclear transmutation techniques (NTT), including nuclear data, computational modeling (TNudy), partitioning and transmutation of long-lived fission products (LLFPs), accelerator-driven molten salt reactors, ADS cyclotron design, life cycle assessment of ADS, and applications (detectors, medical, etc.) at KAERI (Korea Atomic Energy Research Institute), SNU (Seoul National University), and SKKU (Sungkyunkwan University) in Korea.

Introduction

The Emerging Nuclear Technology Group was formed in 2009 as a multidisciplinary, international group with the aim of developing thorium-based accelerator-driven system (ADS) technology for future nuclear energy production. Figure 1 shows the current status of nuclear power plants in Korea. Nuclear energy accounts for 31.2 % of electricity produced in Korea. There are about 23 nuclear power plants operating and, even after the Fukushima accident, we are still constructing three new power plans and a few power plans are under license review [1].

Past ADS Development in Korea: ADS Research in KAERI

The Korea Atomic Energy Research Institute (KAERI) has been working on ADS since 1997. The KAERI ADS system is called HYPER (HYbrid Power Extraction Reactor). HYPER research started as a 10 year nuclear research program funded by the government. ADS research at KAERI consists of three stages. A basic concept of HYPER was established in the first stage (1997–2000) of the development. The basic technology related to HYPER was investigated in the second stage (2001–2003) while upgrading the design. The third stage of research started in March, 2004. The conceptual design of the HYPER core will be completed in the third stage (2004–2006). Investigation of key technologies will be continued in the third stage. The conceptual design of the HYPER core was almost finished in the second stage. The upgrade of the core design and transient study will be done in the third stage. Regarding experimental research, fuel and Pb–Bi studies were performed during the second stage. Uranium surrogate fuel was fabricated and tested. KAERI joined the MEGAPIE project in 2001 for Pb–Bi research. KAERI also installed a static Pb–Bi corrosion test device in 2003 and started experiments. KAERI will complete the construction of the Pb–Bi corrosion loop in 2004. The I-NERI program related to lead-alloy experiments will be launched in 2004. For the fuel/fission product (FP) target study, KAERI plans to perform fission product irradiation tests using KAERI's research reactor HANARO. The HYPER (HYbrid Power Extraction Reactor) PROJECT was designed to transmute transuranic elements (TRU) and some fission products, such as ^{129}I and ^{99}Tc. HYPER is a 1000 MW_{th} system and its k_{eff} is 0.98. Figure 2 shows a schematic configuration of the HYPER core with 186 ductless hexagonal fuel assemblies. As shown in Fig. 2, the fuel blanket is divided into three TRU enrichment zones to flatten the radial power distribution. In HYPER, a beam of 1 GeV protons is delivered to the central region of the core

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to generate the spallation neutrons. All specification of HYPER are listed in Table 1. No thorium was considered for use in HYPER [2, 3].

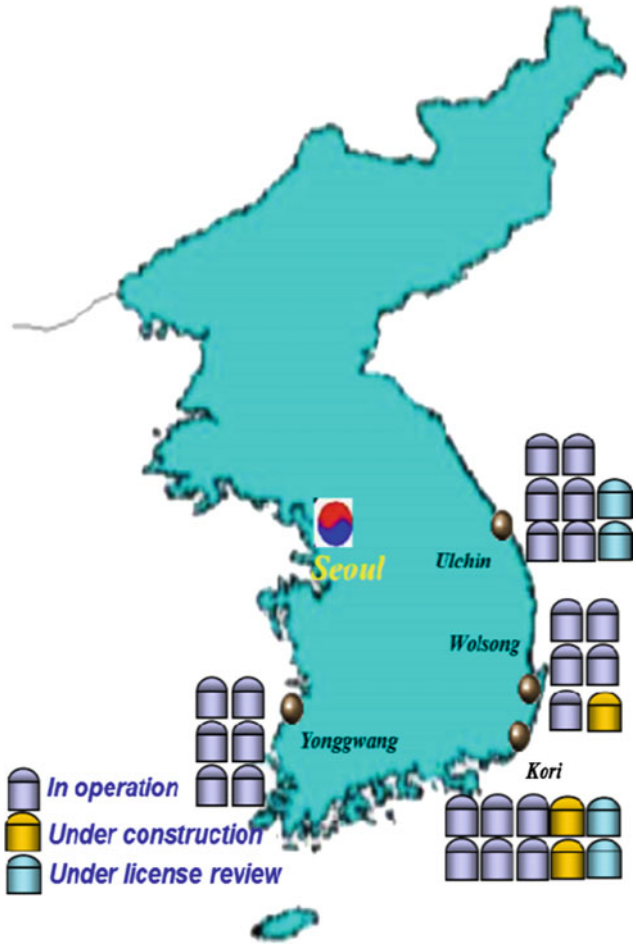

Fig. 1 Current status of nuclear power plants in Korea

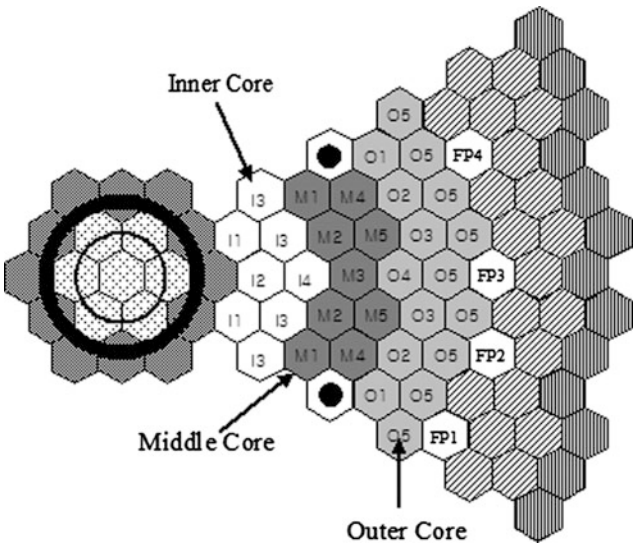

Fig. 2 Schematic diagram of the HYPER core

Table 1 Specifications of HYPER

Item	Value
Capacity	1000 MW_{th}
Keff	0.98
Proton beam	1 GeV, 10.6/16.4 mA (BOC/EOC)
Fuel type	TRU Zr-metal alloy
Coolant/target	Pb–Bi (not separated target)
Transmutation	TRU, FP(Tc-99, I-129)
TRU tran. rate	282 kg/year
FP tran. rate	27 kg/year (Tc-99), 7 kg/year (I-129)
Support ratio	3.2
Temperature	Inlet = 340, Outlet = 470, Max. = 542 °C

Present ADS Status in Korea: ADS Research in Universities

The Emerging Nuclear Technology Group was formed in 2009 as a multidisciplinary, international group with an aim of developing thorium-based accelerator-driven system (ADS) technology for future nuclear energy production. This paper describes the current status on research and development (R&D) of accelerator-driven system (ADS) and nuclear transmutation techniques (NTT), including nuclear data, computational modeling (TNudy), partitioning and transmutation of long-lived fission products (LLFPs), accelerator-driven molten salt reactors, ADS cyclotron design, life cycle assessment of ADS, and applications (detectors, medical, etc.) at Sungkyunkwan university.

TNudy (ROOT Nuclear Data Library)

The ENDF format for nuclear data was designed many years ago [4]. The ASCII format it uses is still practical for human inspection and portability, but it is not very effective for manipulating and displaying the data or for Monte Carlo applications. We propose a new prototype for a nuclear data manipulation package (TNudy) based on the ROOT system [5]. The object-oriented C++ framework of the ROOT system has been the de facto standard in high energy and nuclear physics experiments worldwide for the last ten years. Starting from the ENDF format, the data will be stored in a machine portable binary format. Root files are internally structured with direct access to their different sections and are automatically compressed upon writing and uncompressed upon reading, thereby minimizing the disk occupancy. Moreover, ROOT offers a complete library of visualization and mathematical routines that allow easy display and manipulation of the data (Figs. 3 and 4). The-ROOT framework includes the so-called Virtual Monte Carlo system, which allows us to run different major

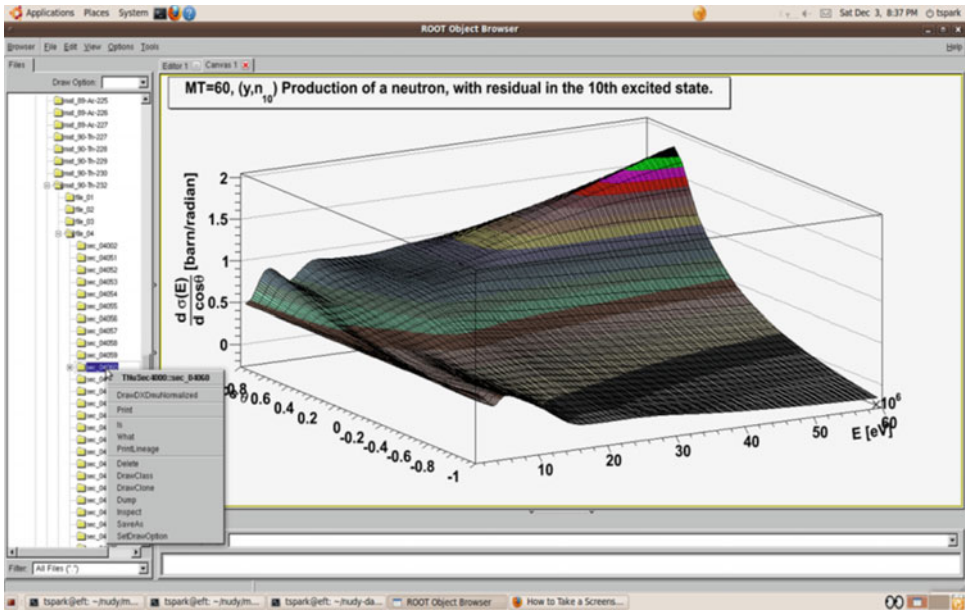

Fig. 3 ENDF data

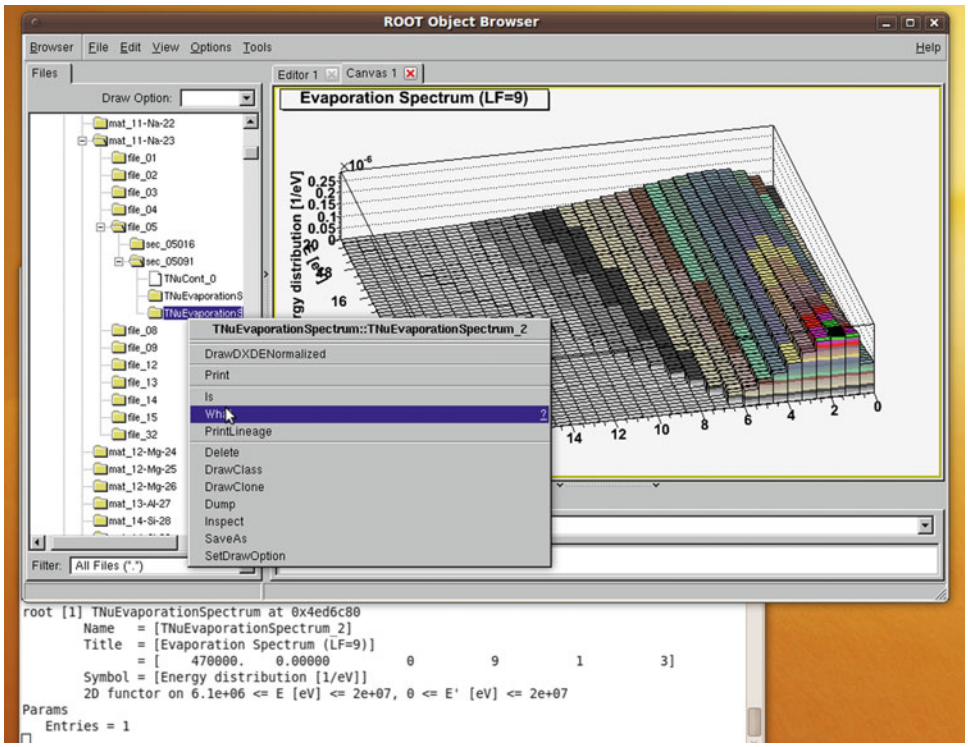

Fig. 4 ENDF data display

transport Monte Carlo simulations (FLUKA, Geant4, Geant3) with a common geometry modeler, which is also part of the ROOT system. Various routines have been developed for ENDF sub-libraries, data visualization, and user-friendly application. Extending these routines and verification of libraries is in progress [6].

Partitioning and Radiochemistry of Thorium

Actinide partitioning is a strategy for the management of high-level nuclear waste (HLW). Green extractants have been designed and deployed for the separation of long-lived radionuclides, including the actinides $^{241/243}$Am, ^{237}Np,

and ^{231}Pa, from HLW. Pyro-reprocessing is considered the most appropriate technique to recover these radionuclides from the discharged spent nuclear fuels as well as from the irradiated actinide targets. It is amenable for short-cooled fuels and has the advantage of compactness over the conventional aqueous technique.

However, this technology is still in its infancy and will be available for industrial application in the distant future. In the meantime, these long-lived alpha emitters are needed to pursue basic research nuclear data such as cross sections of neutron capture and fast neutron fission of ^{232}Th and activation products like ^{233}Pa and ^{233}U are pivotal to the use of Th in an energy amplifier. Developing efficient procedures to separate valuable isotopes such as ^{137}Cs, ^{90}Sr, and ^{99}Mo from irradiated Th is required. Fission products like ^{137}Cs, ^{90}Sr, and ^{99}Mo find extensive applications in industry and in nuclear medicine and cumulative yields of ^{90}Sr during fast fission of ^{232}Th (7.32 ± 0.36 %) and of ^{233}U (6.39 ± 0.33 %) are larger than those from the fission of ^{235}U, ^{238}U and ^{239}Pu. The inert nature of ThO_2 (and its fission products) and high solubility of SrO in acid media provide incentives to leach preferentially Sr over Pd, Y, Zr, and Th in dilute nitric acid/perchloric acid. Sr-selective chromatographic extraction resin was employed for purification of Sr from ^{232}Th and fission products, a process that took place in a perchloric acid medium [7].

Transmutation of LLFPs

Long-lived fission products (LLFPs) are the main cause of the long-term radiotoxicity and risk of nuclear plants. Among other FPs, ^{99}Tc is one of the major LLFPs (8.54 kg/GW/year for pressurized water reactors (PWRs) with 3.2 % ^{235}U); it is long-lived ($t_{1/2}$ = 211,000 years), water-soluble, and suitable for transmuting with the adiabatic resonance crossing method.

The novel method of transmutation by adiabatic resonance crossing (TARC) was pioneered by C. Rubbia et al. at CERN in the 1990s [8]. Transmutation of isotopes can be an effective way not only to reduce the long-lived fission product inventory, but also for the production of medical radioisotopes. Based on this concept, we have completed a design study and heat transfer analysis to determine the main physical parameters of the TARC system by considering the neutron production, energy deposition, and other related data. We assume the use of a 2 kW proton beam of 50 MeV with a 40 μA current, which can be delivered by the MC-50 Cyclotron at KIRAMS (Korea Institute of Radiological And Medical Sciences). A TARC setup is designed, including a neutron converter target and a heat removal system, by using the Monte Carlo particle transport code FLUKA and the ANSYS-CFX package [9]. The neutron converter target geometry is determined to capture completely the primary proton beams. The composition of the target and associated cooling system are optimized for the highest production of neutrons, for which a fin-shape beryllium solid target cooled by helium is proposed, as beryllium is to be preferred over lithium for practical engineering reasons. An elaborate design for the heat removal system is necessary, given the relatively high power deposited over a very localized region of the target. An optimum cooling system must take into account limiting factors such as material melting point, maximum velocity of fluid in the channels, etc., to obtain reasonable temperatures as well as adequate neutronic performance. Simulations of transmutation of ^{99}Tc for an experiment with KIRAMS MC-50 proton cyclotron have been carried out.

Life Cycle Assessment for ADS

We have developed a system model for the life cycle assessment (LCA) of pyro-processing for a closed nuclear fuel cycle. The LCA is developed by the United Nations Environmental Program (UNEP) and the Society of Environmental Toxicology and Chemistry (SETAC) for the purpose of estimating the comprehensive environmental impact of a nuclear power process. The impact covers the complete life cycle of the nuclear process, including material mining, milling, conversion, and fabrication of nuclear fuels as well as material collection, construction, operation, and disposal of nuclear waste at a power plant. Previous studies have analyzed the life cycle impact of the nuclear power process, however, these studies dealt with a once-through cycle only and excluded the reprocessing phase. To focus the technology development of next generation nuclear technology on being more sustainable, the environmental burden of the closed fuel cycle should be considered. In this work, we address the system boundary of pyro-processing (as one of the reprocessing technologies) for the LCA analysis and estimate its associated life cycle impacts. The study is a pilot research endeavor for applying LCA to nuclear technology. As a next step, a LCA would be applied to an ADS system to estimate its environmental performance [10].

ADS Cyclotron Design at Sungkyunkwan University

Recently, we have started a cyclotron design for ADS (Fig. 5). We have a three-stage cascaded connection: first, four injector linear accelerators at 70 MeV, 3 mA, then, four separated-sector cyclotrons, followed by two injection folded radiofrequency cavity cyclotrons. To achieve beams

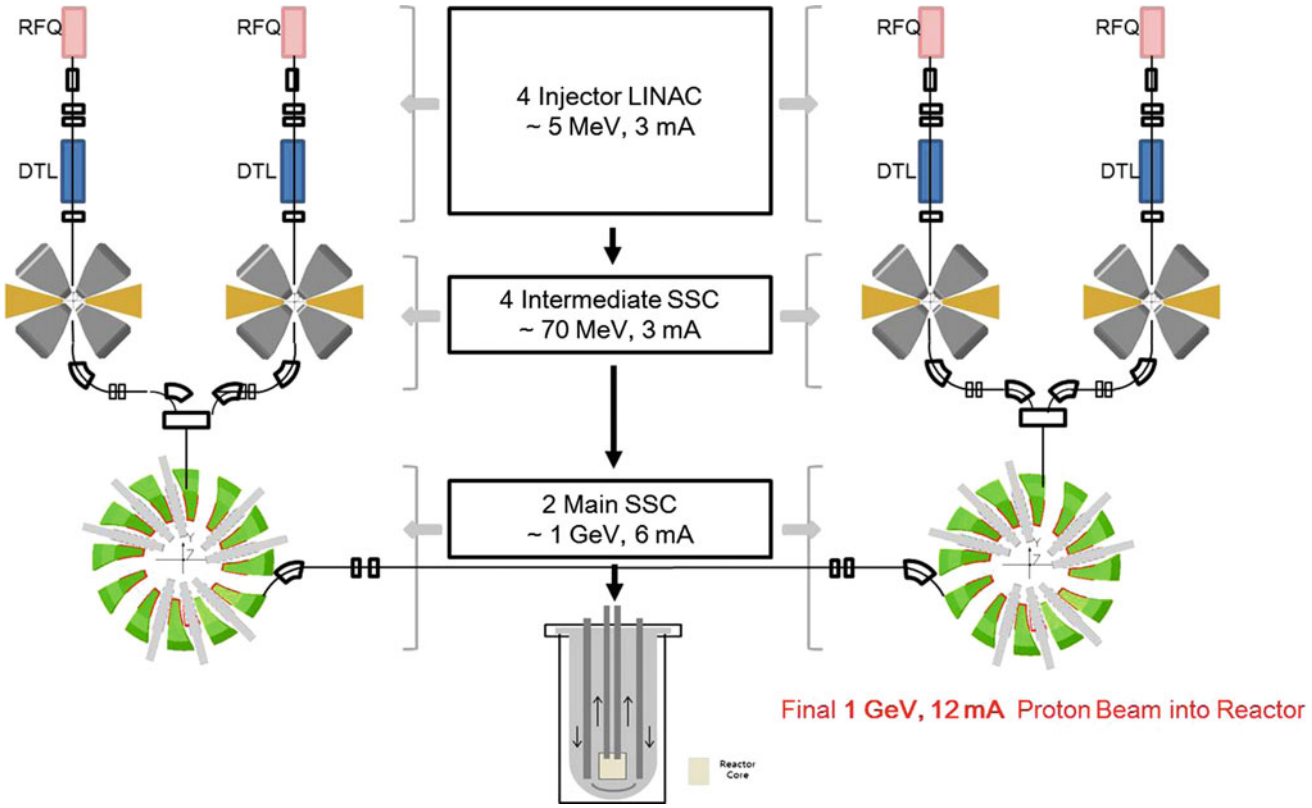

Fig. 5 Conceptual structure of an ADS for thorium energy generation. The characteristics of the intermediate SSC are given in Table 2

Table 2 Specifications of the intermediate accelerator

	Parameters	Values
Beams	Injection energy/extraction energy	8 MeV/70 MeV
	Estimated maximum beam intensity with 1 injector	1 mA
	Emittance	4 mm mπrad
	Type of extracted Ion	Proton, deuteron
Magnet	Pole radii—r_{min}/r_{max}	1 m/4.3 m
	External diameter	8.6 m
	Number of magnet sectors	4
	Magnetic gap	5 cm
	Sector angle on injection radius	30°
	Approx. total iron weight	424 Tons
	Average field	0.385 T
	Relative field variation	0.38–0.415 T
RF system	Frequency/Harmonics Number	74.3 MHz/6th
	Type of resonator	Double
	Dee Number/Dee angles	2/28°
	Q-value	14292
	Peak Voltage (Inj./Ext.)	150/200 KVolts
	Peak radial gain/turn (Inj./Ext.)	50/20 mm
	Estimated total accelerator efficiency	22 %

over 10 mA, we have used two duplicate systems. As known for higher beam currents (over 5 mA), space charge problems exist in cyclotrons. Thus, we have two machines and have inserted two direction beam injection in each: one upper beam and one lower beam, each with a current of 3 mA.

One of the first decisions to be made in the design process was the number of sectors in the magnet and the angular width of each sector necessary to achieve the final energy of approximately 70 MeV. To avoid the effects of resonances $\nu_z = 1$ and $\nu_r = N/2$, we have selected four sectors and a magnet sector angle of 30° ($\alpha = 30/90 = 0.333$). To achieve these goals, first hand calculations were done to determine the basic geometry of the magnet; the isochronous magnetic field was calculated by:

$$B_{\mathrm{iso}}(r) = \gamma \cdot B_{\mathrm{c}} = (1 + T/E_0) \cdot B_{\mathrm{C}}$$

The relativistic factor, γ, is a function of radius, which was calculated from:

$$B(r)/B_0 = \gamma(R) = \left[1 - (qB_{\mathrm{c}}/m_{\mathrm{i}}c)^2\right]^{-1/2}$$

2D simulations have been carried out to determine the basic magnet dimensions and trim coil. LANL Poisson 2D code was used. The advantage of 2D simulation is the ability to use small mesh sizes for areas with high magnetic field gradient.

Summary

Research activities have been focused on computational modeling, TNudy, partitioning and transmutation of LLFPs, accelerator-driven molten salt reactors, ADS cyclotron design, life cycle assessment of ADS, and applications (detectors, medical, etc.) at Sungkyunkwan University in Korea. A new nuclear data library based on the ENDF and ROOT system has been developed. Production of radioisotopes and transmutation has been studied based on the concept of the transmutation by adiabatic resonance crossing. Thermodynamic analyses of a multi-channel helium-cooled device were performed with the computational fluid dynamics (CFD) code, CFX. As a strategy for the management of high-level nuclear waste, actinide partitioning studies have been conducted using 'green' extractants. Preferential recovery of ^{90}Sr from irradiated ^{232}Th has been studied. We have conducted so-called life cycle assessment analyses for estimating the cost of the conventional and modified fuel cycles of nuclear power.

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Proposal of the ADS Research Stand Based on the Linac of the Institute for Nuclear Research of the Russian Academy of Sciences

S.F. Sidorkin, A.D. Rogov, L.I. Ponomarev, and E.A. Koptelov

Abstract

The results of discussions about a research ADS stand at the Institute for Nuclear Research (INR) with a thermal power of up to 5 MW for nuclear transmutation investigations are summarized in this paper. The basic approaches to the design process are stated. The physical and design features of the research ADS stand fit comfortably in the existing infrastructure of the experimental area of the INR linear accelerator (linac). Among other aspects, the physical and technical safety, the horizontal input of the beam, additional barriers for safety, which exclude loss of coolant, are discussed. Features of the tungsten and uranium targets providing the maximal yield of neutrons and rather high lifetimes owing to the distributed thermal load on the beam window are described. Assembly of the stand featuring modules with a potential high degree of prefabrication (modularity) is proposed. Possible structures and geometries of blankets are discussed. Results of the initial design studies are given.

Introduction

Among the global problems of modern nuclear power engineering are the issues of accumulation of long-lived radioactive products of nuclear reactions in the form of fission fragments and minor actinides (MA), processing and recycling thereof, as well as the potential risk of accidents, including those related to the loss of control over the nuclear chain reaction. Closure of the nuclear fuel cycle, as well as development of nuclear power engineering based on the uranium–thorium cycle ($^{232}Th + n \rightarrow {}^{233}U$) cycle in which the accumulation of minor actinides (MA) is minimal (negligible), can potentially avoid some problems, unlike the uranium–plutonium cycle ($^{238}U + n \rightarrow {}^{239}Pu$) with the entire spectrum of such isotopes.

One of the possible ways to overcome these problems is the development of accelerator-driven systems (ADS). Significant advances in accelerator technology has given further impetus to the expansion of work in the field of ADS, raising the possibility of creating accelerators with a beam power of about 5–10 MW and, accordingly, subcritical blankets with a thermal power of more than 100 MW [1, 2].

Fast reactors are also being considered for transmutation of minor actinides, but this technology brings the number of minor actinides in fast-reactor-based nuclear power engineering to the equilibrium level and does not ensure their total transmutation. However, in some ADS patterns, for example, MA transmutation in the direct proton beam, their number can be reduced to virtually zero.

It should be noted that, unlike thermonuclear neutron sources, accelerator-driven systems have no unresolved fundamental physical problems, such as the controlled thermonuclear reaction.

ADS technical implementation raises no concerns about safety: discussions are mainly held about the stability of the accelerated beam of protons and the economic feasibility of ADS, compared with transmutation in fast reactors and disposal in geological formations, as well as about the selection of the optimal ADS configuration and technical issues (input of an

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intense proton beam into the target, creation of long-lived thermally stable targets, etc.).

ADS installations have the following advantages:

- Operation in deep subcritical mode, which significantly reduces the likelihood of nuclear accidents;
- Operation in subcritical mode significantly extends the use of fissile materials with a low fraction of delayed neutrons (^{239}Pu, ^{233}U, MA);
- The MA fission reaction threshold is quite high (0.25–0.5 MeV). Therefore, the use of MA as a fuel in conventional critical fast reactors would lead to a substantial positive void reactivity effect, particularly in the case of accidental loss of coolant. In ADS systems operated in deep subcritical mode, this problem is less severe and it becomes possible to build an ADS with a large fraction of minor actinides in the fuel. In particular, an ADS can be used in thorium blankets for production of ^{233}U and minor actinides as fissile materials to ensure the necessary level of subcriticality.

The comprehensive assessment of different ADS concepts requires a research stand with a thermal power of about 5–10 MW, which would allow the creation and study of almost any configuration of subcritical blankets and targets, the study of physical, technical, and technological problems arising in the course of ADS creation, followed by post-irradiation studies of individual fragments of the installation in hot cells. Such an ADS stand with a thermal power up to 5 MW can be created on the basis of the high-current hydrogen ion accelerator at the Institute for Nuclear Research of the Russian Academy of Sciences (INR RAS) located in Moscow (Troitsk area). This seems to be the only location in Russia, and probably also in the world, where it can be realized in a relatively short period of time and at relatively low financial cost. It should be noted that, according to experts, there is no other prepared experimental site in Europe with an operable high-current accelerator that would meet the requirements of research reactors [3–6].

Moreover, as the spectral characteristics of the external neutron source of the INR RAS accelerator, with a proton energy of about 600 MeV and with a beam energy of about 1.3 GeV, at relative maximum of neutron yield, is identical to the full-scale installation in the proposed ADS stand [6], it allows the simulation of its main neutron physical characteristics.

The proposal can thus be compared in terms of its scope to reactor BR-5 (after upgrading to BR-10) [7] at the Institute of Physics and Power Engineering (IPPE, Obninsk), a reactor that played a key role in the successful program of development of fast reactors in Russia, the testing of new types of fuel compositions, and was a prototype for fast reactors BOR-60 and IBR-2 [8].

Another no less important motivation for this proposal, is the possible use of the ADS stand of the INR RAS as a second neutron source. This would allow the number of neutron guides and, thus, neutron spectrometers designed for research in the field of condensed matter physics, to be doubled.

Infrastructure Specifics of the INR Linac and Neutron Complex

Linear Accelerator (Linac)

The linear accelerator [9] produces proton pulses 200 μs long in a shape close to a rectangle. The design parameters of the accelerator include: proton energy, 600 MeV; average current, 500 mA; pulse repetition rate, up to 100 Hz. During the accelerator operation, the proton beam can be spontaneously interrupted. Heat shocks determined by these factors will directly influence the target and blanket performance. The design of the stand shall have special features similar to those provided in the design of pulsed reactors.

Some Characteristics of the Experimental Complex

The path of the beam in the horizontal plane runs at a height of 1.5 m above the zero (ground) level throughout the experimental complex site (Fig. 1). Accordingly, input of the beam into the neutron source and other installations is effected sideways at this height. Input of the beam into targets from above at a small angle to the horizontal is possible in principle, but requires significant changes to the beam layout and additional radiation protection measures. Input of the beam from below is excluded.

The handling of full cycle fissile materials is not available on site at the INR RAS, therefore, these works are to be performed in cooperation with industrial sites and institutions of the nuclear industry.

Neutron Source Complex

The neutron source complex [10–12] provides thermal and radiation protection, with a total thickness of about 10 m, the water-cooling system of the first and second circuits, and two boxes for installing neutron sources (Fig. 2). The high density of radiation protection and its large thickness are related to the emission of high-energy neutrons by the secondary radiation spectrum of targets, up to the energy of the original proton beam. The upper part of the protection is

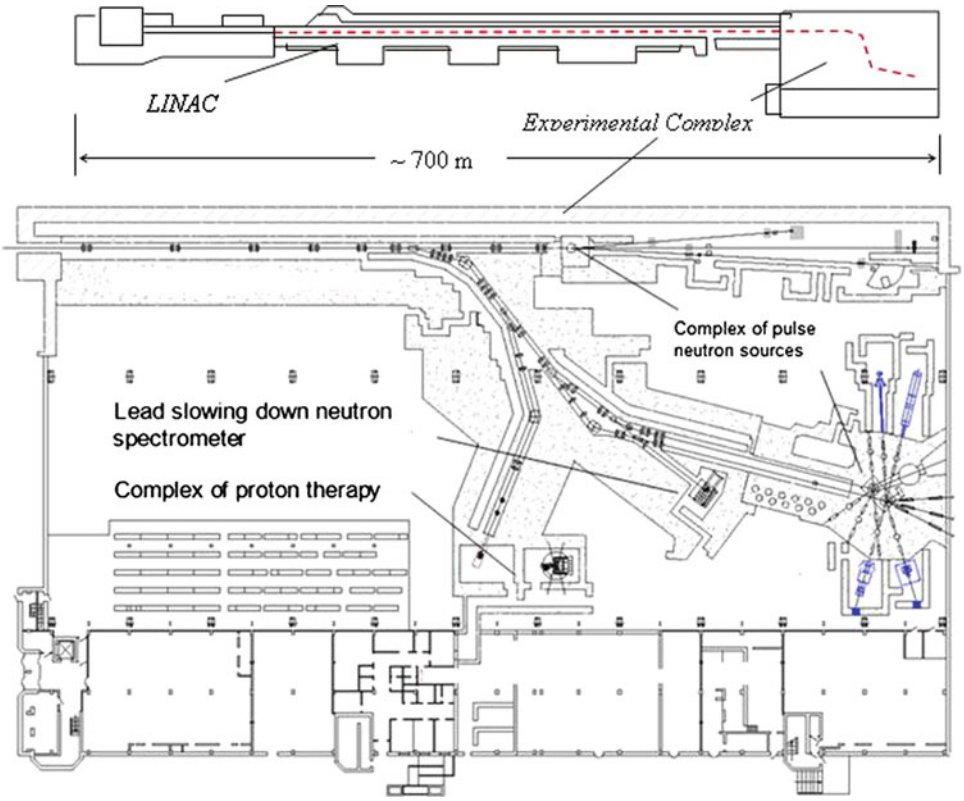

Fig. 1 Linac and experimental complex

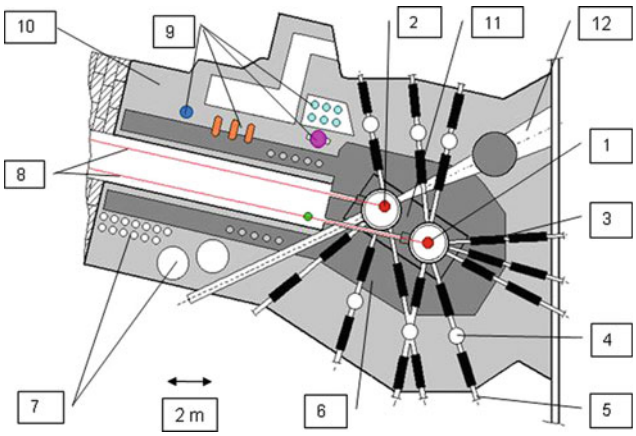

Fig. 2 Assembly of the neutron source: *1* cell of the pulsed neutron source; *2* cell of demonstrated research stand; *3* neutron gates; *4* vertical channels for additional equipment; *5* neutron guides; *6* iron shield; *7* storage of radioactive tanks and ampoules (modules); *8* proton guides; *9* equipment of the first water loop (pumps, filters, heat exchanger, etc.); *10* heavy concrete shield; *11* heat shield; *12* the wide aperture channel

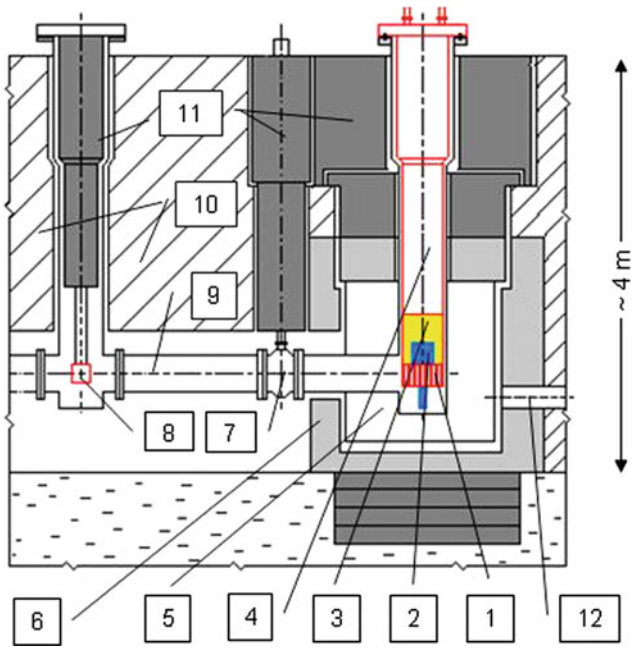

Fig. 3 Schematic of the pulsed neutron source: *1* tungsten core; *2* moderators; *3* beryllium reflector; *4* ampoule (module) with shielding plug, W core, Be reflector, and moderators; *5* gas tanks; *6* heat shield; *7* remote-controlled vacuum seal; *8* beam position sensor; *9* proton guide; *10* iron shield; *11* removable steel plugs; *12* neutron guides

removable, which makes it possible to assemble and disassemble the main equipment.

The first box contains an intense pulsed neutron source designed for study of condensed matter physics using external neutron beams. Its layout is shown in Fig. 3 [12–14].

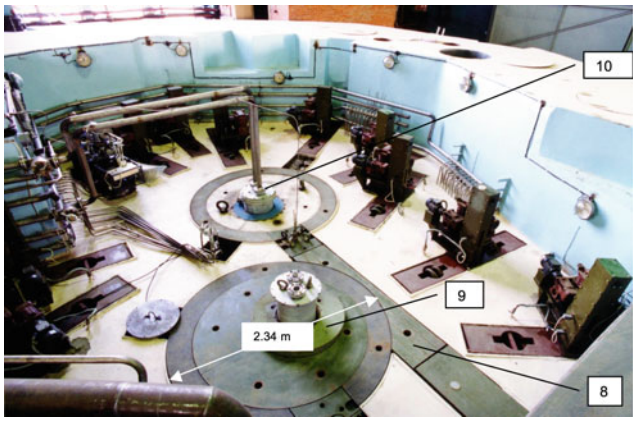

Fig. 4 Photograph of the beam target area showing the second cell position. The overview includes references to the steel shield of the proton guide (*8*), the ADS experimental cell (*9*), and the pulsed neutron source cell (*10*)

The neutron source consists of the following basic elements:

- A water-cooled ampoule, about 4 m in height, in the bottom part of which there are the target, moderators, and reflector, and above which there is a safety plug with channels for the coolant flow;
- Gas tank with vacuum bend for reducing the background of reflected neutrons at the lower moderator and joining the target and ion guide vacuum system;
- Remotely releasable seal, allowing disassembly of the whole equipment of central part of the neutron source;
- Proton beam parameter measuring system.

The side surface of the target module located inside the vacuum chamber joined to the ion guide forms the beam window. Such neutron source layout is flexible enough to allow the use of modules with different targets (W, U, PbBi, etc.) and moderators. It also permits us to perform a total replacement of all the equipment in the central part and to modify the source. Targets are cooled by Field's scheme, with the coolant input and output coming from above. It is assumed that the configuration of the ADS stand will be similar.

The ADS stand is proposed to be located in the second free box of the radiation protection (Fig. 4), the main dimensions of which are shown in Fig. 5. Basic dimensions of the stand are largely determined by the box dimensions together with the beam height level.

The neutron complex infrastructure was designed and built according to the requirements of the research reactors;

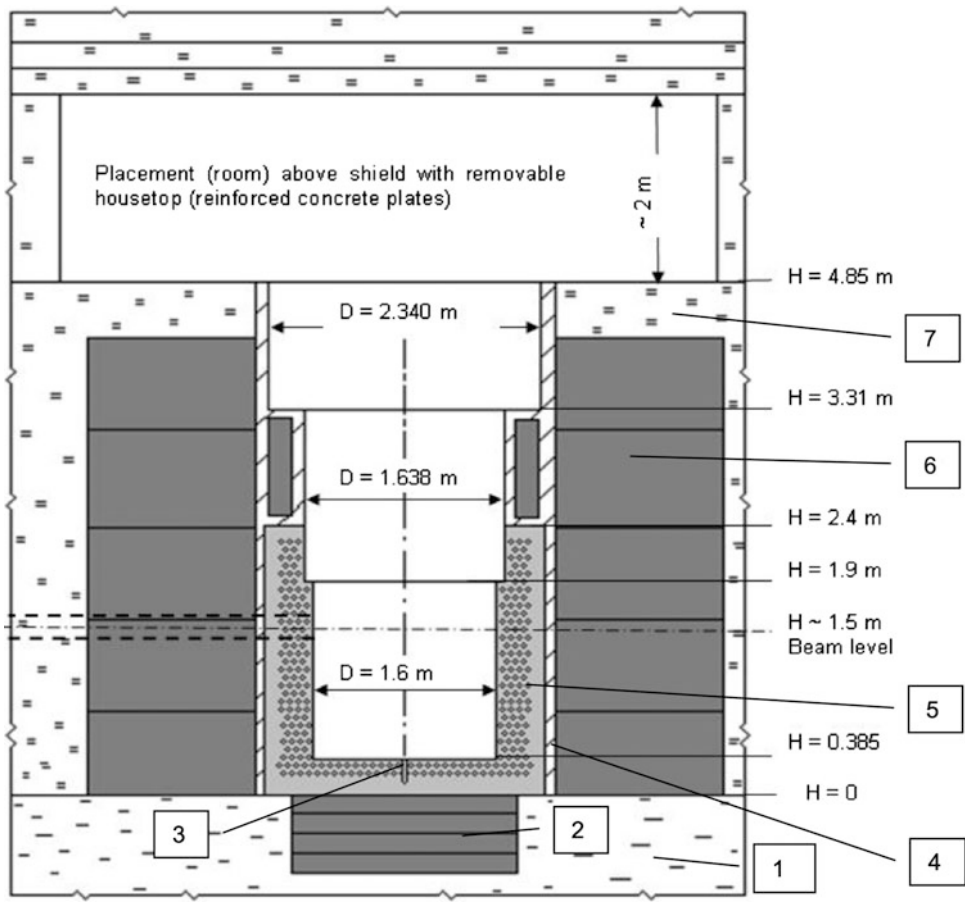

Fig. 5 Schematic of the second cell of the shield and its basic sizes: *1* basis, ~2.5 m; *2* steel plates (additional protection of subsoil waters against activation); *3* radioactive water collector in case of an accident; *4* layer of serpentinite; *5* water-cooled heat shield; *6* steel shield (thickness ~2.5 m); *7* concrete shield (density 2.8–3.5 g/cm^3)

it is consistent with a prospective increase in the accelerator current to 1 mA and with the use of uranium targets and it allows for high local densities of heat deposition in targets [12–14].

Installation Capacity and Basic Requirements

If the proton energy is 500–600 MeV, $Y \approx 8$–10 neutrons are produced per incident proton in PbBi or W targets. In uranium-based targets with cylindrical fuel elements, the neutron yield, Y, is $\sim$1.4 times higher [13, 14]. Thus, the installation capacity can be calculated by using the following formula: $P = YI_P M\omega/(v \cdot c)$, where I_p is the average current of protons, M is the multiplication factor; its value depends on the demonstration installation layout and shall be determined in the course of development according to the nuclear safety rules for subcritical stands. The parameter ω is the importance of the external neutron source, ν is the average number of neutrons produced during uranium nuclear fission ($\sim$2.5), and finally $c = 3.1 \times 10^{16}$ fissions/(MW · s) is the conversion factor. Depending on the multiplication factor (10–50), the stand thermal capacity can be $\sim$3–20 MW. However, owing to a number of limitations on the thermal protection and on the multiplication factor for the experimental complex, the proposed estimated thermal power would be limited to below 5 MW. Table 1 shows the average power estimation for the ADS stand with a proton energy of 300–600 MeV, a value that depends on the target type and composition, the average current of protons, I_p, the multiplication factor, M, and beam energy. The value of the neutron source, ω, is ≈1.3.

The neutron yield of the uranium target with *cylindrical* fuel elements and, thus, the stand thermal capacity are $\sim$1.4 times higher compared with W or PbBi targets. The use of the uranium-alloy-based targets with *plate* fuel elements similar to those used in the Rutherford (RAL) and Argonne (ANL) laboratories would increase the neutron yield and the stand capacity by $\sim$1.7 times compared with W or PbBi targets [15]. However, experience at neutron sources such as ISIS (RAL) and IPNS (ANL) show that uranium targets with plate fuel elements have a short lifetime ($\sim$3–6 months) in a direct proton beam with a current of $\sim$150–200 μA [16–18].

Table 1 shows that the power of $\sim$5 MW in the deep subcritical assembly mode, that is, when k_{eff} = 0.95 (M = 20), can be achieved with an average beam current of $\sim$200–250 μA.

A further increase in the multiplication factor is impractical due to more stringent nuclear safety rules and the increase in power and, thus, the background of neutrons between pulses. To reduce it and ensure stand operation in the mode of pulsed neutron source, one would require a reactivity modulator similar to that used in reactor IBR-2 [8]. If it is used in the blanket of fissile isotopes with a low fraction of delayed neutrons (e.g., ^{239}Pu, β ≈ 0.0023), the stand can be operated as a pulsed neutron source with $M \leq 20$, without a reactivity modulator.

Main Goals, Objectives, and Technical Requirements of the ADS Stand [3–5]

The ADS shall ensure the following:

1. Study of the physical, technical, and technological problems of various types of blankets, target devices, and combinations thereof with the accelerator.

Table 1 Expected average power of the research ADS stand depending upon proton current (I_P), multiplication coefficient (M), proton energy (E_P), and type of target

Average proton current I_P (μA)	Multiplication M (k_{eff})	Average power of blanket, P (MW), for proton energy 300–600 MeV							
		W (plates) or PbBi target				Natural uranium target (cylindrical fuel elements)			
		300	400	500	600	300	400	500	600
100	10 (0.90) 20 (0.95)	0.36 0.73	0.58 1.16	0.83 1.66	1.04 2.08	0.51 1.02	0.81 1.62	1.14 2.29	1.46 2.91
150	10 (0.90) 20 (0.95)	0.55 1.10	0.88 1.75	1.25 2.50	1.56 3.12	0.77 1.54	1.22 2.44	1.72 3.44	2.18 4.36
200	10 (0.90) 20 (0.95)	0.73 1.47	1.16 2.32	1.66 3.32	2.08 4.16	1.03 2.05	1.62 3.25	2.29 4.58	2.91 5.82
250	10 (0.90) 20 (0.95)	0.92 1.83	1.46 2.91	2.08 4.16	2.60 5.20	1.28 2.57	2.03 4.06	2.86 5.72	3.64 7.28
300	10 (0.90) 20 (0.95)	1.09 2.19	1.75 3.5	2.50 5.00	3.12 6.24	1.54 3.08	2.43 4.86	3.43 6.86	4.37 8.74
Neutron yield, Y (n/p)		3.5	5.6	8	10	4.9	7.8	11	14

Importance of the primary neutrons, $\omega \approx 1.3$

2. Study of the various fuel compositions with different compositions of minor actinides and various designs of fuel rod arrays (FRAs) in PbBi eutectic.
3. Transmutation of macro quantities of fission fragments and minor actinides.
4. Possible use of the stand as a neutron source.
5. Quick access to the experimental channels of the blanket, removal of irradiated fuel assemblies, and transfer thereof to hot chambers for research, loading new samples, and FRAs.
6. Relatively quick and easy reassembly of the blanket, replacement of targets, and assembly of other ADS configurations.
7. Fast neutron spectrum in the blanket for study of burnout of minor actinides.
8. Thermal neutron spectrum in the blanket for the study of transmutation of long-lived fission fragments and production of neutrons for research in the field of condensed matter (i.e., possible use of the stand as a pulsed neutron source).
9. Assessment of possible arrangements of neutron guides and their output outside of the blanket.
10. The design of fuel assemblies, FRAs, and configuration of the stand shall accommodate possible spontaneous interruptions and recovery of the accelerator proton current and related abrupt changes in the average power and temperature of the blanket, as well as the occurrence of dynamic, thermal, and mechanical stresses in the structural elements of the core.

It is quite difficult to meet these contradictory requirements within the standard design of a PbBi reactor as relatively frequent and quick access to the reactor pool with a hot ($\sim$300 °C) coolant (PbBi) is needed.

Therefore, the research ADS stand is proposed to be built on a modular principle (highly prefabricated modules) [3–6] in order to ensure relatively quick and easy assembly and disassembly of the installation, interim storage, transportation, and to allow research on the highly active irradiated parts. The modules are installed in a vertical position, hung on the safety plug through flanges or mounted on the support grid. The coolant input and output are from above, and inside each module there is a Field's circulation circuit. In some cases, depending on the purpose, the modules may contain an integrated pump and heaters in the top sections (e.g., PbBi target) as well as inlets for maintaining the chemical composition and sampling.

Stand Conceptual Layout [3–5]

In the central part of the housing there is a target surrounded by the fast water-cooled blanket that is based on highly enriched fuel, and a tight grid of fuel assemblies (the ratio of the grid spacing to the diameter is ≈1.03), as shown in Fig. 6. The proton beam is introduced horizontally.

Inside the blanket there are several zones, with PbBi coolant, that are constructed as separate modules (closed airtight plugs) in which different fuel compositions and fissile isotopes, such as FRAs with UN, UO_2, MOX fuel or with minor actinides, etc., can be used (Fig. 7).

Target

In this ADS stand configuration, the following is to be used:

- A tungsten target composed of plates;
- A target based on cylindrical fuel elements with metal-uranium alloy. These fuel elements are used in the breeding zones of the commercial fast reactors BN-350 and BN-600; thus, it is possible to use existing technologies. The disadvantage of this configuration is a relatively short lifetime of such targets ($\sim$1 year) and more frequent replacement of the target module.

The neutron spectrum of the PbBi target is similar to the spectrum of the W target; therefore, the PbBi target can only be used as a prototype target for commercial ADS.

PbBi Inserts

The PbBi alloy is circulated inside each insert; the upward flow of coolant is in the center and the downward flow is on the periphery. The insert is water-cooled by the first circuit along the side surface. To maintain temperature and ensure safe operation, the insert has a double wall. Between the walls there is a gap of $\sim$1 mm, which is a heat barrier (Fig. 7).

Each insert has its own heater, which allows us to:

- Vary and maintain the specified temperature level in each ampoule;
- Ensure pre-melting of the PbBi alloy and heating of the entire ampoule after its transfer to the experimental area of the INR and installation in standard position;
- Partially compensate for decreased temperature of the PbBi coolant and prevent its solidification by increasing the heater current load automatically in case of a sudden stop of the accelerator or beam loss. Moreover, the insert can be equipped with its own electromagnetic pump. This allows the rate of PbBi coolant flow and its temperature to be varied.

This approach allows the amount of liquid metal to be minimized and the use of PbBi modules that can be assembled and tested at industrial sites of the nuclear industry, similar to fuel assemblies of conventional reactors, and then delivered in a sealed solid (frozen) state to the experimental

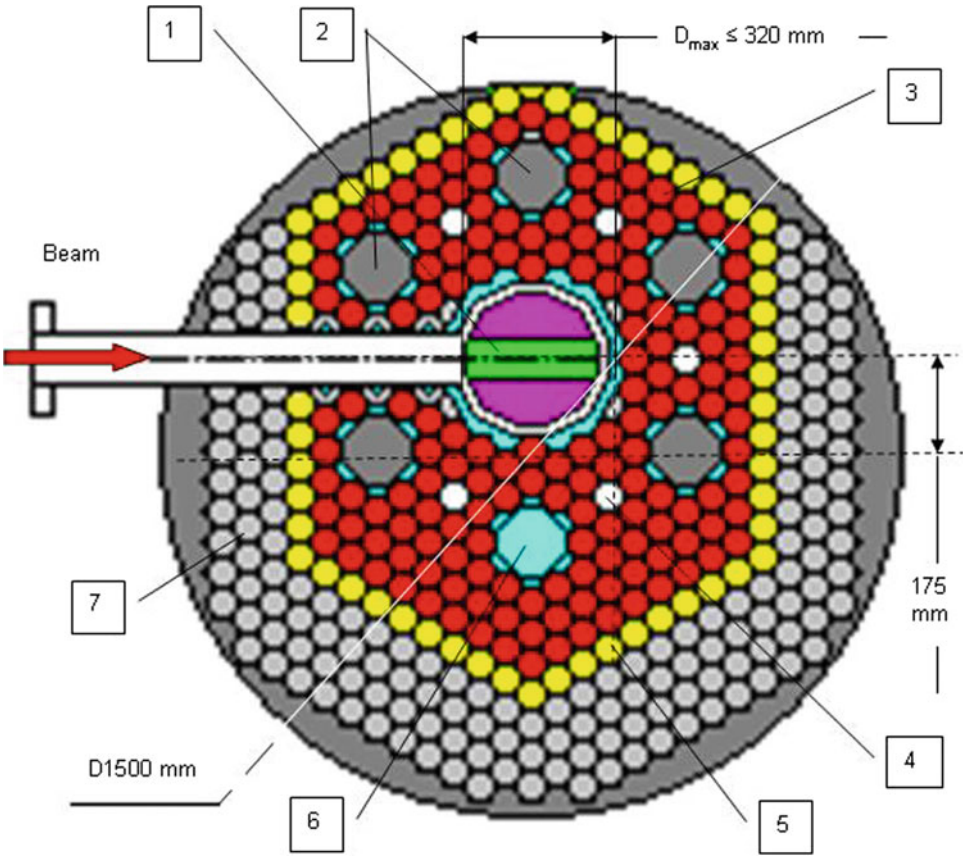

Fig. 6 Conceptual scheme of one of the possible variants of the research ADS: *1* target module; *2* hermetical PbBi capsules with highly enriched fuel and minor actinides; *3* fuel assemblies of the water-cooled blanket; *4* module of the control systems; *5* decoupler (if required); *6* thermal neutron trap (moderator), which can be placed anywhere; *7* reflector

area of the INR RAS and installed in the blanket. After loading into the stand and transition to the liquid phase, and once the required fluence has been achieved, modules are cooled again and returned in the solid state to the nuclear complex industrial site for post-irradiation research and follow-up disposal.

According to experts at IPPE, such a module is able to work for approximately 2 years without support systems and any cleaning of the PbBi coolant, which is compatible with the term of its operation.

Such a scheme of cooling a PbBi module by ordinary water (~1 atm) along the lateral surface with a thermal barrier was proposed in reference [19]. More detailed modeling of similar PbBi modules with air cooling has been reported in references [20–22].

Some Technical Aspects on Safety

Fuel rods with minor actinides and testing of the associated working life when placed inside a PbBi capsule creates three additional barriers to the spread of fissile materials and fission fragments: the PbBi array and two shells made of stainless steel. In operating conditions, if one of the two module shells is destroyed, the water flow that surrounds it creates conditions for local solidification of the PbBi eutectic, which results in leak mitigation ($C_{PbBi} \approx 0.147$ kJ/(kg · K) $\ll C_{H2O} \approx 4.2$ kJ/(kg K)). A similar situation occurs in the case of local destruction of both module shells.

Any manipulation of the PbBi modules and fuel assemblies, which include macro quantities of minor actinides, are only allowed in the hot chambers of the fuel cycle company's industrial site. Therefore, the size of the ampoules shall be limited and be in conformity with the dimensions of hot chambers. If the fuel rods are destroyed and cannot be removed from the PbBi capsule, the latter may be sent in its entirety for long-term storage with follow-up disposal.

The safe overload of the solidified PbBi modules can be carried out by means of the usual technological container, as in a research reactor. Therefore, the necessities of fully automated processes and the creation of completely independent and expensive systems to access the reactor pool, such as in the MYRRHA project [23], are not required. With the approach suggested in this paper, the practically absolutely safe transportation of PbBi modules is provided for, as the fuel rods are enclosed within solid PbBi eutectic, and are surrounded by double stainless steel walls once the module is located in the transport container.

Some Characteristics of the Blanket

The fuel assembly tight lattice in the water-cooled part of the blanket and the high enrichment (~40–80 % U-235)

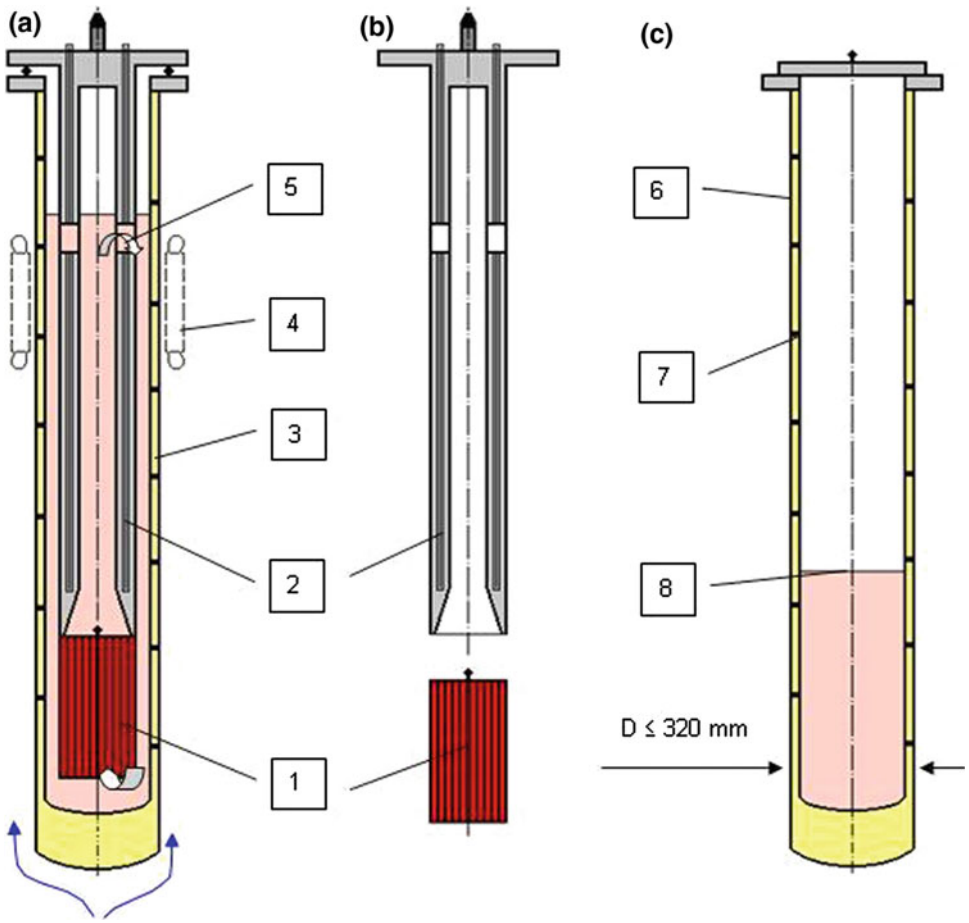

Fig. 7 PbBi module conceptual scheme: **a** assembled capsule, **b** fuel assembly and displacer (removable elements), **c** body of capsule with PbBi after removal of fuel assembly and displacer. *1* Fuel assembly; *2* spreader of up-going and down-going flows with built-in heater, the displacer of liquid metal, and holder of fuel assembly; *3* binary body; *4* electromagnetic pump; *5* direction of liquid metal flow; *6* gas gap (~1 mm) of the heat barrier; *7* spacer; *8* level of liquid metal after removal of displacer and fuel assembly

provide a low volume fraction of water (~15–18 %) and a fast neutron spectrum. In this lattice, a neutron collides six to eight times before absorption or leakage from the core and does not have enough time to significantly reduce its energy (in water a neutron undergoes about 18 collisions on average before reaching thermal energy).

As water cavities are formed by filling the space between hexagon cassettes of fuel assemblies and cylindrical PbBi modules, the neutron spectrum around PbBi may be varied by using displacers. For example, it can be made faster through water displacement around the PbBi modules or, alternatively, softer through changing the thickness of water around the module.

Spectrum-hardening in PbBi modules and reducing fission cross sections can lead to a reduction in the level of heat output. This effect can be compensated for by:

- Using higher density fuel (UN, UC, etc.) and through enrichment compatible with PbBi. For example, uranium is ~20 % enriched in the water-cooled part of the blanket and ~70–90 % enriched in the PbBi modules. For the water-cooled part of the blanket, UO_2 or UO_2–PuO_2 is still the only fuel type actually compatible with water;
- Creation of a PbBi module as a FRA with a large number of fuel rods (more than in the FRA of the water-cooled portion of the blanket), thus increasing the volume fraction of fuel;
- Increasing the power-supply density in peripheral fuel assemblies of the PbBi module and, thus, in the whole capsule using moderated neutrons due to the presence of water in the space between the hexagonal FRAs and the cylindrical shell of the PbBi capsule. In the central part of the module, where minor actinides can be present, the neutron spectrum will still be fast as slower neutrons are absorbed by the peripheral fuel assemblies. This also allows us to collect and study the cascade blanket if fuel based on threshold fissile isotopes is used in the peripheral fuel assemblies of the PbBi modules [2, 24, 25].

In this way, an arrangement can be implemented in which the main heat power will be extracted in PbBi inserts, while the water-cooled blanket will be used to support and maintain the specified multiplication level at a relatively low power supply. However, the heat output density in the PbBi inserts shall be maintained at a level of no less than ~100 kW/L.

Physical Aspects of Safety

During assembly and reload of fuel cassettes of the fast water-cooled blanket or replacement of PbBi modules, water fills into the space thus vacated, which becomes a source of thermal and intermediate neutrons. This leads to a positive effect of reactivity, despite removal of the PbBi module or fuel assembly. To prevent this positive effect of reactivity during blanket assembly, replacement of the fuel assemblies, or PbBi module insertion into water, the FRA shells in the water-cooled part of the blanket are to be made of a hafnium-based alloy (thermal neutron absorber). For example, the Hf–Nb(2 wt%)–Zr(20 wt%) alloy, which is a complete analog of one of the zirconium alloys used for production of fuel rod tubes.

Thus, the overall effect of such a design is to guarantee inherent safety. A decrease in the volume fraction of water, for example, as a result of boiling or partial loss of coolant, leads to a faster neutron spectrum and a large negative effect of reactivity. An increase in the volume fraction of water leads to intensified capture of thermal and intermediate neutrons in the hafnium shell of the FRA. If the fuel assembly or PbBi module is removed and replaced with water (trap of thermal neutron), the reactivity effect will also be negative (Fig. 8).

These two effects allow us to:

- Quickly and easily change the configuration of the fast blanket, replace irradiated fuel rod arrays, PbBi modules, and the target under a layer of water, as in pool-type nuclear reactors;
- Place traps for thermal neutrons and moderators in any accessible part of the fast blanket;
- Create an effective reactor control system based on traps for thermal neutrons.

The use of hafnium shells leads to additional absorption of neutrons in the blanket compared with FRA shells made of stainless steel or zirconium. This negative effect is partially compensated for with the following measures:

- Increasing the number of fuel rods in the FRA, for example, from 19 to 37, and, thus, decreasing the amount and volume fraction of hafnium in the blanket;

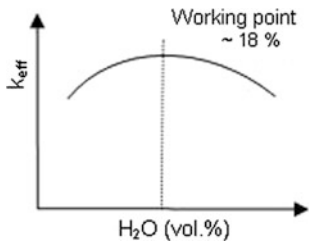

Fig. 8 Relationship between k_{eff} and H_2O volume fraction

- Reducing the thickness of the FRA shell, for example, from ~1 to ~0.5 mm;
- Reducing the percentage of hafnium in the alloy of the FRA;
- Reducing the total volume of the water-cooled part of the blanket by increasing the number of PbBi modules and their cross sections.

These provisions are confirmed by numerous calculations, one of which is given below.

Control System and Compensation Reactivity Effects

Movement of water displacers inside the thermal neutron traps allows us to simply and naturally create regulators, reactivity compensators, and, as necessary, emergency protection. For example, step-by-step movement of the water displacer by gravity or by means of a hollow displacer relying on Archimedes buoyancy, is possible depending on the purpose of the control element. It is much more technically difficult to create a control system in the high-temperature PbBi blanket area.

Furthermore, it is assumed to be possible to compensate for temperature effects of reactivity and, partially, burnout by increasing the beam intensity. The blanket multiplication factor is related to the neutron multiplication factor as follows: $M = k_{eff}/(1 - k_{eff})$. The decrease of k_{eff} by 1 % at the initial value $k_{eff} = 0.95$, leads to the decrease in the multiplication factor $M = \Delta k_{eff}/(1 - k_{eff})^2$ by 4 % and, thus, will require increasing the accelerator intensity by the same value. It means that this method of stand power compensation within ~2 % is well within the technical capabilities of the accelerator.

Fuel Rods

Fuel rods of the fast pulsed reactor IBR-2 (Joint Institute for Nuclear Research) that have been well-proven during long operation in pulsed mode in the liquid metal coolant (Na) at 450–500 °C and have passed further tests for heat shock, are proposed to be used as a prototype for the fuel rods. The fuel rods have special fittings that prevent fuel pellets (PuO_2) from hanging up in the process of heat shock and swelling and are applicable in both aqueous medium and lead-bismuth coolant at ≤400 °C. These circumstances are very important under the conditions of pulsed heat output in the blanket caused by a pulsed mode of accelerator operation and irregular spontaneous interruptions of the average current of protons. Furthermore, rated specific heat loads on fuel assemblies in the PbBi inserts (~100 kW/L) of the research ADS stand correspond to the average loads on fuel assemblies in reactor IBR-2 (design load is ~200 kW/L, operating load is ~100 kW/L), which is also important for justification of the project and licensing.

Thus, this conceptual layout allows all the conflicting initial requirements to be sufficiently satisfied, including fast and thermal neutron spectra, transmutation of MAs and long-lived fission fragments, and creation of moderators and thermal neutron beams. Negative reactivity coefficients are ensured in the process of operation, including in the course of relatively simple reassembly of FRAs and PbBi modules of the fast water blanket under a layer of water and the assembly of other ADS configurations, and experimental research thereof.

For example, the usual fuel assemblies of the water-cooled blanket can be installed in place of the PbBi modules or, alternatively, the number of PbBi modules can be increased by boundary displacement between the blanket and the reflector.

The fast thermal blanket can be assembled on the same support grid as the fuel assemblies and reflector. For example, the width across flats of the fuel assemblies with standard zirconium shells of the water-cooled part of the blanket remains the same, but fuel rod diameters are modified so that their pitch inside the cassettes is approximately equal to 1.3 of the rod diameter, as in thermal reactors. In the peripheral fuel assemblies included in the PbBi modules, to prevent modules overheating by thermal neutrons, fuel based on threshold fissile isotopes, such as NpO_2, can be used.

Using other modules of similar design (instead of PbBi modules), but with other compositions, for example, molten salt with dissolved minor actinides, is possible, too. Possible examples include FLiNaK, with a melting point of 454 °C and possessing high solubility of PuF_3, UF_4, and fluorides of minor actinides [26]. In this case, one should change the parameters of the heat barrier (thickness of gas gap) first of all and use hastelloy as the structural material for the inside shell of the module.

However, with such a conceptual approach, all other stated requirements can also be satisfied more or less easily.

Some Design Characteristics of the Installation

One of the possible designs of the stand is shown in Fig. 9.

Horizontal Beam Input

The horizontal beam input is supposed to remain as in the present configuration of the neutron source located in the adjacent box (Fig. 3). Such an arrangement will allow the target to be placed in the vacuum chamber directly connected to the ion guide, thus removing the need for an additional window. However, in this case it is necessary to take additional measures to prevent the loss of water coolant through the ion guide and blanket exposure, including at the stage when the blanket, after beam removal during the cool-down process, temporarily becomes a fuel storage. For this purpose, the following two additional structural elements are to be included in the stand design:

1. A high-speed vacuum shutter in the ion guide. This is placed on the outside of the stand housing and is triggered by the presence of water vapor in the vacuum system on the blanket side;
2. A thin-walled metal partition inside the blanket covering the ion guide and vertical vacuum chamber and connected thereto for installation of the target module. Its purpose is to ensure the presence of water in the blanket for removal of residual output heat in FRA cool-down mode in case the ion guide and vertical vacuum chamber burn out. In one of the possible variant designs, the partition wall is replaced with a safety enclosure covering the horizontal part of the ion guide.

It should be noted that the layout of beam input diagonally (sideways from above), as shown in Fig. 10, is the most beneficial layout solution. In this case, the level of coolant in the blanket and target is maintained even in case of the destruction of the beam window or target module, access from above to all units of the blanket and ion guide is provided, and all other benefits are still available. Thus, this provides a free space above the reactor as well as spatial separation of ion guide and reactor equipment elements (actuators of control and protection systems, instrumentation, etc.). Furthermore, as the flow of cascade nucleons is directed downwards, radiation shielding can be minimized. However, it is necessary to adapt the stand in the horizontal plane of the beam path, as otherwise the ion guide will need a vertical bend before the beam entrance into the blanket, which is impossible in the existing conditions of the experimental complex of the INR.

Note also, that such input of a beam promotes an increased mean lifetime for the target module and window. However, the proton beam and beam guide should both have the form of a narrow ellipse, thus providing a large cross section for irradiation, and good neutron connection between the two halves of the blanket. Neutron yield also increases because of prolongation of the target material along the beam path. In addition, for the target module with axial symmetry (e.g., PbBi target), the rotation of the module round a vertical axis in principle is possible, which also increases the effective area of interaction of the beam with the target surface, and the mean lifetime of the target module.

Vertical beam input from below requires large basements to be built under the stand for beam layout and for maintenance of ion guide elements. Furthermore, enhanced protection on top of the stand due to the presence of cascade

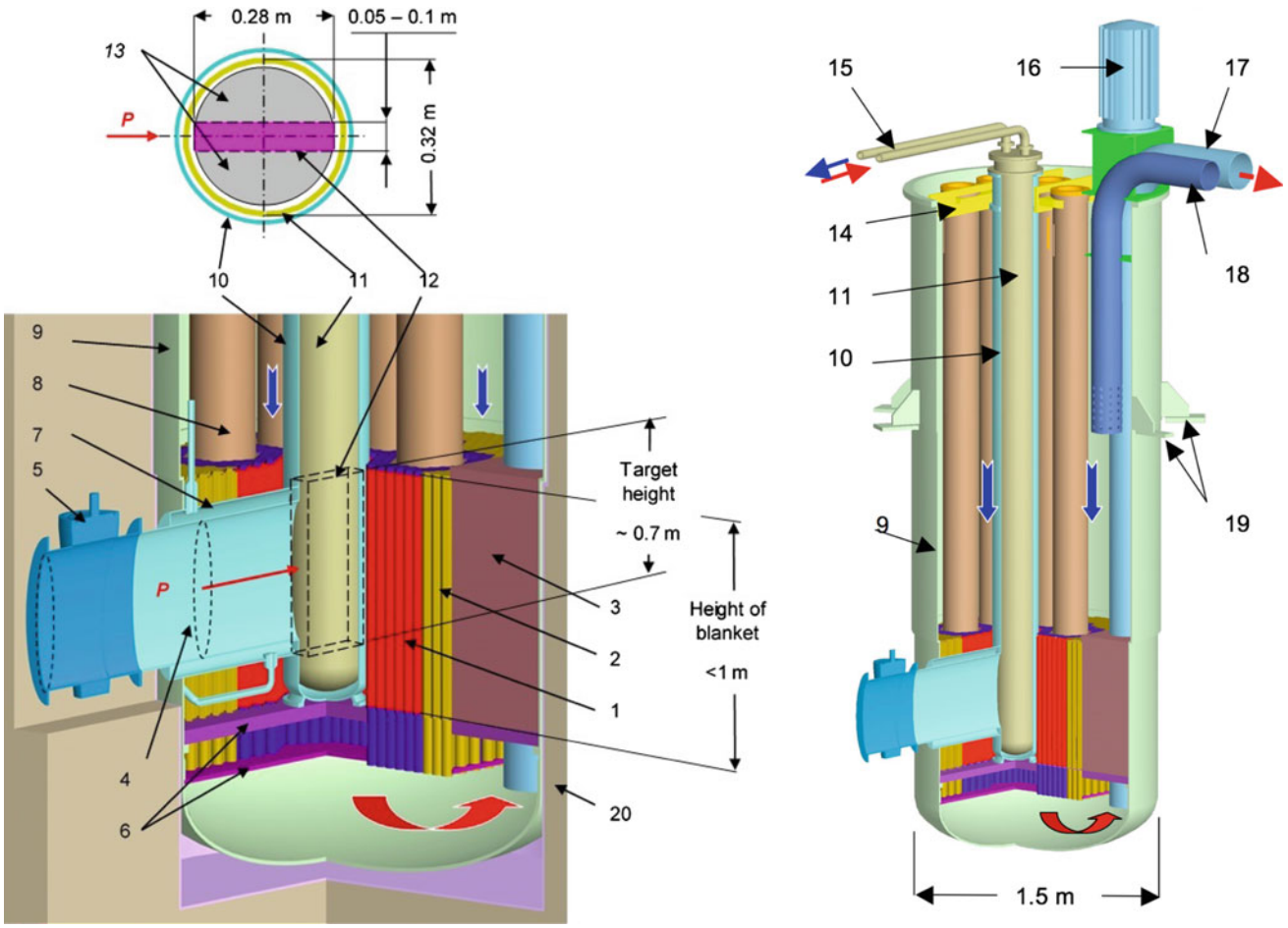

Fig. 9 The main elements of the ADS stand: *1* fuel assemblies; *2* removable displacers; *3* stationary displacer; *4* proton guide, the *dotted line* shows the cross section (*narrow ellipse*); *5* vacuum shut-off gate; *6* support grid; *7* safety shroud; *8* vertical channels for PbBi modules; *9* shell of ADS stand; *10* vertical vacuum chamber (locus for target module), jointed with proton guide; *11* target module; *12* contour of target; *13* displacers, *14* crossbar for PbBi modules; *15* input and output of the coolant of the target module; *16* electric drive of pump; *17, 18* input and output of the water coolant of the blanket; *19* carriers of ADS stand; *20* radiation shield. Typical dimensions of the target in cross section are shown

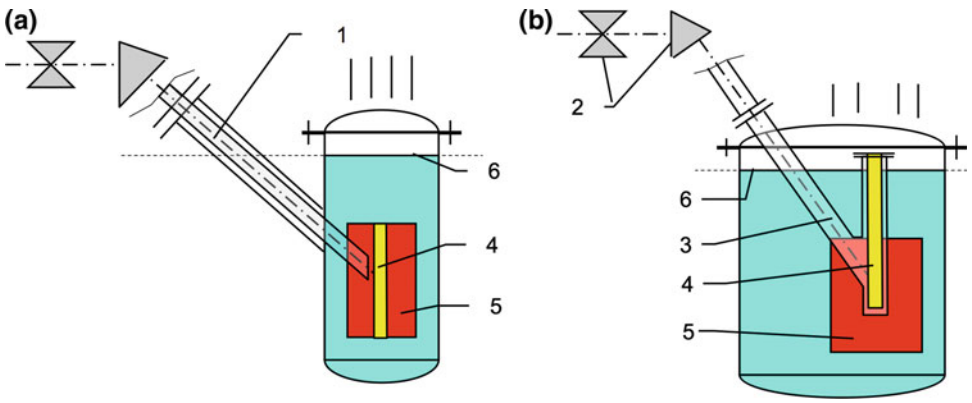

Fig. 10 Schematic of the input of a proton beam diagonally (sideways from above): **a** with separate cooling window on the removable portion of the ion guide; **b** with placement of target module inside vacuum chamber that is connected with the proton guide. *1* Removable part of proton guide; *2* vacuum shut-off gate and beam-bending magnet; *3* vacuum chamber; *4* target module; *5* blanket; *6* water level in body after breakdown of window or target module

nucleons would be necessary, and it does not ensure maintenance of the level of the coolant in case of an emergency. If the beam is introduced vertically from above, the above-reactor space is blocked by ion guide and magnetic optics elements, which makes it more difficult to assemble or reassemble the blanket and replace FRAs and individual modules.

Target Module

The target module (Figs. 3 and 9) is installed in the vertical vacuum chamber joined to the ion guide. The space between the cylindrical housing of the target module and the target (Fig. 9) is filled with inserts that provide minimal absorption and reflection of neutrons emitted by the target. The vertical target dimension of up to 70 cm (lateral dimension of up to ~9 cm in the case of uranium targets) provides a relatively large area of interaction of the proton beam with the target materials, thus reducing the radiation and heat load and, hence, increasing the average lifetime of the target module. Therefore, the ion guide will be in the shape of a rather narrow ellipse or rectangle. Furthermore, a narrow ion guide enhances interaction between the adjacent parts of the blanket.

PbBi Module Positions

The position of the PbBi modules against the target may be varied. This can be effected by moving the assembly of brackets using channels along the support bars. To facilitate adjustment of the channel axes, the bars have constructive samples that may be divided according to the seat spacing in the support grid.

The initial asymmetry of the box design against the proton beam and the relatively large dimensions of the box allow us to create a housing structure for the stand with integrated circulation pumps for blanket cooling. Such modification allows the design to be significantly simplified without compromising safety and provides coolant input–output from above through the upper cover of the installation.

Simulation of Neutron Physical Characteristics [5]

To confirm the physical and technical properties included in the conceptual design of the ADS stand, several configurations of the blanket and the target were simulated by using the Monte Carlo method. Calculations were made in several stages.

1st Stage

Searching for the optimal configuration of targets to ensure the maximum neutron yield from the target.

The main research objectives are:

- To determine the optimum target thicknesses of U and W, which ensure the maximum neutron yield into the blanket per proton;
- To determine the optimum displacer between the target and the cylindrical body of the target module.

2nd Stage

Examination of the basic neutron physical characteristics of the blanket. After a series of calculations in which $k_{\text{eff}} \rightarrow 0.95$, the following version was taken as a basis of the physical properties that were investigated further (Fig. 11 and Tables 2 and 3).

Fuel: a mixture of natural uranium dioxide (80 %) with plutonium dioxide (20 %) enriched with ^{239}Pu by 95 %. During calculations the percentage varied.

The number of PbBi modules is six; each module replaces seven fuel assemblies of the water-cooled part of the blanket. The total number of fuel rods in the PbBi modules is 85; wherein 19 central fuel rods contain NpO_2 for simulation

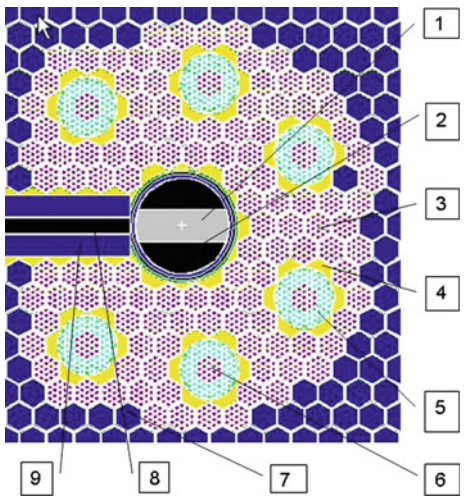

Fig. 11 The basic computational scheme of ADS stands for the study of principal physical characteristics: *1* target; *2* hollow or Al displacer between target and cylindrical body of target module; *3* assembly of 19 fuel rods; *4* water cavity between the cylindrical body of the PbBi module and the fuel assemblies; *5* PbBi module, *6* 19 central fuel rods with NpO_2 in the PbBi module (MA imitation); *7* elements of Al reflector; *8* proton guide; *9* displacers and constructional elements between proton guide and blanket

Table 2 Fuel assembly parameters

Width across flats of the assembly grid	4.3 cm
Outer width across flats of the assembly	4.2 cm
Assembly canning (hafnium alloy)	1 mm thick (Fig. 12)
Number of assemblies with MOX fuel	112

Table 3 Fuel rod dimensions (reactor: IBR-2M)

Fuel rod grid spacing	0.89 cm
Fuel pellet diameter	0.71 cm
Outer diameter of fuel rod shell	0.864 cm
Inner diameter of fuel rod shell	0.77 cm
Clearance between fuel rod	0.03 cm
Number of fuel rods in the assembly	19 (Fig. 12)

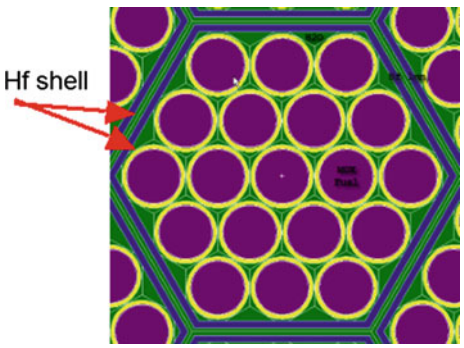

Fig. 12 Schematic of the fuel assembly

of minor actinides, the remainder of the rods contain PuO_2. Geometrical dimensions of the fuel rods are similar.

The height of the blanket is 90 cm.

During calculations the following parameters were varied:

1. Number of fuel assemblies in the water-cooled part of the blanket, which were replaced with Al inserts;
2. Number of PbBi modules (from six to three);
3. Spacing between PbBi modules and the target;
4. Percentage of PuO_2 in the fuel of both parts of the blanket;
5. Thickness of the Hf shells of the fuel assemblies;
6. Composition of the ion guide environment;
7. Core height.

The relative position of the blanket and the target were investigated in order to find the maximum importance of the external neutron source, ω, as well as of the effect of replacement of PbBi with D_2O, the influence of water between the FRA and cylindrical shells of the PbBi modules and of its substitution with different materials, along with other characteristics.

Main Results and Conclusions [5]

Target Optimization

Target optimization allowed us to find configurations that provide the maximum neutron yield from a W target of approximately 9.7 n/p and from a U target of approximately 14 n/p with a proton energy of 600 MeV. A void is the best displacer for surrounding the rectangular target inside the cylindrical target module (Fig. 9). Therefore, hollow displacers of Al alloy (15 % of the volume being metal) were further investigated. Solid displacers made from Be, Ni, and Al reflect too many neutrons back to the target, thus reducing the neutron yield.

The energy output distribution in the central cross-layer is shown in Fig. 13.

The target dimension along the proton beam path and, thus, the diameter of the target module can be reduced as the neutron yield is significantly reduced on the opposite side of the beam input. The diameter of the target module in this case can be limited by approximately (1.2–1.3)R (R = range of proton). The remaining part of the neutron source and neutron yield can be assigned to the blanket. This measure will also contribute to increasing the importance of the neutron source and to reducing the volume of the blanket.

Energy Output

Figure 13 clearly shows bursts of energy output in the PbBi modules and heat output reduction in their central portion in the area of the fuel rods with NpO_2.

Energy output distribution in the blanket is as follows: in MOX fuel, fission energy output is 58.2 %; in PbBi modules, it is 41.8 % (by separate ampoules at: 7.2, 7.8, 7.7, 8.4, 5.1, and 5.4 %). PbBi modules located next to the ion guide have a relatively low heat output. Therefore, it is necessary to examine configurations with four or three modules of large diameters, corresponding to replacement of 19 fuel assemblies in the water-cooled part of the blanket, while reducing enrichment in both parts of the blanket. This measure will also contribute to the redistribution of energy output in favor of the PbBi modules.

Reactivity Effects

Reactivity effects as a result of fuel assembly and PbBi module removal and their replacement with water were as follows (Table 4).

FRAs with hafnium shells 1 mm thick were taken from different parts of the blanket for better accuracy. Removal of four FRAs with hafnium magazine shells 0.5 mm thick gave

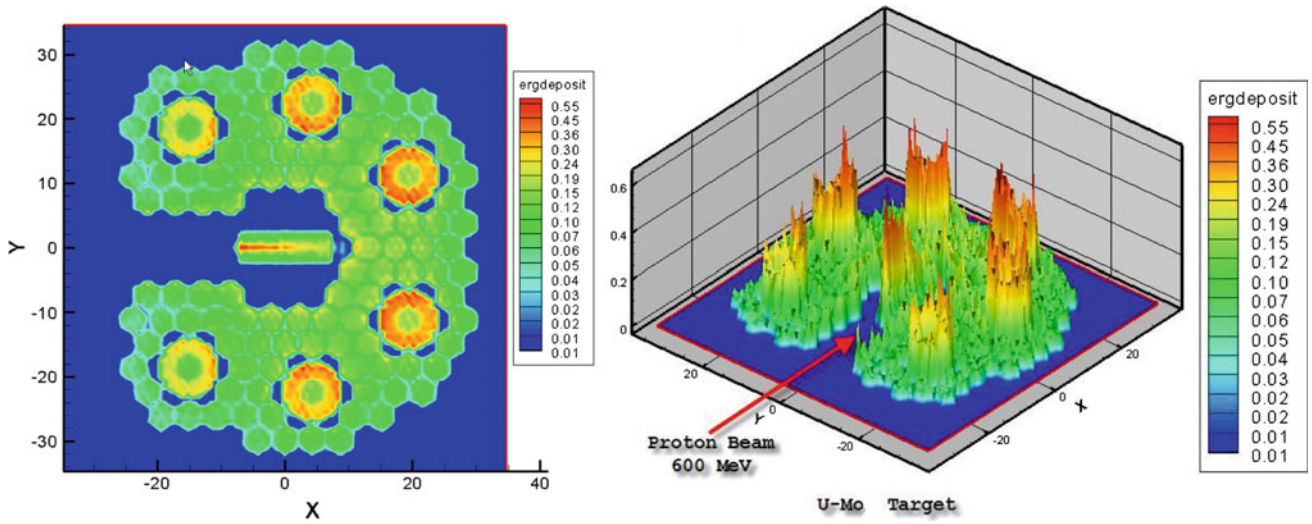

Fig. 13 Calculated distribution of heat over the cross section of the central cross-layer of ADS stand with a uranium target per 600 MeV proton

Table 4 Reactivity effects—Replacement of fuel assemblies and PbBi module with water

Removal of one PbBi module	0.029 k_{eff} (–2.9 %)
Removal of one fuel assembly	0.001 k_{eff} (–0.1 %)
Removal of four fuel assemblies	0.010 k_{eff} (–1.0 %)

a value of –0.004 k_{eff}. During calculations, the Hf density was decreased twice, from 11.4 to 5.7 g/cm^3, at the same thickness. However, in the presence of FRAs with thin (0.5 mm) hafnium shells, k_{eff} increases from 0.95 to 1.013.

If the assembly shells are made of stainless steel (1 mm), the effect of removal of four assemblies becomes positive +0.0057 k_{eff}, and k_{eff} with such assemblies increases from 0.95 to 1.172. This means that, in this case, it is impossible to reassemble the blanket under a layer of water without further measures.

Thus, calculations show that Hf canning of fuel assemblies provides the negative reactivity effects if FRAs and PbBi modules are removed, thus allowing the reassembly of the blanket and assembly of other configurations under a layer of water, as in pool-type thermal reactors.

There are three variable parameters capable of reducing the hafnium volume fraction and for the minimization of the additional neutron capture while keeping the negative effect of reactivity:

- Percentage of hafnium in alloy, for example, Hf(78 %)–Nb(2 %)–Zr(20 %);
- The wall thickness of the cassette, for example, approximately (0.5–1.0 mm);
- The number of fuel rods in the assembly, for example from 19 to 37.

Importance of the Neutron Source

The maximum importance of the initial neutron source obtained during optimization of the ADS stand configuration is $\omega \approx 1.35$.

PbBi versus D_2O

In the course of the examination of physical characteristics, possible replacement of PbBi coolant with D_2O was investigated. For comparison, the base variant was taken (Fig. 9) in which PbBi eutectic in liquid metal modules was replaced with D_2O. Numerical results are shown in Table 5 and in Fig. 14.

From the data shown in Table 2 and Fig. 14, it follows that in the case of replacement of PbBi coolant with D_2O, the spectrum is not significantly changed in the tight grid of fuel assemblies. In the area of neutron energy, $E_n > 0.25$ MeV, with fission of virtually all minor actinides, the spectra in PbBi and D_2O modules are almost the same. The fission ratio between modules and the water-cooled part of the blanket remains almost the same.

It should also be noted that in real reactors cooled by PbBi eutectic, widely spaced fuel rod assemblies are normally used [27–31]; therefore, the average neutron energy in

Table 5 Comparative parameters of ADS stand (base case) with PbBi and D_2O modules

Parameters	Coolant in modules	
	PbBi	D_2O
k_{eff}	0.95423 ± 0.001	0.95024 ± 0.001
Neutrons per fission	2.947	2.947
Number of fissions in modules by:		
Thermal neutrons (E = 0–0.645 eV)	10.50 %	10.58 %
Intermediate neutrons (E = 0.645 eV–100 keV)	42.36 %	44.67 %
Fast neutrons (E > 100 keV)	47.15 %	44.75 %
Energy release:		
MOX fuel (water-cooled part of blanket)	58.2 %	57.69 %
PuO_2 (PbBi and D_2O modules)	41.8 %	42.3 %

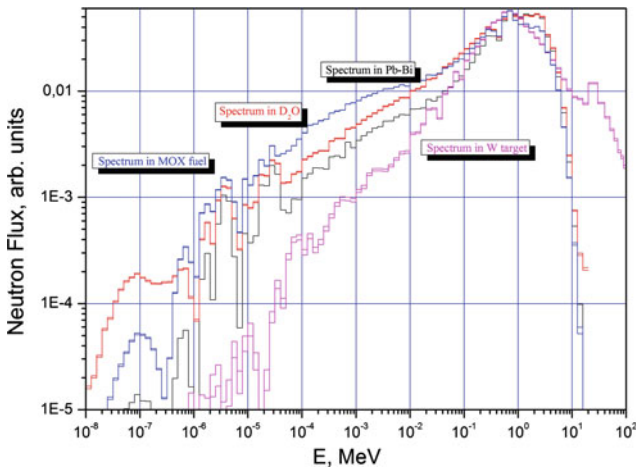

Fig. 14 The neutron spectra in the target in the water-cooled part of the blanket and in the modules cooled by PbBi or D_2O

the spectrum of such a fuel assembly grid will be lower than in the tight lattice with highly enriched fuel. Thus, this article describes the marginal (envelope-case) situation with the fastest spectrum in PbBi modules.

These circumstances mean that:

- In the future, the diameter of PbBi modules will be increased so that, in the case of the widely spaced fuel rods, energy is preferentially output to these modules rather than the water-cooled part of the blanket;
- The ADS stand configuration based on the tight lattice of fuel assemblies cooled by heavy water (or a mixture of D_2O and H_2O) is very promising, especially as the cross section of tritium production will be low due to the fast neutron spectrum.

It should also be noted that in the proposed concept of the ADS stand, all fuel rods inside the PbBi modules can be loaded with minor actinides, despite a relatively small cross section of the stand, thus providing the appropriate level of subcriticality for the stand through increased enrichment of fuel in the water-cooled part of the blanket. However, the water-cooled part of the blanket can be relatively small in volume. For example, the volume of the core in projects on multiplying pulsed neutron sources with PuO_2 fuel is approximately 15 dm^3 [13, 14]. Clearly, in this situation the heat output will be redistributed in favor of the water-cooled part of the blanket. This means that detailed optimization of the stand configuration is required in terms of the maximum minor actinides burnout rate per source proton.

Thus, calculations and previous developments confirm the physical and technical properties included in the concept of the ADS stand and their practical feasibility.

Directions of Further Research

Use of ^{237}Np as a fuel could exclude the necessity to use weapons-grade plutonium and highly enriched uranium and will reduce the neutron background between pulses because of the lower effective fraction of delayed neutrons, β_{eff}, for neptunium when using the ADS stand as a pulsed neutron source.

Use of molten salt modules with dissolved minor actinides (instead of PbBi modules) will be investigated.

Transmutation of minor actinides in the proton beam will be analyzed, too, as the high solubility of MA in melts of LiF–NaF–KF allow us to use it as a target for the proton beam. Operation with very low coefficients of multiplication reduces the equilibrium level of MA in the target and allows burnup of MA to be almost complete.

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A New Concept for ADS Spallation Target: Gravity-Driven Dense Granular Flow Targets

Lei Yang and Wenlong Zhan

Abstract

For Chinese accelerator-driven system (ADS) projects, the main purpose is to dispose of nuclear waste. Spallation targets in the tens of MW range are necessary for ADS to provide enough neutrons to drive the subcritical blanket, which is not an easy task. We propose the concept of the gravity-driven dense granular flow target, where heavy metal grains are chosen in order to produce enough neutrons and the temperature of the grains can be 1000 °C. The heat of the high-power proton beam deposited in the heavy metal grains will be dealt with off-line. Another important aspect is source availability and cost of operation, which is affected by radiation effects, other damages, and the radiotoxicity. The gravity-driven dense granular flow targets have the potential to easily deal with these problems and would be a cost effective candidate for targets in tens of MW range.

Keywords

Accelerator-driven systems • Spallation targets • Dense granular flow

Introduction

Accelerator-driven systems (ADS) are subcritical assemblies driven by proton accelerators through a high-power spallation target. For instance, a proton accelerated to 1 GeV can create several tens of neutrons when impinging on a heavy metal target through spallation reactions, and then these neutrons are injected into the subcritical fission blanket. This is a hybrid system, which can be described as subcritical assemblies of fissionable material activated by a powerful external neutron source. The hybrid systems can be used for:

1. Incinerating long-term radioactive nuclei into stable or short-term nuclei, which is usually called "waste transmutation", although transmutation in general means a change in the nuclear entity [1–3];
2. Transforming non-fissile nuclei, namely U-238 or Th-232, into highly valuable fissile nuclei, Pu-239 or U-233, respectively, (along with the additional use: to produce tritium);
3. Generating energy (mainly through fissions reactions) [4].

For "waste transmutation" an ADS is suitable for the destruction of minor actinide (MA) through transmutation [5]. Because ADS do not require fuels containing uranium, they are more efficient at destroying MA waste. The European Union has taken the ADS as a core strategy for nuclear waste treatment and disposal. The Ministry of Atomic Energy Industry of the Russian Federation launched the ADS Development Plan in 1998. The United States Department of Energy formulated a route map for accelerator-driven nuclear waste transmutation processes in 1999, called the ATW plan. Japan started its OEMGA plan for long-term research and development of final nuclear disposal in October 1988. Furthermore, South Korea and India have made ADS research plans. For an industrial-scale ADS MA burner (Table 1), the reactor would have to be on the order of 500 MW (thermal) to 1500 MW (thermal) per

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Table 1 Parameters of an industrial-scale ADS MA burner [6]

	Industrial-scale transmutation	US-ATW	EU-EFIT
Beam power	10–75 MW	45 MW	16 MW
Beam energy	1–2 GeV	1 GeV	0.8 GeV
Beam time structure	CW	CW	CW
Subcritical blanket thermal power	500–1500 MW	850 MW	400 MW

burner [5]. To minimize capital costs, several such burners could be driven by a single accelerator, as envisioned in the US ATW plan, where a 45 MW (1 GeV@45 mA) beam would drive four 850 MW burners. In the European ADS Roadmap, a single 16–50 MW accelerator driving a single large burner, the thermal power of which ranges from 400–200 MW, is contemplated.

The aim of the Chinese-ADS project is to dispose of nuclear waste. Physical and technological foundation research on ADS as part of the Key Basic Research Development Plan (973 Plan) was implemented in 1999 by the collaboration of the China National Nuclear Corporation and Chinese Academy of Sciences. Based on several rounds of high-level expert consultation and assessment from 2009–2010, the "Future advanced fission energy-ADS transmutation system" was approved as a strategic leading science and technology program in January 2011 to speed up ADS R&D. Owing to the considerations of the accumulated growth of nuclear waste in China, the roadmap of ADS technology in response to nuclear energy development in the future has to be discussed. The development of ADS should have several phases. The first phase is principle verification. In this phase, the aims are to solve key technological problems of ADS units, to realize small-scale system integration, and to develop the design system; the key technologies of accelerators, spallation targets, and subcritical cores have to be applied to an industrial facility. The second phase is demonstration of facility building. The aims here are to implement medium-sized technological integration, build and run $\sim 100\ MW_{th}$ ADS driven by a 25 MW accelerator beam power and start transmutation experiments. The third phase is the industrial facility and full-sized technological integration. During this phase, a ~ 1000 MW system driven by full-energy low-intensity 50 MW beam power will be built and run and its operational reliability will be verified (Table 2).

To achieve the objectives of the ADS program, high-power proton beams are needed to bombard the spallation target in order to generate neutrons driving the subcritical cores. For the tens of MW beam power required for the industrial-scale ADS concepts, one of the most difficult challenges is the high-power spallation target as currently only MW-class spallation target exist at the Spallation Neutron Source (SNS).

Challenges of ADS High-Power Targets and the New Concept

In the early days of spallation target development, 1980s, several solid targets at beam power levels of several hundred kW were in operation, where the material of the targets was solid U, Ta, or W. However, in these solid targets, heat removal is limited by the heat conduction of the target material and convection cooling; thus, increasing the target power will ultimately reach a limit. If the heat deposited could be dealt with off-line, heat removal from the target could exceed the limit of the solid targets. Based on this concept, heavy metal liquid targets with a beam window have been designed and operated at ~ 1 MW for the MEGAPIE and SNS projects. The MEGAPIE project was started in about 2000 to design, build, and operate a liquid metal target as a key experiment on the road to experimental ADS [3, 5].

The MEGAPIE project and research on heavy metal liquid (HML) targets show the challenges of the design. Heat removal from the lead–bismuth eutectic (LBE) window target would be limited by corrosion and erosion of the LBE material, which usually limits the temperature to less than 550 °C and the velocity to less than 2 m/s. The potential damage to the window structure can arise from cavitation and shock waves of the HML. The MEGAPIE design is very close to the acceptable limits for these effects. The chemical and radiotoxicity of the LBE target made the operation, safety, decommission, gas control, etc., difficult and there

Table 2 Parameters of the Chinese-ADS

	Initial facility	Demo facility	Industrial facility
Beam power	5 MW	25 MW	50 MW
Beam energy	0.35 GeV	1.5 GeV	2 GeV
Beam time structure	CW	CW	CW
Subcritical blanket thermal power	10 MW	~ 100 MW	~ 1000 MW

have been incidents with the lead–bismuth loop in the intermediate circuit during operation.

For ADS, the spallation targets have to use compact targets to provide high-flux beams; the crucial factors are power deposition, heat shock, radiation damage, and so on. Thus, the first issue for a high-power target is how to remove the heat from a proton beam of high current density. The second issue is that the lifetime of the target is limited by the radiation damage, heat shock, corrosion, etc. Thus, the stability and safety of operation, the cost of decommission, and the complexity of the system will be the challenges to overcome to implement a high-power spallation target. Nowadays, new concepts are becoming the focus of more research activity for high-power targets, including the concepts of windowless heavy metal liquid targets, large-scale rotating solid targets, fluidized bed solid targets, and so on, although these concepts would be not applied for ADS.

Here, we propose a new concept for high-power spallation targets: the gravity-driven dense granular target (DGT), which has a compact structure and the potential for the high-power operation. The target material is a collection of a large number of discrete solid particles, namely the granular materials, which can show both semi-solid and semi-fluid behavior. For example, the granular flow driven by gravity shows the semi-fluid behavior of the granular materials. The science and technology of granular flow has a long history, for example, the hourglass is a typical application of gravity-driven dense granular flows and is believed to have originated in medieval Europe. In an hourglass, the domed interior contains sand, and time is proportional to the amount of outflow as determined by the design of the geometry.

For a gravity-driven DGT (Fig. 1), the heavy metal grains flow into the container under gravity, where the proton beam interacts with the grains. If the geometry of the container is suitably designed, the flux of outflow could be constant. The heat of the high-power proton beam deposited in the grains will be brought out of the container via the outlet by the dense granular flow. Furthermore, the issues of the conventional approaches to spallation targets are neatly sidestepped by allowing the granular particles to be damaged and then replacing them on-line.

The granular particles in a DGT would run like fluids and the choice of materials for the discrete solid particles is wide. Compared with HML targets, where only several possible metals could be used, the DGT has the potential to maximize benefits through particle selection; possible factors to consider include: high neutron yield and low radiotoxicity for neutronic optimization, high specific capacity and thermal conductivity for potential higher temperature difference, low chemical toxicity, and low corrosion for convenient operation.

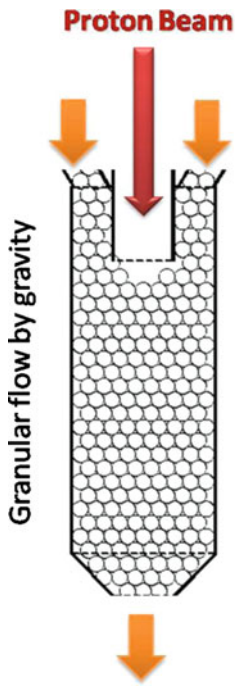

Fig. 1 Concept of the gravity-driven dense granular target (DGT); the geometry of the container needs to be designed so as to provide a constant flux of granular outflow

In the HML target concept, non-fissioning elements usually can be used in the spallation target for high neutron yields; these include tungsten, tantalum, lead, bismuth, mercury, osmium, and iridium. As precious metals are rare, naturally occurring metallic elements of high economic value, they would not be used in targets; this rules out osmium and iridium. Of the remaining elements, tungsten has the highest volume heat capacity and melting point. Thus, tungsten grains could remove the higher deposited power from the beam–target interaction region.

In general, the advantages foreseen for the gravity-driven DGT include:

1. Behavior of the DGT is analogous to the windowless HML target; high-power deposited heat would be removed off-line, the same as with a liquid; the granules will flow away from the interaction region and will be cooled externally using heat exchangers. Tungsten granules have the possibility to withstand very high power proton beams before cooling.
2. In the granular flow, the beam-induced stress waves are contained within each separate grain of material, and cannot generate splashing, cavitations, shock waves, and other hydrodynamical problems.
3. The target material is continuously reformed, and the grains can be replenished and disposed of off-line; the DGT sidesteps corrosion problems and can avoid

chemical toxicity and reduce radiotoxicity through suitable choice of material.

4. Granular engineering is a mature technology developed for the conveyance of granules in the chemical, materials, food processing, and even nuclear industries. Such experience can be exploited in the design of various components for the development of a complete target system.

For the DGT system (Fig. 2), the proton beam will be transported by high-energy beam transport and a beam pipe line to the target to interact with the tungsten particles in the spallation region, which is formed of a silo. The proton beam interacts with the tungsten grains that are confined between walls of the silo. The dense granular flow in the silo is driven by gravity, and the grains discharge from the spallation region through the bottom of the silo. The circuit for continuous grain supply includes a heat exchanger, grain filter, grain elevator, grain storage, and a cover gas system. This design configuration is compatible with an axisymmetric blanket.

A small-scale test system of the DGT has been set up; the scale ratio is 1:5, the particles are a tungsten alloy with a spherical diameter of 1 mm, the diameter of the cylindrical chamber is 56 mm. Figure 3 shows a photograph of the test system, which operates with a total of 1200 kg tungsten alloy grains. The tungsten grains are conveyed by a pipe chain lift from a hopper at the bottom to the top of the system through a vertical 2.8 m high pipe with a diameter of 150 mm. The grains fill the chamber, which has the proton beam pipe located in the middle. The coaxial flow of granules is driven by gravity, and the mouth of the discharge nozzle directs the particle flow into a heat exchanger. Automatic control of the circuit is achieved by using a combination of timed processes and feedback from sensors, for example, a load cell to measure the mass of granules in the bottom hopper.

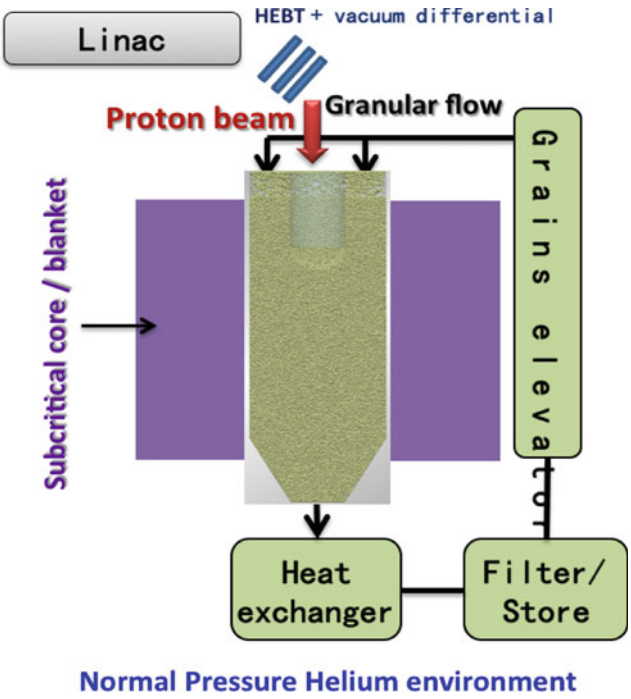

Fig. 2 Schematic outline of a gravity-driven dense granular target with: grain injection and discharge nozzle; grain loading and spallation region; proton beam pipe line; receiver hopper and heat exchanger; grain receiver vessel with filter; grain lift system, and cover gas environment

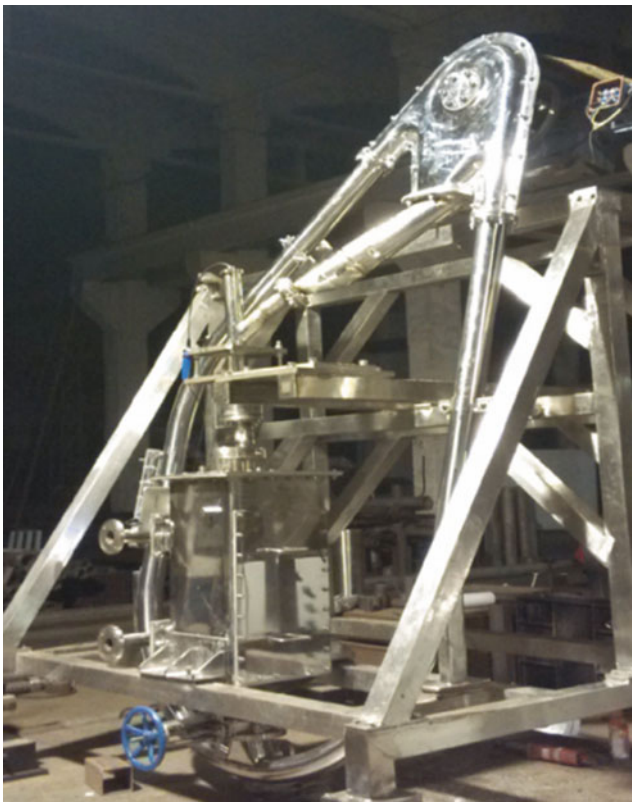

Fig. 3 A small-scale test system of the DGT, including the beam interaction chamber, the heat exchanger, the pipe chain elevator and so on

Discussion and Conclusion

The gravity-driven dense granular target could avoid limitations that exist for purely solid or liquid targets, but naturally they pose new challenges. For example, erosion is almost certainly going to be the key technical issue, particularly in pipes, nozzles, the chamber, and in receiver hoppers. Recently, erosion tests in the room temperature to 10,000 °C range have shown that tungsten alloy granules made of a SiC/tungsten alloy are excellent in terms of wear resistance with a maximum wear amount of $\ll 1$ mm/year. Other

subsystems need to be studied in detail, although some of the components have been used in similar cases, such as the beam entry window used in water-cooled systems, the countercurrent water corrugated plate heat exchanger, and the granular elevators. According to international standards, the NE series hoist is suitable for conveying powder, granular, and small blocks of non-abrasive and abrasive materials. Because the traction hoist is a ring chain, it allows delivery of high-temperature materials up to 600 °C. A general transport height up to 40 m can be achieved with the NE series hoist; however, the TG type hoist can lift up to 80 m.

It has been demonstrated by microscope simulations and small-scale loops that tungsten grains can be readily driven by gravity to form a dense and stable granular flow within a chamber. In general, the advantages foreseen for the gravity-driven dense granular target include the very high heat-removal possibility, long operation life, low chemical and radiotoxicity, and other modest system parameters. The configuration of the DGT has the potential to form the basis of a spallation target in the tens of MW region for a simple ADS target facility.

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Accelerator-Driven Systems for Thorium Utilization in India

S.B. Degweker, Pitamber Singh, P. Satyamurthy, and Amar Sinha

Abstract

Research on accelerator-driven systems (ADSs) is being carried out around the world with the objectives of waste transmutation and power production. Over the last few years, the Bhabha Atomic Research Centre (BARC) has been pursuing research in the area of ADS with the primary objective of thorium utilization. ADSs can play an important role in augmenting India's efforts for the development of nuclear power by way of the three-stage program utilizing indigenous resources. In this article, the research and development efforts in this area at BARC are outlined along with some schemes being studied for thorium utilization in ADSs.

Keywords

Accelerator-driven systems • Th–U cycle • Reactor noise • Monte carlo simulations

Introduction

Over the last few years, the Bhabha Atomic Research Centre (BARC) has been pursuing a research program aimed towards developing accelerator-driven systems (ADSs). Such systems were first proposed for fissile material production [1–3] and later for waste transmutation [4] and would have required very high power (energy ~1 GeV; current–few hundred mA) proton accelerators. The current interest in ADSs was spurred after the proposals by Carlo Rubbia et al. for energy production utilizing Th fuel [5, 6], which required relatively modest power accelerators (approximately 1 GeV–10 mA currents), but ADSs have caught the attention of the world for an equally important role—that of nuclear waste transmutation. It is well known that nuclear reactors generate radioactive waste, which retains its radiotoxicity for hundreds of thousands of years, and disposal of this waste has been a major source of public concern. It is expected that a few ADSs can safely transmute the waste produced by the currently operating critical reactors, while producing useful power.

Indian interest in ADSs is primarily related to the planned utilization of our large thorium reserves for future nuclear energy generation. Thorium has the added advantage that it produces much lower quantities of long-lived radioactive wastes compared with uranium. However, thorium by itself is not fissile and must be first converted to fissile ^{233}U by neutron irradiation, a process called breeding. Considering the importance that the thorium fuel cycle is expected to play in the Indian nuclear power program, we emphasize this role of ADSs in the present article. We present a brief description of the on-going research and development efforts in this area at BARC and some schemes studied by us for Th utilization in ADSs.

Overview of the Program for ADS Development

Accelerator Development

The program is oriented towards development of a high-current, high-energy proton accelerator. The targeted energy and current are 1 GeV and 30 mA, respectively. The

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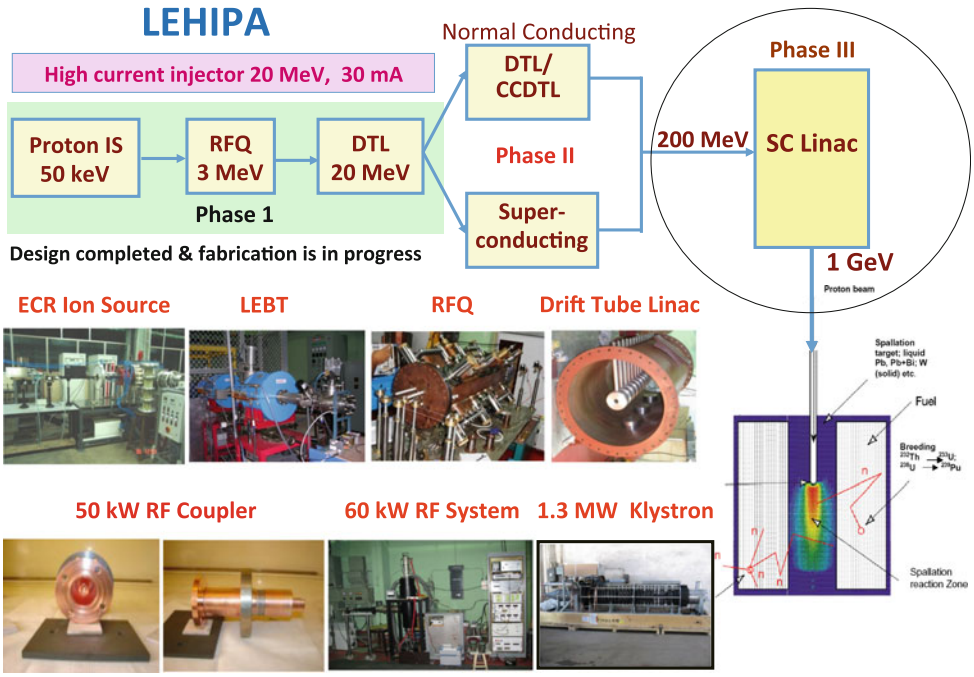

Fig. 1 Schematic of the proposed accelerator for ADS and some of the components developed

first phase consists of the development of a 30 mA, 20 MeV linear accelerator (linac) injector (LEHIPA). The second phase involves taking the beam to an energy of 1 GeV by way of a superconducting linac. Presently, the first stage is under development and several components, such as ion source, low energy beam transmission line, and drift tube linac, have been built. A radio frequency quadruple (RFQ) of 400 keV has been built and tested and this has given confidence for the planned RFQ of 3 MeV. The RF power supply will be through two 1.3 MeV klystrons, which will be procured for this purpose. The general scheme of the proposed accelerator for ADS, and some of the components developed, is shown in Fig. 1.

A major program for augmentation of the infrastructure required for development of a 1 GeV linac has been launched. This includes developments in accelerator physics and facilities for developing spoke resonators, superconducting cavities, helium vessels and tuners for superconducting radio-frequency (SCRF) cavities, vertical and horizontal test stands, cryo-modules for SCRF cavities, high-power solid-state RF amplifiers, low-level RF systems, RF protection systems and related instrumentation, beam diagnostics, monitoring and protection systems, cryo-module test stands (CMTS), and necessary infrastructure for SCRF cavity fabrication, processing, and testing. This program is being carried out at various laboratories in India.

Target Development

Considering the high heat density deposition by the proton beam (approximately a few kW/cm^3) and the high radiation damage ($\sim$100 dpa/year or more) that causes embrittlement, the use of a circulating liquid target is an attractive option. Among the heavy metals, Hg is not suitable owing to low its boiling point and, hence, Pb or lead–bismuth eutectic (LBE) seem to be most suitable target materials. The target development research includes studies related to heat removal and window damage such as irradiation creep, and void swelling due to hydrogen and helium generation and transmutation.

An experimental Hg loop has been in existence for several years and is being used to develop diagnostic techniques such as loops and an experimental target velocity field mapping by ultrasonic velocity profile (UVP) monitoring, carry-under studies, two-phase flow studies by gamma rays, laser-triangulation for free-surface measurement, computational fluid dynamics (CFD) code validation, and gas-driven flow studies. Another LBE thermal-hydraulic experimental test facility (loop) has been designed and is under fabrication (Fig. 2). Its thermal-hydraulic parameters are chosen to be suitable to drive a $\sim$100 MW ADS. Thermal hydraulics code validation, primary coolant development, corrosion mitigation studies, component development, diagnostics, and C&I development, will be carried out in this loop.

A target has been designed and is being developed for irradiation in the proton beam of the medical cyclotron at Calcutta. Presently, civil works and beam line procurement are in progress. A prototype target is being installed and is shown in Fig. 2. The facility will be used for studying the coupling of the beam with the target module, window heat extraction, radioactivity issues, gas handling, irradiation studies, combined control and instrumentation, remote operation, and remote dismantling.

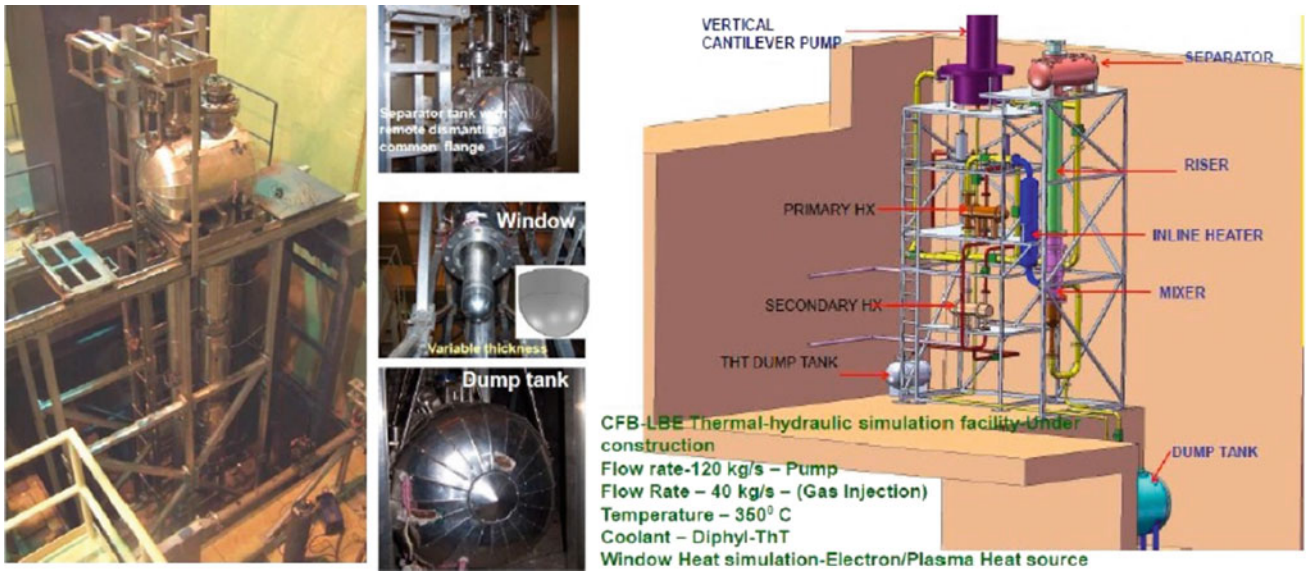

Fig. 2 *Left* Various components of the target to be irradiated in the medical cyclotron, VECC, Calcutta. *Right* LBE loop coming up at Trombay

Reactor Physics Studies and Developments

Computer Codes and Nuclear Data

A program to induct new code systems was developed as part of the ADS project at BARC. For the interactions of high-energy protons (>20 MeV) with the target, the codes FLUKA [7] and a recent version of CASCADE [8] are used. For neutron energies below 20 MeV, we have developed a transport theory and Monte Carlo-based codes to perform static and burnup calculations [9]. Efforts to develop a comprehensive kinetics capability, including calculation of alpha modes, natural modes, analysis of experiments for subcriticality measurements, treatment of feedback effects, and definition and calculation of kinetics parameters, are also underway.

We have also upgraded our database for reactor physics with the nuclear cross-section data for about 300 nuclides, which includes almost all actinides, fission products, coolant and structural materials of interest to ADSs, fission product yields, and decay data for burnup, and delayed neutron yields and spectra for fission in various actinides for kinetics studies. We have also upgraded our transport theory data to include the recently prepared WIMS 172 group library [10] based on ENDF-6 files.

Experimental Program: The Purnima Subcritical Facility

A subcritical facility for carrying out basic reactor physics experiments by using an existing 14 MeV neutron source at Purnima labs, Trombay, has been set up. The neutron generator has been upgraded to give a yield of 10^{10} neutrons per second, with a tritium target. A facility for pulsing the beam has been added. The subcritical reactor assembly consists of natural uranium metal rods moderated by high-density polyethylene (HDP) and reflected by BeO. The k_{eff} of the assemblies is estimated to be about 0.89, which is also confirmed by preliminary measurements. The experiments planned include measurement of flux and spectrum, neutron multiplication and power, and pulsed neutron and noise studies for subcriticality measurement. Figure 3 shows a photograph of the system.

Theoretical Studies Related to the Experimental Program

Theoretical studies in support of ADS-related physics experiments have been initiated. These include the development of a new theory of noise in ADSs [11], a method for calculating the fundamental and higher alpha modes [12], and development of a noise simulator [13] to aid the planning and interpretation of these experiments. Some of these activities are discussed briefly in this section.

Measurement and monitoring the subcriticality of an ADS will be an important and essential safety requirement. Pulsed neutron, source jerk, and noise methods have been proposed and experiments have been conducted around the world to test these methods. Reactor noise in ADS is different from that in critical reactors and requires a new theoretical framework for interpreting noise experiments. This is because traditional reactors have neutron sources from

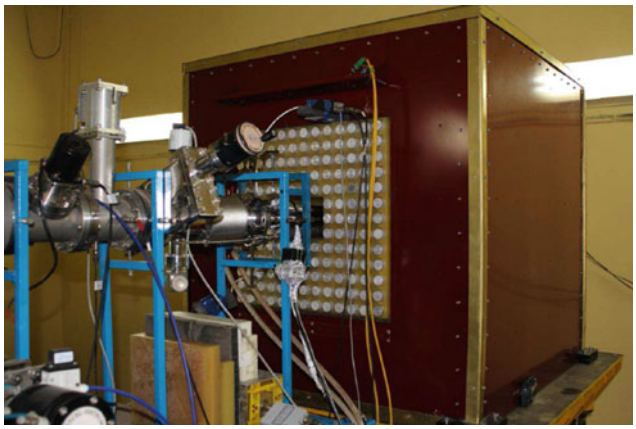

Fig. 3 Experimental subcritical facility at Purnima laboratory, Trombay

radioactive decay and are Poisson in character. Accelerator sources are different on account of periodic pulsing, fluctuations in intensity (typically a few percent), and correlations in these fluctuations. For these reasons, accelerator sources cannot be assumed to be Poisson sources. A new theory of noise in ADS, taking into account these differences, has been developed by us [11] at BARC. Available experimental evidence seems to support the theory and it is now generally accepted as the theory of ADS reactor noise [14].

As part of the planning of the experiments, simulation of the kind of results that might be expected with different detector locations and counting and analyzing setups is necessary, particularly in view of the fact that the k_{eff} is rather low and higher modal effects will be large. Simulations with standard code packages are not appropriate because of several non-analogue features built into such codes. An analogue Monte Carlo (MC) simulation can give information about the magnitude of, for example, the variance-to-mean ratio for given values of system parameters such as k_{eff}, detection efficiency, etc., and how it compares with the background random noise for a given counting time or the magnitude of the space-dependent effects, delayed neutron effects, and dead time effects. This allows us to assess the feasibility and accuracy of the proposed subcriticality measurement. As the usual analogue MC simulation takes a very long computing time, we have developed a diffusion-based analogue Monte Carlo code [13] for carrying out quick simulations. We have also developed a transport-theory-based analogue Monte Carlo simulator for obtaining more accurate results [15].

Simulation of pulsed neutron and noise experiments using these tools has been carried out. Both types of experiments will benefit from placing the detectors at the zeros of the first symmetric harmonics. The main difference between the detector configurations in pulsed neutron and noise experiments is that it is necessary to have detectors located at all eight intersections of the zeros of the first three symmetric harmonics in the noise experiment, (whereas a single detector placed at any of the zeros is adequate for the pulsed experiment) and the combined output of all detectors is used for the analysis.

Thorium Utilization in Accelerator-Driven Systems

The three-stage program, described in greater detail elsewhere (for example, in [16]), represents the standard route to Th utilization in the Indian context. ADSs could serve to augment our efforts in this direction and present alternative scenarios. The CERN energy amplifier (EA) [5, 6] is an ADS concept for power production using Th in a self-sustaining cycle. One of our earliest studies on ADSs examined the possibility of reducing the required accelerator current (power), while maintaining a safe level of subcriticality. The answer was the one-way coupled fast thermal ADS [17], which permitted operation of such an ADS at much reduced accelerator currents.

We will, however, describe other schemes being studied by us for breeding ^{233}U in the ADS, which are more interesting in the Indian context, rather than the self-sustaining Th–U cycle. All these schemes would use some fissile seed fuel at the initial stage so that they produce power from day one. Even so, the first and second schemes are basically for power production, using Th fuel in ADS mode, whereas the third scheme is mainly for breeding. Which of these modes would be used will depend upon economics as well as technical issues such as the reliability of the accelerators to function continuously for long periods for the purpose of power production. As all these schemes are in the deep subcritical region, the power peaking problem is severe for solid-fueled reactors. Hence, we also examine molten salt reactors (MSR), which may also have the advantages of low fissile inventory and excellent neutron economy.

Traditionally, accelerator breeder concepts [1–3] have relied on the use of (solid) U or Th targets because of the higher neutron yield per proton in these materials—some neutrons are absorbed in the target, leading to fissile material production—and also because a considerable amount of energy is generated in these targets due to fissions, which can be used to produce part of the power needed to drive the accelerator. However, such targets pose severe problems with regard to heat removal and radiation damage and in recent years there has been greater interest in the use of liquid Pb or LBE targets [5, 6] owing to the ease of heat removal and because they pose no structural damage issues. In the following discussion we assume such targets.

The Power-Producing Breeder ADS

In this scheme, the ADS is fueled with a mixture of ^{233}U and Th and requires part of the ^{233}U to be recycled back into the subcritical core to maintain the k_{eff}, while the excess ^{233}U that has been bred is utilized in other (critical) reactors. The fast Th–U cycle can be used in a critical system, but the breeding can be improved in an ADS. The primary difference in breeding rate in an ADS over that in a critical reactor is due to the externally produced neutrons. In fact, it is simple to show that the difference is exactly equal to the external neutron source strength. Additional factors that account for the higher breeding in an ADS include the absence of control rods, which are necessary to maintain criticality in traditional breeder reactors. Conversely, the doubling time depends upon the specific power, which is lower in an ADS of low k_{eff} because of greater power peaking. We are studying a system driven by a higher power accelerator (the 30 MW linac described earlier), with a view to keeping a lower k_{eff}, taking these factors into account.

In Situ Breeding and Burning in Once-Through Cycles: The Thorium Burner

An interesting alternative to breeding ^{233}U in ADS reactors for use in critical reactors is to breed and burn in the same (ADS) reactor. Such a system does not need costly reprocessing and refabrication of the Th fuel and in some versions can completely avoid reprocessing or enrichment at any stage. The idea of breeding and burning has been studied in different forms, for example, the Linear-Accelerator-Driven Reactor (LADR) [18] concept, in which the fuel is burnt without reprocessing in a sub-critical system. The Traveling Wave Reactor (TWR) [19] under study by several groups is another example of such systems run in critical mode.

We have proposed and studied such accelerator-driven systems [20, 21]. The concept is illustrated in Fig. 4. The main fuel is Th, which is fed continuously into the system. The system is maintained at a k_{eff} in the range 0.9–0.95 as a result of the ^{233}U bred internally. The system is driven by an accelerator of a suitable current to produce the required power. After recycling, a fraction of the power is fed back to the accelerator, and the balance is exported to the grid. The fuel is discharged after high burnup and can be sent for disposal (Th produces miniscule quantities of long-lived minor actinides compared with U), or in case it is decided to recover the ^{233}U, for intermediate storage. In the initial phase of operation there is no internally bred ^{233}U and a seed fuel is added to produce k_{eff} in the range 0.9–0.95.

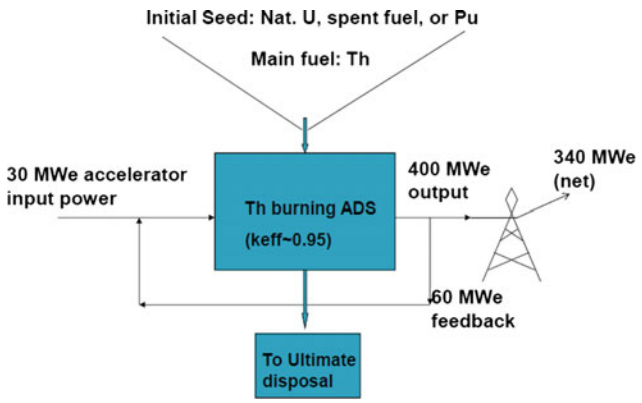

Fig. 4 Schematic of the Th burner concept

Heavy Water Moderated Thorium Burning ADS

A pressurized heavy water reactor (PHWR) ADS is operated in a once-through Th cycle. This process starts with a core made up from both natural U and Th bundles. The reactor is continuously refueled as usual with natural U but the Th bundles are allowed to reside longer. During operation of the reactor, ^{233}U will be generated and this adds reactivity to the system. The additional reactivity can be compensated by replacing some U by Th. As a result, the amount of Th in the system will increase progressively, whereas the U inventory will decrease. Ultimately, a situation will be reached where the core will consist of only Th and no natural U. Fresh Th bundles are loaded in a blanket surrounding the core before they are introduced into the power-producing core region. Consequently, neutrons leaking from the core will be captured in these bundles and, hence, it can be assumed that the loss on account of leakage is practically zero.

Such a subcritical system would have several advantages, such as not requiring enrichment or reprocessing at any stage, a high burnup (about 10 %) of the Th fuel, and it also acts as a breeder for ^{233}U. The consumption of natural U is a small fraction of that in a purely uranium-fueled reactor. However, this system has a low k_{eff} of about 0.9 and a gain of less than 20, thereby requiring an accelerator power of 30 MW (1000 MeV, 30 mA protons) for a 200 MW_e ADS. About one third of this power has to be fed back into the accelerator, assuming an efficiency of 50 %. Should the efficiency be lower, a larger fraction has to be fed back. The required exposure is also much higher than the best achieved to date in water-cooled reactors.

Thorium Burning Fast ADS

The fast system has some inherent advantages over thermal systems, such as better neutron economy owing to lower parasitic capture in fission products and higher values of the product $\eta\varepsilon$. Thus, one might expect that a fast Th–U system

might very easily achieve a k_{eff} equal to 0.95. However, the principal disadvantage of a fast system is that the equilibrium ratio of U:Th is about 0.1, so the system loses a much larger quantity of fissile material when a fuel assembly is discharged. An equal amount has to be made up by breeding. This disadvantage almost offsets the advantage of better neutron economy in a fast system.

Our calculations [21] suggest that it is possible to get a relatively higher value of k_{eff}, equal to that of about 0.95 for metallic fuel. Parasitic absorption (other than by structural material (stainless steel), coolant (Na or Pb), and fission products, which is accounted for in the calculations) must be minimal, and neutron loss on account of leakage should be almost zero. This is possible by completely surrounding the core with a Th breeder blanket. Good shuffling of fuel elements will be necessary and nearly continuous fueling may be required. The fast systems will require an initial charge of fissile material, such as Pu, to kick start the system and a fuel design that can withstand burnups of 400 GWd/tonne. This is more than two times the best obtained so far in fast systems.

Accelerator Production of ^{233}U for Use in Critical Reactors

Consider the irradiation of a Th blanket with neutrons from a spallation source. As irradiation proceeds, the fissile (^{233}U) fraction increases and, hence, the fission rate of the contained ^{233}U also increases. The k_{eff} of the blanket, and consequently, the source multiplication in it also increase. One consequence of increased source multiplication is an increase in the neutron flux and a consequent increase in the rate of fissile material production. Another consequence is that more and more of the fissile product starts burning, which reduces the rate of fissile ^{233}U production. As the $\eta\varepsilon$ value for U–Th fuels is substantially greater than 2, the former effect is generally speaking the greater of the two. Burning, however, also causes an increase in the fission product concentrations, thereby increasing losses due to parasitic capture.

These effects are illustrated in Fig. 5, which shows the variation of different parameters [22] with exposure time at discharge for thermal and fast Th blankets driven by a 1 GeV, 30 mA proton beam on Pb. For both systems, the rate of ^{233}U production shows a slow and small rise until a peak is reached and the rate falls off thereafter. The fast system has a somewhat higher peak breeding rate owing to the higher value of $\eta\varepsilon$ and lower parasitic capture in fission products. The k_{eff} value increases almost linearly with exposure, and saturates thereafter at about 0.9. Similarly, the ^{233}U fraction in the fuel first increases linearly and rapidly flattens off at a value of about 1.4 % for the thermal system, and at about 10 % for the fast system. The power and flux follow an S-type curve and, unlike k_{eff} and the fissile fraction, the saturation is much slower. In both systems, it is seen that by irradiating the Th for sufficiently long, it is possible to attain close to the maximum fissile fraction and produce at least as much power as is required to run the accelerator (assuming 30–50 % efficiency) without significant reduction in the fissile ^{233}U production rate. The fast system produces considerably higher power, which can be fed to the grid.

Thorium Utilization in Molten Salt Reactors

The Monte Carlo burnup code, MCBurn, has been modified for application for molten salt reactors by taking into account the on-line addition of fissile/fertile material and the removal of fission products or actinides, such as Pa, by adding necessary source terms and (non-radioactive) decay constants. The source terms are adjusted at the beginning of each burnup step to maintain criticality (or a prescribed value of k_s in the case of ADS). This is somewhat tricky and a method has been developed to ensure that the system does not depart significantly from criticality or the targeted value of k_s. In molten salt reactors, mixing of the fuel salt takes place over time scales much shorter than the burnup and, hence, the salt concentration can be assumed to be uniform throughout the reactor at any given time and one burnup zone suffices (two burnup zones for two salt reactors) even though the reactor is neutronically heterogeneous. This results in good statistical precision in the reaction rate tallies used to obtain effective cross sections.

The code was used to study the three MSR systems shown in Fig. 6, [23]. The first is a single-salt thermal critical reactor of 2500 MW_{th} power, which is a variant [24] of the molten salt breeder reactor (MSBR) designed by the Oak Ridge National Laboratory (ORNL) group [25]. The second is a 1000 MW_{th} two-salt fast reactor, whereas the third is the same as the first but operated as a subcritical reactor at a k_{eff} of 0.96. The first shows a small breeding ratio of 1.04 over 50 years, whereas the second shows a much higher breeding ratio of 1.14 over 30 years. However, the doubling time is about the same (about 35 years) as the specific power in the fast system is lower. Compared with the critical thermal reactor, the thermal ADS shows a much higher breeding ratio, 1.14, at a k_{eff} of 0.96. If it is driven by a 30 MW proton accelerator, it can produce a power output of 1500 MW_{th} and give a linear doubling time of 15 years or an exponential doubling time of a little over 10 years.

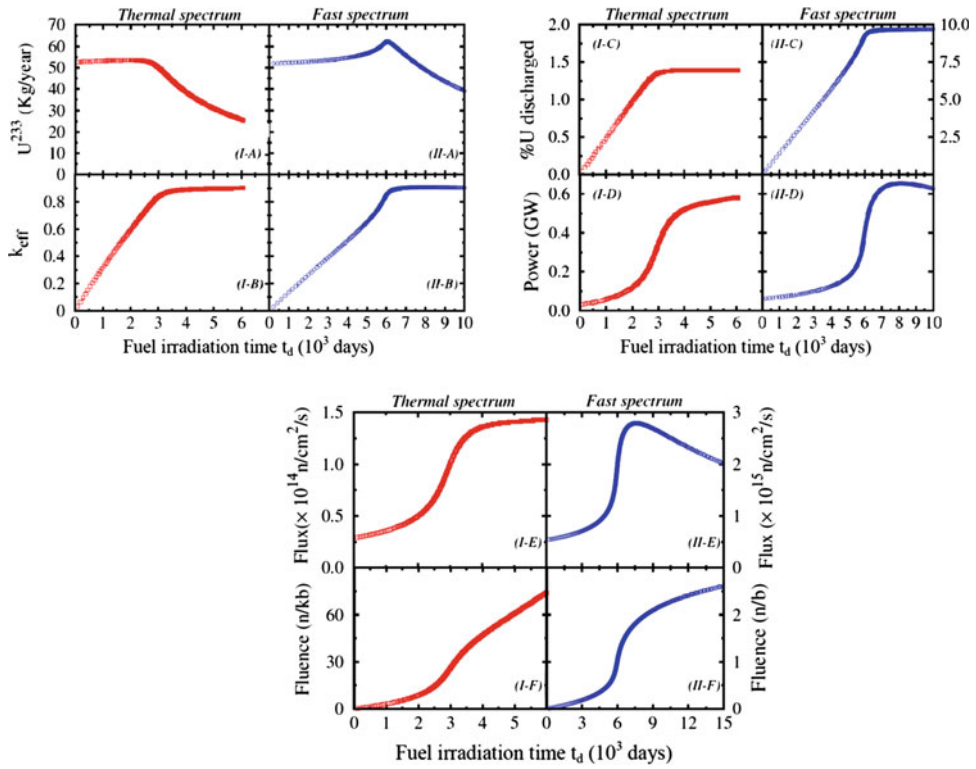

Fig. 5 Variation of ^{233}U production rate, k_{eff}, percentage of ^{233}U in discharged fuel, reactor power, neutron flux and fluence as a function of fuel irradiation time in thermal (*red*) and fast (*blue*) systems

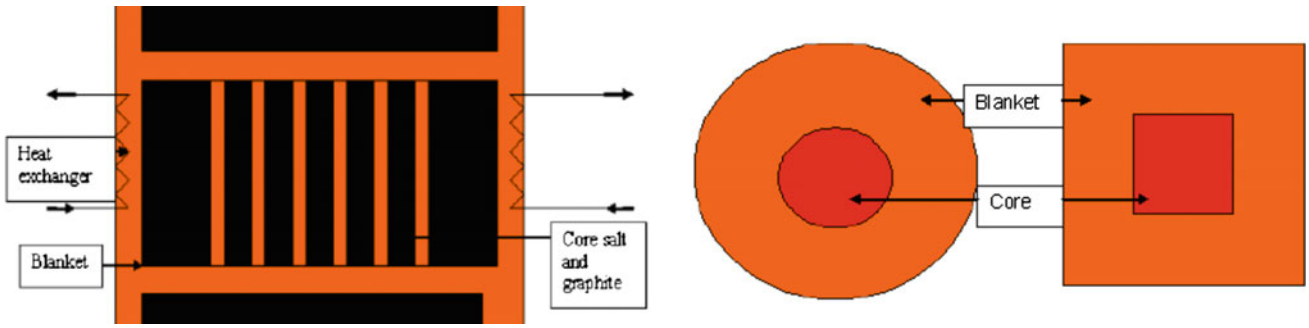

Fig. 6 The thermal (*left*) and fast (*right*) MSR systems studied as critical reactors and subcritical ADSs

Conclusion

ADSs show considerable advantages over traditional critical reactors for transmutation of waste and Th utilization and are, hence, gaining interest around the world. BARC has initiated a program for the development of ADSs in India primarily for the purpose of Th utilization. We have briefly described this program and some of the routes to Th utilization in the ADSs being studied by us viz., power production with recycling of ^{233}U in a breeding cycle; ADS reactors fueled with fertile Th to produce fissile ^{233}U for use in critical reactors; in situ breeding and burning of Th fuel in fast or thermal ADS reactors with a fissile (seed) fuel only in the initial stage; and the use of molten salt reactors in subcritical ADS mode.

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iThEC's Approach Toward Nuclear Waste Transmutation and Energy Production with Thorium

Yacine Kadi

Abstract

The international Thorium Energy Committee (iThEC) was recently created in Geneva to promote thorium-fueled accelerator-driven systems. iThEC was initiated by members of the CERN scientific community under the auspices of Carlo Rubbia, the inventor of the ADS system he refers to as the Energy Amplifier (EA). The committee also comprises business leaders, politicians, and members acquainted with public relations, in an effort aimed at broadening the support for the ADS concept. Nuclear policy today in Europe and more particularly in Switzerland has reached a point where crucial decisions must be made, affecting the security of electricity supply, the fulfillment of commitments to the Kyoto protocol for combating climate change, while addressing public concerns that increased following the Fukushima accident. Nuclear power has its advantages, yet the perennial problem of what to do with nuclear waste, the perceived risks due to criticality, and the fear of nuclear weapon proliferation have long been a thorn in the side of nuclear power, preventing it from taking its place as a core carbon-free technology of the 21st century and beyond. In addition, policy makers are generally ill-informed about alternative nuclear energy systems, such as the possibility of ADS using thorium fuel. The main purpose of iThEC is to work on reversing the current negative trends in Europe, by providing better information to the public on thorium possibilities, by supporting thorium ADS technology developments and by actively engaging the scientific, political, and business leadership. iThEC is preparing a proposal for the construction of an ADS demonstrator.

Introduction

Providing energy to a growing and increasingly prosperous world is the greatest challenge facing humanity today, one which will largely determine its fate. The rise in numbers and in wealth of the world's population are inexorable over the century to come. World energy consumption will have to triple in order to provide humanity at the end of the 21st century with a consumption level comparable to that of today's European population; an inevitable rise despite increased energy efficiency.

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The international Thorium Energy Committee (iThEC), created in late 2012 as a non-profit organization, is hosting at CERN the current ThEC13 international conference on thorium-based nuclear energy production. The goal of the new organization is to promote thorium as an alternative fuel and more generally to advance the cause of a sustainable energy solution by promoting the construction of a prototype thorium reactor controlled by a particle accelerator system called an "ADS" or accelerator-driven system "as proposed by the Nobel Prize Laureate in Physics"; Carlo Rubbia at CERN [1, 2] (Fig. 1).

The benefit of C. Rubbia's solution is its ability to answer the energy challenge while minimizing the ecological impact any such production entails. The goal of iThEC is to promote the concept and thereby:

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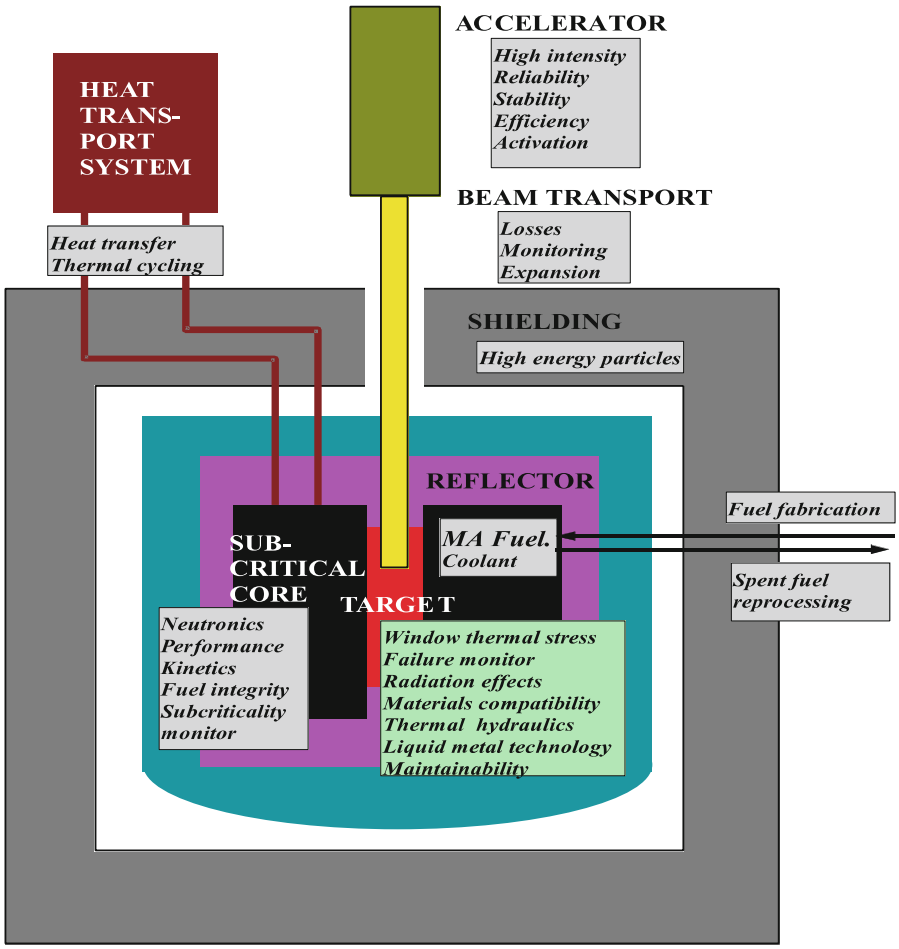

Fig. 1 Carlo Rubbia's concept of the energy amplifier (*MA* minor actinides). *Source* PSI

1. Demonstrate that the proposed energy production method can contribute significantly to economic development without compromising the eco-system.
2. Demonstrate on an industrial scale that much of the volume and the lifetime of nuclear waste can be eliminated; thus, reducing the risk of deep disposal, along with the costs and fears that it generates.
3. Foster know-how and progress in the nuclear field, to ensure effective control of existing nuclear power plants and decisive safety improvements in future nuclear power plants.

It would seem wise from the perspective of iThEC to undertake an international research effort centered on this innovative and sustainable concept.

A Path to Sustainability; the Thorium-ADS

Current nuclear power systems find their origins in the pioneering work of Physicist Enrico Fermi in the 1940s who carried much of his work under the constraints of the wartime period. His elegant and simple solution to harnessing nuclear power hinged on his discovery of the inherent properties of nuclear fuel; delayed neutrons and counter-reactions such as the Doppler Effect in the fuel. For the past 70 years, it is a solution that has served the nuclear power production industry well. However, this method does not result in the only or indeed the best possible use of nuclear fission. In the intervening years, many fields of science and technology have progressed and opened up new avenues for harnessing nuclear fission. After the accidents in Chernobyl and Fukushima, it would appear reasonable to question whether such other methods using nuclear fission would be more acceptable to society.

Integrating the ADS into existing nuclear power production will be a progressive transformation, closing the nuclear fuel cycle and making it sustainable. An ADS as a new reactor is not per se the answer to all the world's energy needs. Rather it fits into a strategy for reducing the waste and maximizing use of re-sources and it would be deployed in parallel with existing light water reactors (LWR) and generation III/IV reactors (Fig. 2). Future technological developments, difficult to predict today, may well lead to a

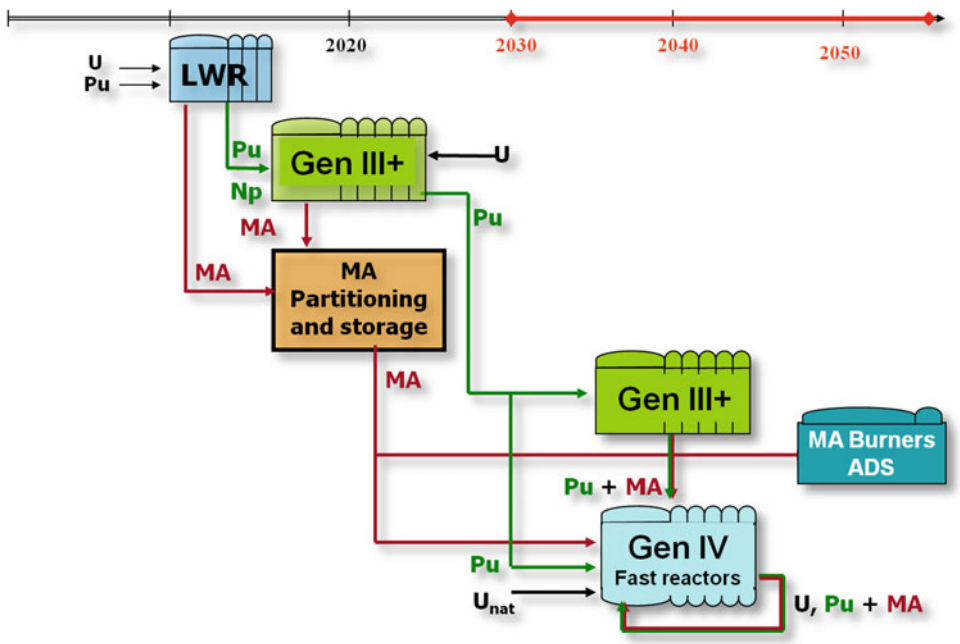

Fig. 2 The ADS in an integrated nuclear fission strategy

dominance of ADS reactors, once operational feedback has been established.

Thorium resources are abundant, several times higher than uranium, and mainly located in politically stable countries (India, USA, Australia, Norway, etc.). They would ensure, for many centuries, a reliable carbon-free supply of electric power. In addition, used in an ADS, thorium would greatly facilitate wider use of renewable energy by ensuring a back-up supply of electrical power that can be adjusted to demand and is continuously available.

Advantages of an ADS

An ADS cooled by natural convection of liquid metal offers significant advantages in terms of maximizing use of resources, inherent safety, non-proliferation, and reduction of existing and future waste.

The members of iThEC are convinced that the extensive experimental work carried out at CERN by C. Rubbia has shown how coupling particle accelerator technology with subcritical nuclear reactors, using thorium as a fuel in a so-called thorium-ADS, offers a viable alternative method of nuclear energy production from fission. C. Rubbia's essential intuition was that by providing an external controllable source of neutrons instead of relying on delayed neutrons, enhanced safety, reliability, and flexibility in the use of the fuel would ensue. The latter being of utmost importance if nuclear waste is to be mixed into the nuclear fuel so as to eliminate it from the existing stockpile.

Production of long-lived waste such as plutonium and minor actinides is significantly curtailed compared with uranium-based fuel. With a thorium-ADS, it is possible to eliminate much of the waste resulting from the operation of existing conventional plants; thus, reducing the size and complexity of long-term nuclear waste storage sites. The key to this unique characteristic is the ability of the ADS to accept fuels incorporating minor actinides and plutonium without compromising safety as the ADS does not depend on the so-called beta factor, the proportion of delayed neutrons, for control of reactivity. Once these substances are eliminated, the waste becomes far more manageable over shorter periods of time (Fig. 3).

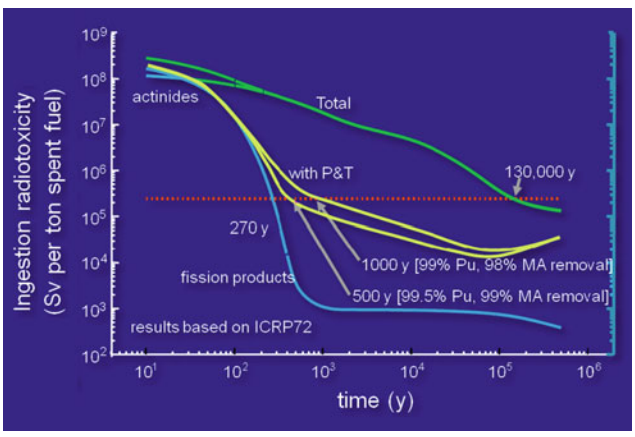

Fig. 3 Waste reduction scenarios

Table 1 Transmutation rates (kg/TW_{th}) of plutonium and minor actinides and long-lived fission products (LLFPs)

Nuclides	ADS Studies			PWR (UO_2)
	EADF ($ThPuO_2$) ENDF/B-VI	EADF ($UPuO_2$) ENDF/B-VI	EADF ($UPuO_2$) JENDL-3.2	
^{233}U	+31.0	–	–	–
Pu	−42.8	−7.39	−5.55	+11.0
Np	+0.03	+0.25	+0.24	+0.57
Am	+0.24	+0.17	+0.14	+0.54
Cm	+0.007	+0.017	+0.020	+0.044
^{99}Tc prod.	+0.99	+1.07	+1.22	+0.99
^{99}Tc trans.	−3.77	−3.77	–	–
^{129}I prod.	+0.30	+0.31	–	+0.17
^{129}I trans.	−3.01	−3.01	–	–

[a]Note that positive rate values denote production, negative destruction
EADF Energy amplifier demonstration facility

Inherent safety is guaranteed by immediate shut-down of the accelerator, thereafter shedding the decay heat through natural convection of the liquid metal coolant, which remains well below the boiling point. Unlike water, the chemically inert liquid metal coolant carries no danger of formation of hydrogen as in Fukushima. Furthermore, natural circulation is independent of pumps and does not require a power supply.

Military proliferation risks are greatly reduced as plutonium production is negligible. The main fissile element is uranium-233, but present in an isotopic mixture unfit for military operational use.

Transmutation of Plutonium with Thorium

Thorium offers demonstrably superior capabilities in terms of reducing the waste. Table 1 shows the results of different studies carried out for ADS loaded with either uranium or thorium in comparison with the baseline UO_2-loaded pressurized water reactor (PWR). In terms of plutonium destruction, the ADS loaded with thorium burns five times as much as an ADS loaded with uranium, not mentioning, of course, that a conventional PWR actually produces plutonium. Plutonium, because of its proliferating nature, is a particularly challenging type of waste.

The Way Forward Proposed by iThEC

The strategy proposed by iThEC [3] is to make maximum use of prior experience and existing equipment to facilitate and accelerate the development of an ADS. A staged development, gradually increasing the size of the prototypes, should enable risk-informed decisions to be taken, minimizing the likelihood of technological or financial impediments to the development of a full-scale ADS. Inevitably, the timescale of such a staged project is likely to span several decades. However, given that the fundamental matters at stake relate to the future of mankind's energy supply, an extended timescale should not be seen as an obstacle.

As described in Table 2, the first stage would start with a relatively modest beam power, one that is currently available in many existing accelerator facilities in Switzerland (PSI, CERN), the US (ORNL), or Russia (Troitsk). In this regard, Switzerland has a unique strength in ADS technology, as the Paul Scherrer Institute (PSI) in Villigen has developed a cyclotron with a proton beam with the capacity to drive a nuclear waste incinerator. PSI also managed in January 2007, a pioneering experiment with the operation of a megawatt-class neutron spallation source; MEGAPIE [4, 5]. With a proton beam power on the order of 1 MW, such a neutron source would be sufficient to eliminate about 30 kg of plutonium and minor actinides per year. It is evident for iThEC that Phase 1 should be hosted by an existing accelerator facility and the organization is actively pursuing this route through its many contacts. A realistic timeframe for this first stage would be 2015–2020.

The following Phase 2 and Phase 3 would derive vital knowledge necessary for the optimization of any new nuclear system from this first pioneering Phase 1. Although it is difficult to foresee all possible difficulties, past experience in developing fast reactors lend credence to the belief that the next stage—Phase 2—could be developed over a period of 10 years. This should be enough to develop the correct licenses and operate the system for at least a few years, prior to the full-scale industrial prototype in Phase 3. Given the main industrial interface with the operation of the existing diverse fleet of nuclear reactors and the likely

Table 2 Staged development of an ADS facility

	Phase 1 2015–2020	Phase 2 2020–2030	Phase 3 >2030
Proton driver power	250 MeV * 3 mA = 0.75 MW_{th}	250 MeV * 6 mA = 1.5 MW_{th}	900 MeV * 6 mA = 5.4 MW_{th}
Gain G_0	0.75	0.75	2.5
Subcriticality, k	0.95	0.975	0.975
Gain = $G_0/(1 - k)$	15	30	100
Thermal power	11.25 MW_{th}	45 MW_{th}	540 MW_{th}

diversity of reactor generations that will be in operation in 2030, predictions for the final phase are difficult. At the very least, starting a program of this type in earnest would offer a realistic chance of addressing the major challenges of the nuclear power industry beyond 2030.

Conclusion

A thorium-fueled ADS offers unique possibilities in eliminating long-lived waste, one that is unrivalled in terms of sustainability safety and flexibility. The system not only transforms waste into stable products but at the same time pro-vides energy.

As with any technological development, uncertainty as to its affordability persists at this early stage. iThEC has therefore proposed a phased approach, consisting at first in an assessment of the technologies with a laboratory-scale prototype, followed by a second phase for testing innovative technologies at a prototypical level, and finally a third fully industrial phase, which would integrate the ADS concept into the existing nuclear power infrastructure.

iThEC aims to use its network and promote this innovative approach through international collaboration, to contribute to solving one of the most pressing needs of the present times.

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Part VII
Summary of the Round Table Discussion

National and International Thorium Programmes (Sessions 1, 2)

Anil Kakodkar

National and International Programmes

Well friends, I think you will agree with me that we had a very good and lively conference. It is important for us to recognize that today nearly 1/6th of the global electricity comes from nuclear power. This, however, has reached a plateau on account of a number of issues that we all are aware of. Thorium, which is a larger resource compared with uranium, seems to have the potential to resolve many of the issues that impede the rapid growth of nuclear power. Even so, we can't imagine thorium replacing uranium any time soon. I think we need to position thorium in the context of some special niche areas that truly belong to thorium. There are several issues that have remained unresolved during the pursuit of the uranium cycle. I think for us success would lie in exploring at least some of those issues, if not all, that can be resolved through the use of thorium and not project thorium as a replacement for uranium, at least to start with.

I believe that in most of the existing commercial power reactors there are opportunities to introduce thorium with a view to getting a larger energy value from natural uranium. We should explore opportunities where this is likely to be commercially more attractive. Secondly, we need to recognize the fact that we need energy in forms other than electricity. Nuclear energy playing a major role in replacing fossil hydrocarbons, for example, would be very important for climate change considerations. Producing nuclear energy at higher temperatures is important in this context. Thorium has a unique opportunity here in becoming a major source in the primary energy supply. Molten salt reactors and accelerator-driven subcritical reactor systems have been discussed in depth in this meeting and so we are all very clear about the opportunities with thorium there.

I have been asked to make some remarks or suggestions in terms of what co-operation one could think of. Ideally, if everyone can converge on an agreed system configuration for a thorium reactor, that could be the theme for international cooperation. That, however, is a very difficult convergence to achieve. There are several core technologies that are important for a number of reactor types relevant to thorium. Molten salt systems, high-power proton accelerators, and high-temperature systems are examples of such core technologies. It is perhaps easier to reach convergence on the identification of such core technologies, on which one can set up cooperations. One can think of looking at some of them or all of them. The simplest thing to begin the cooperation, however, would be to explore opportunities for thorium utilization in existing systems and to work on nuclear as well as thermo physical data for different aspects of thorium use. We did talk about nuclear data, but I think thermo physical property data sets are equally important, particularly, when we are trying to deal with different sets of materials.

Now, to make a cooperation program work, you need a structure to drive it and I get a sense that the international Thorium Energy Committee (iThEC) does have that interest or the ambition. We also need willing participants and I am sure the Committee would ask or invite people to participate and we hope that the participation will grow. We also need an agreed program. Given below are some ideas, in increasing order of challenge, based on what I have said earlier.

(1) Studies in agreed areas of common interest.
(2) Development of core technologies that are of common interest.
(3) International project on a thorium reactor.

Apart from building consensus on areas of cooperation, one would need to negotiate financing arrangements and that is another problem I hope the organizers would be able to tie this up.

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Innovative Thorium Reactor Concepts (Sessions 3, 4, 5)

Hamid Aït Abderrahim

Thorium Reactor Concepts

The morning session's request for more discussion time was fulfilled. A lot of lively exchanges took place. Warm thanks to all the speakers and to the audience!

India's Thorium Programme

Dr. Vijayan reminded us of the very original "three-stage strategy" set forward by Prof. Homi Bhabha, in the 1950s for India's nuclear power development. The Indian thorium programme is very well structured, very goal-minded, applies sound technology (Indian-developed), and is timely. The Indian thorium programme is the first, in a long time, that provides for full-scale validation of:

(1) Heavy water reactors using natural uranium fuel to breed plutonium;
(2) Sodium-cooled fast reactors using plutonium fuel to breed ^{233}U;
(3) Advanced heavy water reactors using thorioum + ^{233}U fuel;

Steadily, India masters all steps of the thorium cycle. High-temperature reactors, molten salt reactors, and accelerator-driven systems are also under development for future deployment.

UK Study

Dr. Rob Arnold presented some projections based on scenario studies for a potential deployment of nuclear power in the UK based on the U–Pu fuel cycle.

He reviewed three scenarios:

(1) No new conventional nuclear plants in the UK (not even those currently proposed);
(2) Up to 75 GW open-cycle reactors around the mid-21st century;
(3) Up to 75 GW closed-cycle reactors around the mid-21st century.

His presentation included a study on thorium-based molten salt reactors (MSR), and showed agreement with findings from a Grenoble study (EVOL FP7 project, Elsa Merle).

Is the Thorium Cycle Feasible? Is It Desired?

Can Raj Seghal's conclusions and recommendations, summarized here, be considered consensual?

"I believe that it is prudent to develop an alternative to the U–Pu fuel cycle for future generations". However, it is not easy to make a case for commercial thorium fuel utilization only on the basis of economics. Uranium fuel prices are still much lower than 100 $/pound or 220 $/kg, which is considered as the break-even price for the introduction of an alternate fuel cycle (fast reactors or thorium cycle).

"The thorium cycle does provide an efficient platform to burn and transmute ^{239}Pu; in light water reactors (LWRs) or fast reactors". The ^{233}U formed cannot be easily used for weapons manufacture, as it contains much ^{232}U. Long burnup operation of the Pu–Th fuel could also burn the ^{233}U formed from thorium, thereby producing additional energy. Such an open fuel cycle could indeed be economically competitive with the open fuel cycle currently employed for LWRs. A "mission" of disposal of accumulated plutonium could be justified for thorium.

"In the future, when fuel cycles are closed, the ^{233}U made in Pu–Th-fuelled LWRs or fast reactors could be extracted and employed for power generation in LWRs". This would

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require shielded facilities for reprocessing and re-fabrication, which would be expensive, but feasible.

"Research and pilot-scale developments for the thorium cycle will need public financing. If governments assume that responsibility, commercial use could become feasible and desirable."

Alex C. Mueller's Personal View

The CERN DG made the point with the success story of the Higgs particle: "In order to achieve its goals, a community must federate around one single project to which everybody is committed". As much as the particle and nuclear physics community federated around the Large Hadron Collider, our community must unite around MYRRHA, the only well-advanced project for which at least partial funding exists.

Either we get this project through together or ADS in general and Th-ADS in particular will be killed. Nothing is more harmful than a dispersed community.

Other Important Points

Dominique Greneche reminded the audience that like any other technology, thorium-cycle-based nuclear energy also has some drawbacks. The warnings contained in Dominique Greneche's engaging talk should be seriously taken into account.

A beautiful lesson of the great advances for the physico-chemistry of molten salts and the potential capabilities of atomistic modelling to derive molten salt material properties have been shown by Paul Madden.

These R&D progresses will open the door to industrial perspectives such as presented by Kirk Sorensen (Flibe liquid fluoride thorium reactor (LFTR) strategy development) but one should not underestimate the time needed for civilian application of nuclear technology.

Other Important Views on Thorium

Luc van Den Durpel from AREVA, delivered good news: AREVA and Solvay (two large industrial groups with significant knowledge on thorium and nuclear energy) are interested in the thorium fuel cycle. They are investing in a R&D programme and invited the audience to join them if interested.

But he also mentioned that AREVA, as a nuclear industry representative with fuel cycle development as part of its strategy, doesn't want to mix the U–Pu and Th–U cycles. Instead, the Th–U cycle should be introduced progressively in the present fleet of reactors. This is a very long endeavour (>100 year).

Prof. Muammer Kaya announced the Turkish Government's decision to embark on nuclear power generation, but based on the U–Pu fuel cycle, whereas the country is the second largest country with thorium reserves. His aim is to stimulate awareness in decision makers of the thorium potential for nuclear power in Turkey and to create a Thorium Research Center (ThoReC).

Tony Donaldson from Rolls-Royce presented a review of the advantages and challenges of molten salt–thorium-based reactors for commercial applications in a fully neutral approach.

Dr. Jan Uhlir reported on the Czech National programme in the field of molten salt reactors (MSR) development associated with pyro-reprocessing of LiF-based fuels: MSR and pyro-reprocessing R&D has been a research topic for decades in the Czech(oslovac) Republic. Since 2000, a renewal of these activities has been stimulated by the support of the Ministry Of Industry and Trade and allowed the Nuclear Research Institute, Rez, (NRI) and other Czech partners, such as Skoda Nuclear Machinery, to be important players in the European Commission's framework programmes 5, 6 and 7 projects (ALISIA, MOST, EVOL). The objective is to position the Czech programme (material research, pyro-reprocessing, tests in the LR0 reactor, reactor design) in a larger international framework.

Thorium-Fuel Cycle and Transmutation (Sessions 6, 7)

Yacine Kadi

Thorium, occurring as a fertile Th-232 isotope, is far more abundant than uranium, yet it has so far eluded any use in the nuclear power industry. Far from being unknown, thorium was envisaged as a nuclear fuel from the beginning of the nuclear age, yet its nature as a fertile nuclear material, rather than a fissile one as Uranium, has impeded its development.

As demonstrated by Prof. Dr. Şahin [1], much of the present nuclear power generation is concentrated in industrialized countries utilizing a uranium open cycle, which is inherently limited by the finite supply of uranium. In view of the needs of the developing world, thorium could present a long-lasting solution to their energy needs, if an efficient method for breeding fissile U-233 from Th-232 can be found. The different options include alternative mixed fuels in CANDU reactors (use of TRISO particle for deep-burn of Pu and MA circa 400 GWd/t), nuclear fusion energy systems (different blanket configurations for breeding tritium based on LiF–BeF_2–thorium molten salts, or Li-cooled ThO_2 fueled with Pu and minor actinides (MA)), and accelerator-driven systems. The CANDU reactor has the attraction of being able to breed thorium using seed nuclear material such as plutonium with conventional technology. A more advanced option would be to breed U-233 by using a high-power neutron source, derived either from a fusion driver or from a spallation target.

The United Kingdom's National Nuclear Laboratory has examined closely the complete fuel cycle aspects relating to the introduction of thorium [2]. The advantages and disadvantages in terms of sustainability, proliferation resistance, economics, and radiotoxicity were studied with dedicated fuel cycle simulation computer programs to assess the impact of different fuel cycle scenarios. Among the options evaluated were a once-through fuel cycle with Th-232 as a fertile material using a U-235 or Pu-239 driver fuel in a thermal reactor. Options in a fast reactor included Generation IV concepts such as sodium-cooled fast reactors, molten salt fast reactors (MSFR), and accelerator-driven systems (ADS). One of the advantages identified in converting to a thorium–plutonium mixed oxide (MOX) fuel could be plutonium disposal. Also, from a strategic perspective, sustainability remains one of the main drivers of such a cycle. On the negative side, the study found the economics of thorium are still an unknown, in particular, significant R&D programs are required to progress towards technical maturity. In all instances, the time frame for introducing thorium would stretch over decades.

One of the attractions of thorium relates to its purported beneficial effect on waste management, an advantage that can only be exploited if the appropriate reprocessing technology can be developed. India [3] has for some time being developing such a technology as it possesses vast reserves of thorium, which could provide it with near complete energy independence. It has developed a three-stage program, starting with pressurized heavy water reactors (PHWR) using natural uranium, which provide plutonium. In a second stage, the plutonium can then be used for breeding U-233 from Th-232 in a fast breeder, which is currently under construction. Finally, in the third stage, once a large enough U-233 inventory has been bred, it would be used as a nuclear fuel in an advanced heavy water reactor (AHWR) dedicated to power generation. The need for thorium fuel has driven a research program focused on irradiating thorium fuel elements in the existing PHWR fleet. Facilities have been built to reprocess these irradiated fuels and some success has been registered in recovering U-233. The main difficulty lies in the dissolution of thoria fuel using fluoride as a catalyst. Vitrification of thorium wastes has also been achieved on a prototypical basis.

Reprocessing thorium fuel is also of interest to the French CNRS, which has set about investigating aqueous, and pyro-reprocessing [4]. The pyro-chemical route was examined based on a molten salt reactor cycle. The research

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group proposes a complete reprocessing scheme, identifying for each element an appropriate extraction method as well as closing any gaps in the available database. It is hoped that in this manner an experimental device may be developed to test the efficiency of the proposed reprocessing. The aqueous treatment of solid oxide fuel would center on the Thorex process with dissolution in nitric acid followed by liquid–liquid extraction of Th/U by 30 % tributyl phosphate (TBP). Another process also used in India starts with dissolution in nitric acid, selective liquid–liquid extraction of U by 5 % TBP, leaving thorium in the waste. The CNRS research is continuing with support from Areva, Solvay, and the Euratom consortium.

Investigations led by the Paul Scherrer Institute (PSI) in Switzerland [5] indicate that the molten salt reactor (MSR) has excellent neutron economy in the U–Th cycle as the capture of U-233 is low, but parasitic absorptions by the carrier salt and graphite are also quite reduced. The fact that the fuel is in a liquid state gives the reactor additional flexibility compared with solid fuel rods and inherent safety. Indeed, it is possible to remove on-line highly mobile gaseous and volatile fission products and the absence of a cladding eliminates the risk of it failing. The PSI has, thus, established a cooperation with research centers and universities to study the feasibility of the MSR using advanced computational tools and focusing on reprocessing possibilities offered by this concept as well as the simulation of the core.

The Chinese efforts [6] in evaluating thorium for its use in a nuclear reactor have led to a program for improving the nuclear data used in neutronic studies of a thorium reactor. The fission product yields of Th-232 induced by 14 MeV neutrons were measured by direct gamma spectroscopy and, by taking into account the uncertainties, noteworthy discrepancies with the existing data were found. A system study of thermal cross sections found that there is a lack of sound experimental data for Th and Pa. Recommendations were issued for fission product yields for Th-232 based on a combination of ENDF/B-VII.0 and JEFF-3.1.1. The nuclear reaction data for Th-232 and U-233 in CENDL were revised and improved, based on the feedback from benchmark testing.

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Thorium-Reactor Physics (Session 8)

Robert Cywinski

The deployment of thorium as a nuclear fuel has had a long and largely successful history, stretching back to the 1960s when the thorium-fueled Peach Bottom nuclear plant pioneered the commercial exploitation of high-temperature gas reactor technology. However, since the closure of the thorium-fueled 300 MW HTGR in Germany in the late 1980s, thorium reactor physics has been largely neglected, except perhaps in India. We now appear to be on the brink of a considerable resurgence of interest in thorium as a potential nuclear fuel and, coupled with the development of new reactor designs and modes of operation, this has presented new opportunities, but also many new challenges. Session 8 of IThEC focused upon both the opportunities and challenges.

Firstly, Srinavasan Ganesan highlighted the considerable breadth of activities of the Nuclear Data Physics Centre of India with a clear emphasis on the need for and progress in the measurement and curation of neutron and charged particle induced cross sections associated with the Th–U fuel cycle and other processes, particularly with a view to improving the reliability of Monte Carlo codes for reactor design and simulation.

This theme was continued by Frank Gunsing (CEA) who provided a comprehensive overview of the exploitation of phases I and II n_TOF (neutron time of flight) facility at CERN between 2001 and 2012 for the measurement of capture and fission cross sections relevant to both the thorium and uranium fuel cycles and the transmutation of nuclear waste.

Konstantin Mikityuk (PSI) followed with a presentation on the history and technology of fast reactors whilst exploring factors such as sustainability, safety, proliferation resistance, and radiotoxicity of the waste associated with the thorium fuel cycle. Although calculations show that in principle an equilibrium closed cycle fast reactor fueled with thorium is possible, the thorium cycle has both advantages and disadvantages. Particular advantages are the very low quantity of minor actinides in the EQL-Th fuel cycle; the fact that the Doppler effect is stronger and void effect are weaker in EQL-Th fuel compared with EQL-U; and the radiotoxicity and decay heat of EQL-Th fuel are lower during the first 10,000 years of cooling compared with EQL-U fuel. However, the disadvantages include the fact that U-232, the precursor of hard gamma emitters, is produced in the Th–U cycle with the (n, 2n) reaction being higher in a fast spectrum; k_∞ of equilibrium fuel is lower for the Th–U cycle compared with U–Pu, so to reach iso-breeding, blankets of fertile material are required; and the fissile precursor of U-233 (Pa-233) has a longer half-life compared with Np-239, so the potential for separation and decay to pure U-233 is higher. Moreover, it was emphasized that our collective experience with thorium fuels and thorium reactor technology is significantly less than that with U–Pu fuel cycles and deployment.

The final presentation of the session was made by Elsa Merle-Lucotte (IN2P3, CNRS), who gave an introduction to the physics of molten salt fast reactors (MSFRs). Not only were the advantages of a liquid fuel considered, but the particular advantages of the Th–^{233}U cycle over the U–P cycle (for which the neutronic properties of F in the LiF salt are not favorable) were also demonstrated. There then followed a detailed evaluation of the design characteristics, neutronics, fueling, and safety of a realistic MSFR, carried out with support from the Euratom-funded EVOL program.

In the ensuing discussion it was widely accepted that to move the thorium agenda forward there is a need for a continuing international effort to measure, refine, and curate nuclear data related to the thorium cycle. Additionally, deep modeling of whole thorium-fueled reactor systems using that data must be carried out, and this in turn necessitates a willingness to accept or reject appropriate or inappropriate thorium deployment scenarios based upon evidence provided by the data and the modeling.

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Accelerator-Driven Systems: The Accelerator (Session 9)

Mike Seidel

ADS: The Accelerator

Requirements for ADS Accelerators

The thermal output of an accelerator-driven reactor scales with the beam power of the driving accelerator:

$$P_{\mathrm{th}} = \frac{G_0}{1-k} P_{\mathrm{beam}}$$

In principle, the amplification factor can be made large by choosing a value for k close to one, that is, by approaching critical operation. However, for safety reasons k very close to one is not desirable and consequently the driving accelerator must deliver significant beam powers. In practice, beam powers ranging from a few megawatt up to several tens of megawatt are being discussed for ADS systems. The required beam energy is in the range of 0.8–2 GeV. The highest beam powers achieved in accelerators today are just above 1 MW. The most critical aspect of such accelerators is unwanted loss of beam that leads to activation of accelerator components, affecting the serviceability of the accelerator. Furthermore, other aspects are also important and can be summarized in the following list:

- High beam power of several MW must be provided with losses less than 50 W/location or less than 1 W/m for stretched out losses. Thus, relative losses should be in the range below 10^{-4};
- A low rate of trips lasting over a second must be achieved to ensure continuous operation of the reactor. Trip rates of such duration as low as 0.01–0.1 trips per day are necessary. This figure is two to three orders of magnitude better than the performance of existing high-power proton accelerators;
- The energetic efficiency of the driver accelerator should be high for the conversion of grid power into beam power. Efficiencies achieved today are in the range of 10–20 %;
- The thermal power of the reactor depends on the beam power and it should be controlled with reasonable precision. Thus, the accelerator must provide a good stability for beam current and energy;
- The cost of the accelerator should be acceptable.

Review of the Contributions

J.P. Revol, CERN, opened the session with a review presentation on the history and concept of ADS systems. In the last decades, the physics of driven reactors has been studied in great detail and today precise simulations and predictions are possible. The use of thorium in a fast driven reactor is advantageous. The requirements for ADS accelerators, as summarized above, were described.

A.C. Mueller, CNRS, gave an introduction on the physics of particle accelerators. Cyclotrons and superconducting linear accelerators (linacs) are accelerator concepts for high-power operation. For very high powers and in view of low trip rates, superconducting linacs are preferred according to a review by the Nuclear Energy Agency (NEA).

P. Mandrillon, AIMA, reviewed cyclotrons for ADS applications. The PSI cyclotron with 1.3 MW average beam power demonstrates good performance. Newly proposed cyclotron concepts for the Daedalus project and an 800 MeV stacked strong focusing cyclotron by A&M Texas University were discussed. The author focused then on a cyclotron for H_2^+ acceleration with reverse bends for clean stripping extraction and three low-energy beam injections at 60 keV. Thus, no injector cyclotron is needed for this machine.

F. Bouly, CNRS, reported on the MYRRHA project, which is an ADS demonstrator experiment to be built in Mol, Belgium. The accelerator will deliver 600 MeV protons with more than 1.5 MW beam power and will utilize a superconducting linac. The project is in an advanced stage

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and many subsystems, such as source, booster accelerator, superconducting spoke cavities, and superconducting elliptical cavities, are in a prototyping phase. In this study, significant effort has been made to improve the fault tolerance of the linac by automatic energy compensation of individual failed cavities.

R. Barlow, Huddersfield University, focused on cyclic accelerators and considered the Daedalus proposal using cyclotrons with up to 4.8 MW beam power for neutrino production. For Daedalus, the acceleration of H_2^+ molecules with a much higher binding energy than H^- is planned. This would allow clean stripping extraction of the beam. Then, he discussed a non-scaling fixed-field alternating gradient (FFAG) accelerator for high beam intensity, which has constant optics, resulting in a constant tune and a large aperture. These developments are still at the conventional level and technical details are not worked out. As cyclic accelerators are potentially cheaper, more than one could be used to drive an ADS reactor, thereby achieving redundancy for reliable operation.

Conclusion and Round Table Discussion

From the presentations one can conclude that the theory on ADS reactor systems is well developed, including the availability of simulation codes. The requirements for the accelerator are well defined and present a good basis for designing concrete ADS systems. Concerning beam power, today's accelerators are just at the threshold where they can be used for ADS systems. An example for the next step in accelerator design is marked by the European Spallation Source (ESS). The superconducting linac of ESS will deliver 5 MW of beam power, a figure that is already sufficient for many ADS applications.

The two major concepts under discussion are cyclotrons and superconducting linacs. Although cyclotrons as cyclic accelerators are compact and cost effective, their beam dynamics is more complicated, clean extraction is demanding, and tuning of cyclotrons is tedious. Superconducting linacs are costly installations, but have intrinsically low losses due to their large aperture and the absence of extraction elements in comparison to cyclotrons. Overall, one can state that most present projects are planning to utilize linacs, but development efforts are ongoing as well for cyclotrons, with a potential for cost-effective solutions.

During discussions, the question of the efficiency of particle accelerators for converting grid power to beam power was raised. Today, the overall efficiency of high-intensity accelerators does not exceed 20 %. This figure is mainly determined by radiofrequency (RF) sources and RF losses in accelerating resonators. Superconducting accelerators have low RF losses, but are limited by the power needed for the cryogenic plants. With future developments the efficiency could be improved, but likely not above 40 %.

In the summary presentation, the ESS project was mentioned as an example of an advanced accelerator with 5 MW beam power. A comment was made by F. Meot, that the high beam power of ESS is achieved with a beam energy of 2.5 GeV, which is unnecessarily high for ADS systems. It will be more demanding for a linac to achieve the same power at lower energy, for example, 1.2 GeV, as the higher current will require more power to be transmitted per coupler of the linac. Furthermore, he commented that studies are underway to achieve continuous-wave acceleration at fixed frequencies, also in FFAGs, and with electron models such schemes have already been demonstrated. M. Seidel agreed that FFAGs have potential, but the clean acceleration of high-intensity beams remains to be demonstrated with such accelerators.

P. Mandrillon pointed out that the important aspect of the cyclotron concept he presented was its simplicity and the absence of a costly injector installation, resulting in low cost.

Accelerator-Driven Systems: The Spallation Target (Session 10)

Karel Samec

ADS: The Spallation Neutron Source

Neutron sources delivering high-intensity neutron fluxes from the spallation reaction of high-energy protons on heavy nuclei have achieved a remarkable degree of maturity in their development with key test results such as MEGAPIE. Amongst their many possible uses, such neutron sources are a key component of the accelerator-driven system or ADS.

The ADS concept rests on three main components: the accelerator, the central spallation neutron source, and, surrounding it, the subcritical core. Of all three components, the spallation neutron source had been identified as the less well known and was therefore specially targeted for further research in a dedicated test at high power. The MEGAPIE project for "MEGAwatt PIlot Experiment", ran successfully at 0.78 MW of beam power for 4 months in 2006 at the PSI SINQ facility and provided an instant increase of 80 % in neutron flux over the conventional SINQ source. The project was launched in 1999 to design and build a high-power liquid metal lead–bismuth eutectic (LBE) spallation neutron source [1]. It was to operate in the existing Swiss spallation neutron facility, SINQ, at the Paul Scherrer Institute (PSI), replacing the standard solid-target technology neutron source for a limited time. The main achievement for the program was to validate experimentally a completely new concept in a configuration well suited to an ADS application. Indeed, MEGAPIE presents itself as cylinder, of which the lower end, comprising the spallation zone, is 2 m long and 20 cm in diameter [2]. Such a design lends itself to being placed in the center of a subcritical core to serve as a high-intensity neutron source controlling the fission reaction in a subcritical mode. Of particular relevance to ADS development was the demonstration that a compact self-contained spallation neutron source operating with liquid metal and using a solid T91 stainless steel beam window could be operated reliably.

Similar undertakings have been successful in the US with the Spallation Neutron Source (SNS) and with the Japan Proton Accelerator Research Complex (J-PARC) where megawatt-class mercury-type spallation neutron sources are also under operation for pure research, their configuration being less suited to an ADS. In both instances, the facilities are operated in pulsed mode, which has posed some challenges. Indeed, mercury expansion and wave reflection at the vessel interface lead to tension in the mercury and cavitation, a problem that has been solved by the injection of a gas bubble stream into the liquid metal at the point of entry of the beam, just behind the beam window.

Efforts are underway to implement neutron spallation sources in power production plants. An early stage of development for such a project is shown in [3] for a pilot plant to be developed in Venezuela. The concept centers on an accelerator-driven molten salt reactor (ADMSR) in which the fuel is dissolved as a F–Li–Be salt in a reactor core surrounding a central neutron source. The compact neutron source is based on an existing design from the EURISOL program and has been implemented in a neutronics model suitable for evaluating performance. First results indicate that performance in the megawatt range is indeed attainable with a fairly small reactor vessel; the inside diameter containing the fuel being no more than 60 cm in diameter.

Pursuant to the considerable nuclear expertise in the state of Virginia, a public–private initiative has been set up, the Virginia Nuclear Energy Consortium Authority [4]. Stakeholders in the private sector include AREVA, Babcock and Wilcox, Mitsubishi Nuclear Energy Systems (MNES), along with state institutions such as the University of Virginia and James Madison University. Institutions outside Virginia, such as Oak Ridge National Laboratory, have also joined in the venture. An immediate goal of the venture is to develop affordable nuclear medical isotope technologies for use as medical imaging agents and for cancer treatment. The

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developments are to be affordable and ensure a plentiful supply of isotope while improving public perceptions of nuclear medicine and technology. First concrete steps involve linac multi-cell cavity fabrication and the sourcing of niobium end products. The accelerator-driven system is also being evaluated, using resources and local expertise available from pioneers in the field such as Charles Bowman.

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Accelerator-Driven Systems: National Projects (Session 11)

Maurice Bourquin

ADS: National Projects

In this session, two ADS development activities in Asia were reported.

Professor Cheolho Pyeon from Japan presented the first real experiments of their kind on thorium-ADS. In Kyoto University, the ADS consists of the Kyoto University Critical Assembly (KUCA) and the Fixed-Field Alternating Gradient (FFAG) accelerator, which produces a neutron flux with 100 MeV protons on a W target. A 14 MeV neutron source is also available. Experiments are performed in the critical state by irradiating thorium plates and in the subcritical state with a thorium-loaded core. Capture and fission reaction rates are measured and compared with numerical analyses.

Professor Pyeon indicated that the objective of the Japanese ADS is transmutation. The present studies are academic, while waiting for the new ADS facility at the Japan Proton Accelerator Research Complex (J-PARC) to start operating.

Professor Jong-Seo Chai from South Korea first presented the current status of nuclear power plants in South Korea. In operation are 23 units, under construction are three pressurized water reactors (PWRs), and under license review are four PWRs. There is an urgent need to solve the national problem of nuclear waste. In the past, ADS research was performed by the Korea Atomic Energy Research Institute (KAERI), which resulted in a 100 MeV pulsed beam proton linear accelerator (linac) built by KAERI that now provides proton beams of 20–100 MeV for science research purposes. However, now ADS research is being carried out by universities, mostly at Sungkyunkwan University (SKKU). Their R&D plans include the development of a high-energy high-current cyclotron accelerator system of 5 MW for an accelerator-driven thorium-based nuclear reactor. It comprises an injector linac, and intermediate stage cyclotron, a final stage high-energy cyclotron (1 GeV, 12 mA), and a beam transportation system. At SKKU, various research on transmutation and cyclotron design for ADS are going on, including computational modeling.

Professor Chai noted that the motivation for these mostly university research projects stems from the public pressure to stop producing radioactive wastes in operating nuclear power plants and to have available a reliable production of electricity. As wind and solar power will not provide enough power, a solution could be with ADS. To that end, the aim is to have in Korea by 2040 an operational ADS nuclear reactor system, a proposal well supported in the media.

In the discussion that followed, it was mentioned that a similar situation exists in Switzerland and in Germany, where government decisions have been made to forbid replacing existing nuclear power plants. But discussions on developing ADS has not (yet) taken place in these countries. In Switzerland, a popular vote will take place next year in the form of a modification of the federal law on energy.

Professor Chai added that Korea was developing a cyclotron system instead of a linac in order to obtain better stability. He believes that if a worldwide ADS community is formed, the Korean government would support ADS research in Korea. The private sector is concentrating efforts on PWRs, but could change in favor of thorium.

Questions were raised as to how the results of this conference can be best used to advance the case for thorium-based nuclear energy. It was suggested that highlighting the destructive effects made on the economy by subsidized wind and sun power in some countries could change the situation. Indeed, in countries like Korea the solar energy industry, which cannot survive without subsidies, is in great difficulty. The development of wind power is recognized as bringing environmental problems as well.

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Accelerator-Driven Systems: National Projects (Session 12)

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ADS: National Projects

The accelerator-driven system or ADS, based on an accelerator coupled to a spallation source and subcritical core, is attracting renewed interest in the wake of the Fukushima accident due to its unique abilities as a means of reducing the level of nuclear waste and eliminating many accidental scenarios, which afflict conventional critical nuclear reactors. Representatives of many nations outlined their countries' perspective on developing these new types of reactor systems.

The Institute of Nuclear Research, RAS, in Moscow, represented by Dr. Sidorkin [1], has already in operation one of the world's most powerful linear accelerators (linacs) coupled to a pulsed neutron source. The existence of this hardware makes it an ideal candidate for developing an ADS demonstrator at minimal investment, an order of magnitude lower than alternative projects with no pre-existing infrastructure. The thermal power delivered in the core would reach 8.74 MW with a beam of 300 μA/600 MeV on a ^{Nat}U target at a k_{eff} of 0.95. The core physics have been calculated and the essential engineering layout of an ADS fitting in the existing facility has already been elaborated. Such a demonstration ADS would fulfill the important goal of burning minor actinides at a level that would be difficult to achieve safely in a conventional critical reactor. A second goal would be to demonstrate the ability of the ADS system to withstand the thermal transients induced by beam trips.

The People's Republic of China is investing heavily in ADS technology as witnessed by the impressive account of ADS-related experiments in China by Dr. Lei [2]. The Chinese ADS roadmap fixes an ambitious goal of developing a demonstration facility by 2022. Important milestones in the development of the project include the testing of superconducting cavities for a projected linac with an ultimate goal of 250 MeV/10 mA. A computerized engineering mockup has been constructed of the ADS prototype and thermal-hydraulic and structural analysis have been completed both of the coolant flow in the reactor and in the neutron spallation source. Two versions of the reactor are being studied, one cooled by gas, the other by lead–bismuth eutectic (LBE). Test loops with LBE are being used to study the long-term effects of corrosion. Granular flow is seen as an option, in particular for cooling the target, and has been the subject of numerous experiments. The program presented by the Chinese Academy of Sciences is formidable in its scope and breadth and augurs well of the future of the ADS concept in China.

India, a leading nation in Asia in the field of nuclear technology, has embarked on a program of development for the ADS. India is well known for its development of thorium technology initially based on thermal reactors, then followed by fast breeders. ADS technology is seen as part of a later stage of thorium fuel development to improve the breeding of thorium for use in critical reactors. The essential steps in the program are the development of a 1 GeV linac, a spallation neutron source, and a subcritical core with particular attention given to the nuclear data. The essential parts of a high-power linac are undergoing preliminary testing and are still at an early phase. The spallation source is to be based on liquid-metal technology and a first test is planned involving coupling of the beam with the neutron source to validate, in particular, the beam window. Work on the subcritical core centers on validating the nuclear data using a low power neutron source operating at 10^{10} n/s and that is embedded in a subcritical facility consisting of ^{Nat}U rods inside a polyethylene moderator surrounded by a beryllium oxide reflector. As an alternative, a molten salt reactor (MSR)–ADS has been studied from the point of view of the neutronics and has been shown to achieve a good breeding ratio compared with fast critical or thermal MSR. The Indian perspective on the ADS focuses on its complementarity with the existing program for thorium utilization in critical reactors.

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iThEC, represented by Dr. Y. Kadi, outlined past milestone projects in the development of an ADS, most of which occurred in Europe. Starting with the FEAT experiment at CERN, which demonstrated the physics of an ADS, this was followed by TARC, which showed how the spectrum could be tailored to optimize transmutation. The MUSE experiment at CEA and YALINA experiment in Belorussia demonstrated the physics of an ADS in the fast spectrum. Finally, MEGAPIE at the Paul Scherrer Institute coupled a vertical ADS-type spallation source to an accelerator at megawatt level, the first test at prototypical level of an ADS system. Building on these past achievements, iThEC propose a three-phase program with increasing levels of power, starting with an 11 MW_{th} system moving on to 45 MW_{th} and finally demonstrating the ADS at industrial level with a k_{eff} of 0.975 and a power of 540 MW_{th}. By 2015, iThEC aims to set up an international collaboration to build a thorium ADS. In the interim, iThEC will identify material and financial resources and competencies on a world-wide basis and invite interested parties to join in this common undertaking.

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Part VIII
Conclusion

ThEC13 Summary and a Look into the Future

Jean-Pierre Revol

ThEC13: A Significant Success

The purpose of ThEC13 was to review and update the status of thorium energy technologies, from R&D to industrial developments. ThEC13 beautifully fulfilled that goal, as the attendance statistics confirm. About 200 people from 32 countries were actually present at CERN in the Globe of Science and Innovation, including some prestigious scientific and political personalities. In addition, more than 5000 connections were made worldwide to the live webcast.

The conference received broad political and scientific support, which is a clear sign of the renewed interest in thorium technologies, which even reaches into the nuclear industry. AREVA and SOLVAY used the conference to make an important announcement about their long-term strategy concerning thorium:

"AREVA and SOLVAY are joining their know-how to add value to thorium's entire life cycle" [1].

Clearly, in the short time available to me, I cannot do justice to the many excellent presentations given during these three and half days, and my summary will inevitably be somewhat biased.

Government support was also illustrated by the announcement at the conference of a significant industrial grant for thorium chemistry awarded by the Czech ministry of Trade and Industry to the research group headed by Jan Uhlíř of UJV-ŘEŽ in Prague in support of a cooperative venture with the DOE in the US.

Thorium

In his introductory talk, Carlo Rubbia brilliantly summarized the case for considering thorium as a new source of energy. He pointed out that, as it must be used in the breeding mode, "thorium is a practically sustainable source of energy, on the human timescale" [2].

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As ^{232}Th is six neutron captures away from ^{238}U, the production of transuranium elements (TRU), which constitute the long-term component of nuclear waste, is minimized, and this physical property can be used to destroy existing nuclear waste and military plutonium, while producing energy. For the same reason, a thorium fuel cycle has unique potential in terms of nuclear weapon proliferation resistance.

Even though ^{233}U is generally considered better in terms of breeding than ^{235}U and ^{239}Pu (Fig. 1), for reasons of reactor physics, it cannot be simply substituted in place of uranium in a standard critical reactor. In the fast neutron flux required to minimize waste production and to optimize TRU destruction, breeding with ^{233}U is less favorable than with ^{239}Pu (Fig. 1). Therefore, something additional has to be done in order to use thorium efficiently.

Options for the Practical Utilization of Thorium

At ThEC13, three options for the practical utilization of thorium were reviewed:

- Using thorium blankets around reactor cores to breed ^{233}U;
- Continuously circulating the fuel through the core, such as to always have fresh fuel, which can be achieved either in pebble bed reactors (once-through pellet fuel cycle) or in molten salt reactors (MSR), with recirculation of the liquid fuel and requiring reprocessing on-line;
- Using a particle accelerator to provide the extra neutrons needed to sustain fission reactions in a subcritical core, in so-called accelerator-driven systems (ADS). This is the solution proposed, in particular, by C. Rubbia and promoted by iThEC, the main organizer of this conference.

Pebble bed reactors were not discussed at the conference, but it was mentioned that, after the pioneering

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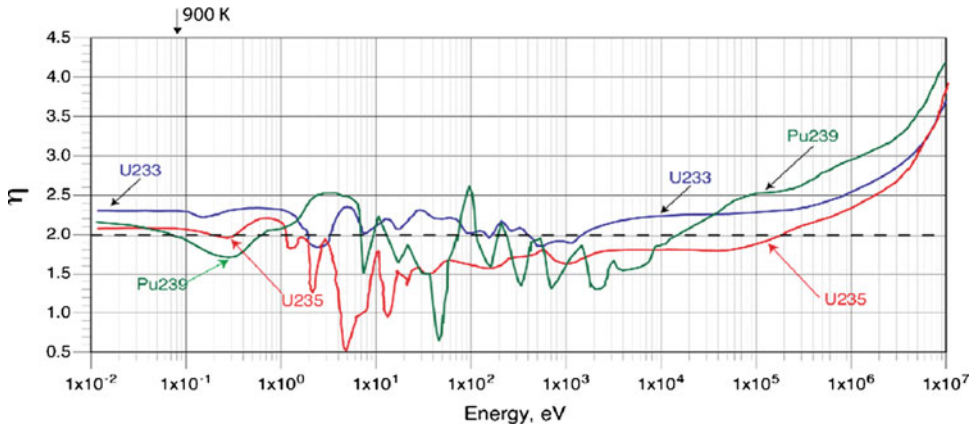

Fig. 1 Number of neutrons produced per neutron absorbed (η) in ^{233}U, ^{235}U, and ^{239}Pu, as a function of neutron kinetic energy, taken from [2]. For breeding to be possible, η has to be larger than 2

THTR-300 project at AVR, Jülich, Germany, was dropped, there were on-going developments in South Africa, United States, and Turkey.

Molten salt reactors (Fig. 2) are based on a technology that seems to focus a lot of interest, if one can judge by the number of talks related to the subject at this conference (ten talks). The subject was well introduced by Merle-Lucotte [4], who presented the physics of thorium molten salt fast reactors. It appeared that MSRs are clearly a line of research where coordination between the various worldwide R&D efforts should be improved, as there is still a lot of work needed to demonstrate the practicality of this scheme on an industrial scale. There is a particularly well-focused and advanced MSR program at the Shanghai Institute of Applied Physics in China, reported by Xu [5]. In the discussion, it was noted that R&D should be extended to other salts that allow a fast neutron energy spectrum, favouring transmutation, and that licensing issues should not be underestimated, but should be addressed from the beginning, as MSRs imply new safety considerations (different confinement barriers,

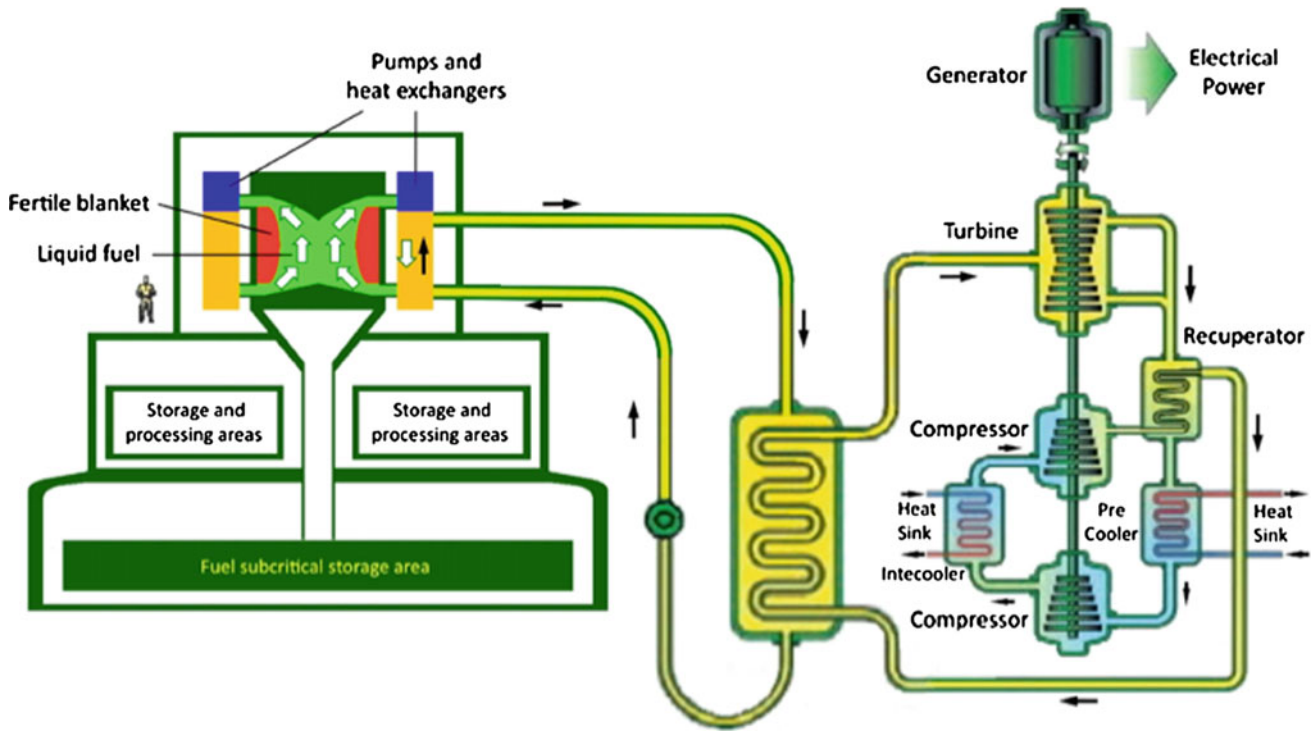

Fig. 2 The molten salt reactor scheme, from Madden's presentation [3], showing the three basic circuits of such systems: fuel circuit, intermediate circuit, and thermal energy conversion circuit

chemical extraction on-line, neutron irradiation outside the core, temporary stops of the chemical extraction flow, corrosion, etc.).

Accelerator-driven systems were the subject of the largest number of talks (fourteen talks). The readiness status of the main technological elements was reviewed:

- Progress in accelerator technology [cyclotrons and linear accelerators (linacs)] is significant, as explained by Mueller [6]. Mandrillon [7] presented an innovative approach to high-power cyclotron beams. Barlow advocates the use of non-scaling fixed-field alternating gradient accelerators (FFAG) [8]. The good news is that, today, both linac and cyclotron proton beams have achieved a sustained power in excess of 1 MW;
- Progress on neutron spallation targets was reported, including the success of MEGAPIE [9], the first MW-class neutron source, which operated successfully for three months at SINQ [10] in Switzerland and the Spallation Neutron Source (SNS) [11] in the United States, now fully operational at a power of 1.3 MW;
- Various core designs were also described.

The most advanced project is MYRRHA [12] (Fig. 3), details of which were presented by its project leader Hamid Ait Abderrahim. Several other ADS concepts, at various stages of developments, were described, including a thorium-fueled reactor for power generation by Rubbia and Aker Solutions [2], and for which a detailed engineering design already exists. A Troitsk demonstrator project [13] in Russia, based on an existing accelerator complex, was presented by Stanyslav F. Sidorkin. Presentations of ADS R&D in China (CADS) [14] and Japan [15] for burning minor actinides, and in India [16], with a view to simplify the present thorium utilization scheme, were also made. In

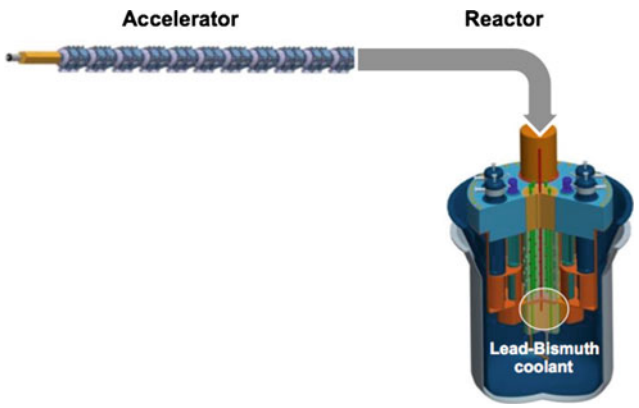

Fig. 3 The ADS scheme, illustrated by the MYRRHA prototype, taken from Aït Abderrahim's presentation [12], showing the accelerator inserting protons vertically onto a target, located at the center of a subcritical nuclear core, that produces neutrons by spallation

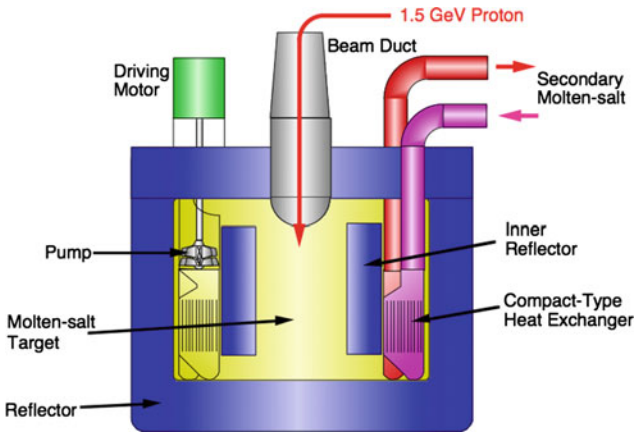

Fig. 4 Example of a molten salt ADS transmuter, as presented by Sasa [18]

Kharkov, Ukraine, the Kharkov Institute of Physics and Technology (KIPT) neutron source [17] is being completed and will be licensed for low enriched uranium (LEU).

Alternative ADS concepts were also presented at ThEC13, in particular the molten salt ADS by Rubbia [2], Sasa (Japan) [18] (Fig. 4), and Chai (Korea) [19].

ADS were also the subject of several other presentations addressing specific aspects, such as reactivity measurements by beam pulses, corrosion and material compatibility, etc.

As in the case of MSR R&D, the need for a coordinated worldwide effort is evident, to avoid duplication of tests, and to progress as quickly as possible from physics concepts to technological choices. The main issue for ADS today is the absence of a demonstrator. This is more of a political issue (funding) than a scientific one. This is clearly where the thorium community should concentrate its effort.

National and International Thorium Programs

India has by far the most advanced practical scheme for using thorium, based on three existing technologies, presented by Vijayan [20]:

- Use of a thorium blanket around heavy water reactors (HWR), CANDU, or light water reactors (LWR) to produce plutonium from the limited Indian uranium supply;
- Use of sodium-cooled uranium–plutonium fast reactors with a thorium blanket to breed ^{233}U. The first 500 MW_e prototype of a fast breeder nuclear reactor is being commissioned at the Madras Atomic Power Station, in Kalpakkam, India;
- Reprocessing the thorium blanket and manufacturing ^{233}U–Th fuel for advanced fast or thermal water reactors.

This strategy, which covers both the front-end and back-end of the fuel cycle, clearly works in principle. However, there remain concerns over the complexity of a scheme requiring the maintenance of three different reactor technologies. In addition, sustainability is questionable, and the present scheme does not solve the issue of nuclear waste.

National programs of Belgium, China, Czech Republic, France, India, Japan, Norway, Sweden, Turkey, and the UK, with different levels of involvement in thorium R&D, were presented. In Europe, the MYRRHA project dominates the scene, even though it is not yet fully funded, and does not include thorium fuel in its program. Nevertheless, it develops the basic elements needed for future thorium ADS.

Thorium Fuel Cycle

Two sessions were devoted to the thorium fuel cycle. The subject had already been briefly discussed by Rubbia, who presented new concepts in his introductory talk [2]. Presentations included various related subjects, such as commercial development of thorium fuel, licensing, back-end of the fuel cycle, pyro-electrical and aqueous reprocessing, etc.

Simulation of Thorium Systems

Ganesan [21] gave a very detailed status of the nuclear data related to thorium, and Gunsing et al. [22] described the impressive performance of the neutron time-of-flight (n_TOF) facility [23] at CERN, which provides the precise data (Fig. 5) needed to simulate new systems. Today, the physics of ADS is well understood and thorium systems can be simulated with high reliability.

Conclusion

At ThEC13, an impressive amount of R&D work was presented on various aspects of thorium technologies. R&D on thorium, regardless of its present specific purpose, should be encouraged. On the other hand, many activities are taking place in parallel worldwide, but not in a coherent way. International cooperation is a necessity in view of the importance of the energy issue, which is a challenge facing the whole world and requiring a global response.

Is it possible to converge on concrete projects, generating partnerships within international collaborations? In the round table discussion, Anil Kakodkar suggested that thorium could very well be a theme for international cooperation [24]. The CERN collaboration model should work for any new significant project. Developments common to MSR and ADS should be encouraged whenever possible (for instance, regarding the fuel cycle).

ThEC13 will be documented in a book of proceedings, which should contribute to this goal and making it a reference for the field of thorium technologies.

The goal of iThEC is to promote international cooperation in the development of thorium technologies, to provide abundant, safe, and clean energy for humankind. This is perhaps illustrated by one photo taken at the ThEC13 banquet (Fig. 6) of representatives from the two main countries, China and India, that are likely to define the energy future of our planet.

On behalf of iThEC and of the ThEC13 Organizing Committee, I would like to thank you all for having

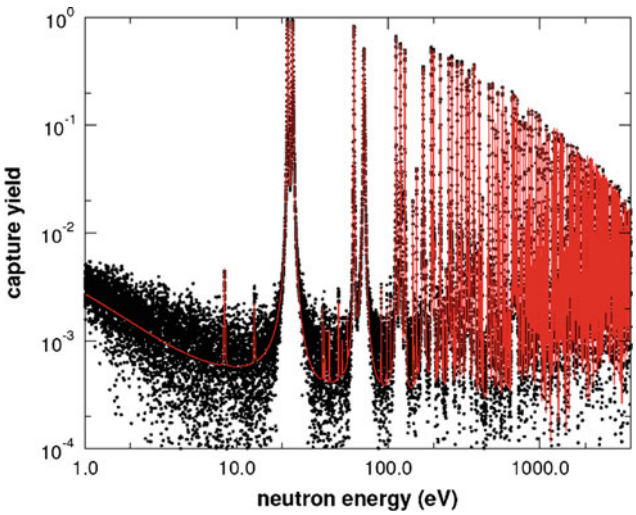

Fig. 5 Measurement of the neutron capture reaction rate on ^{232}Th at n_TOF [23], illustrating the high resolution with which resonances can be resolved. Taken from Gunsing et al. [22]

Fig. 6 Honjie Xu, China, Jean-Pierre Revol, iThEC, and Anil Kakodkar, India, at the ThEC13 banquet. *Photo* Credit Jean-Pierre Revol

contributed in such a professional way to this conference, by making presentations, by preparing posters, by participating in discussions, or simply by your presence here, in the CERN Globe of Science and Innovation.

Acknowledgments I would also like to thank CERN for providing an ideal environment for scientific discussions, and my colleagues from iThEC, who worked enthusiastically to make ThEC13 a great success.

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Part IX

Posters: National and International Thorium Programmes

Neutronic Analysis and Transmutation Performance of Th-Based Molten Salt Fuels

S.-I. Bak, J.S. Chai, J.P. Curbelo, S.W. Hong, E.J. In, Y. Kadi, C. Tenreiro, and Y.J. Zhou

Introduction

There have been many studies on lithium–beryllium fluoride compounds as the molten salt coolant for thorium-based molten salt reactors (MSR), but sodium- or lead-based chloride compounds have not been studied. Early conceptual studies of the sodium or lead chloride based fuels were made for an accelerator-driven molten salt reactor as a transuranics (TRU) burner in Japan in the 1990s [1], but there has been no study on the sodium or lead chloride molten salt with a thorium fuel, until now. Thorium is an attractive fuel material with low minor actinide (MA) production and can avoid plutonium production. MA production is a major concern for the management of spent nuclear fuels. An accelerator-driven system (ADS) was proposed for burning MA efficiently. If a critical reactor is used with a high fraction of MA, it would be very difficult to control criticality. However, a subcritical system can accept a large fraction of MA. The present work concerns a subcritical ADS with molten salt fuels using thorium and MA for investigating the potential of the system to breed ^{233}U and burn MA.

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Models for Reactor and Fuels

An accelerator-driven molten salt reactor system (AD-MSR) was proposed by the Japan Atomic Energy Agency (JAEA) in the 1990s [1]. The system was intended to incinerate MA mainly through fission induced by high-energy neutrons. The molten salt acts both as the primary coolant and as the fuel itself. It also acts as the target material for generating neutrons by spallation of heavy nuclei by a proton beam (Fig. 1).

In this work, we used the model proposed by JAEA [1] and studied the characteristics of different molten salt fuels based on lithium–beryllium fluoride, sodium chloride, and lead(II) chloride for ADS. These fuels have neutron spectra ranging from thermal to fast. We varied the composition of the fuel, which includes Th, U, Pu, and MA, and compared the results. We have calculated quantities such as the neutron spectra, breeding ratio, neutron multiplication factor, and transmutation ratio of MA. The calculations were performed with Energy Amplifier Monte Carlo (EA-MC) [2] and FLUKA [3, 4], and the results are compared for the different fuels shown in Table 1.

Results

To compare the characteristics of different molten salt reactors for Th–^{233}U fuel, the amount of ^{233}U in the composition shown in Table 1 was varied until $k_{\text{eff}} \approx 0.98$ was obtained. The reactors based on the LiF + BeF_2 fuels can achieve $k_{\text{eff}} \approx 0.98$ with a lower ^{233}U ratio than those based on the NaCl or $PbCl_2$ fuels as shown in Fig. 2. The subcritical cores based on NaCl or $PbCl_2$ fuels have fast neutron spectra with considerably higher neutron leakage component compared to the cores based on LiF + BeF_2.

The breeding of ^{233}U from ^{232}Th is efficient in the epithermal neutron energy region where ^{232}Th has large

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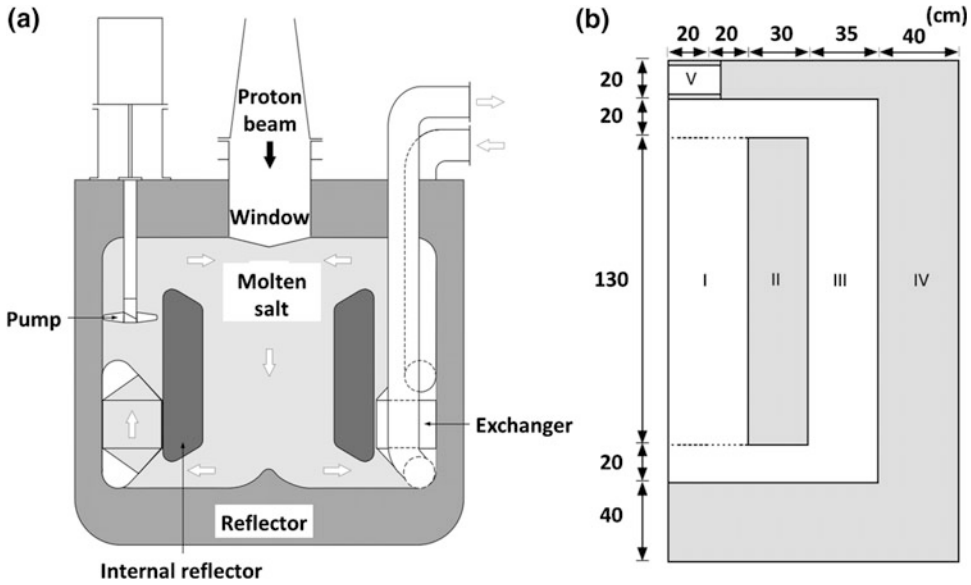

Fig. 1 Concept of the JAEA molten salt ADS (**a**) and a simplified geometry for the simulation (**b**) [1]

Table 1 Molten salt fuels used in this work

Fuel	Composition	Reference
Li–Be fuel No. 1	**64 % ^{7}LiF, 18 % BeF_2**, 18 % (Th, ^{233}U, Pu, MA) fluoride	JAEA [5]
Li–Be fuel No. 2	**73 % ^{7}LiF, 15 % BeF_2**, 12 % (Th, ^{233}U, Pu, MA) fluoride	ORNL [6]
Na fuel	**64 % NaCl**, 36 % (Th, ^{233}U, Pu, MA) chloride	JAEA [1]
Pb fuel	**64 % $PbCl_2$**, 36 % (Th, ^{233}U, Pu, MA) chloride	JAEA [1]

The coolants written in *boldface* were not varied. The fluoride or chloride fuel compositions written in normal font were varied for comparison of the results

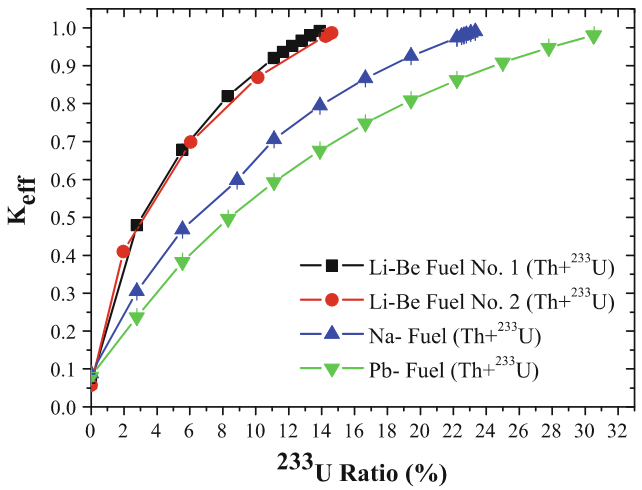

Fig. 2 Neutron multiplication factor as a function of ^{233}U ratio for different fuels, which include only Th and U in this case. The ^{233}U ratio refers to $^{233}U/(^{233}U + ^{232}Th) * 100$ %

neutron capture cross sections; thus, the LiF + BeF_2 based fuel is better than other fuels for breeding. The neutron capture cross sections are larger for chlorine isotopes, particularly at low neutron energies, than for fluorine; thus, chloride-based fuels have disadvantages from the point of view of neutron economy.

The temperature coefficient is estimated for reactor safety. All the fuels have negative values of temperature coefficient and the absolute value of the temperature coefficients for the LiF + BeF_2 fuel is found to be almost ten times larger than those for other fuels (details not shown here).

To compare the transmutation potential of MA in different fuels, ^{237}Np, ^{241}Am, ^{243}Am, and ^{244}Cm were considered, which are the main MA isotopes in spent fuels. The fission per capture (F/C) ratios for different MA in the molten salt burner with pure MA, without U or Th, are shown in Fig. 3a. The transmutation of the MA is more efficient in the fast neutron region; the NaCl and the $PbCl_2$ based fuels have a higher F/C ratio than LiF + BeF_2 based fuels. Figure 3b shows the F/C ratios of $PbCl_2$ based fuels when they include Pu or Th + Pu. By adding Pu to pure MA fuels, the F/C ratios remain more or less the same. The F/C ratio for the Th + Pu fuel is lower than for the Pu fuel. The conversion ratio of Th to ^{233}U and the amount of MA transmutation in different molten salt fuels are shown in Table 2.

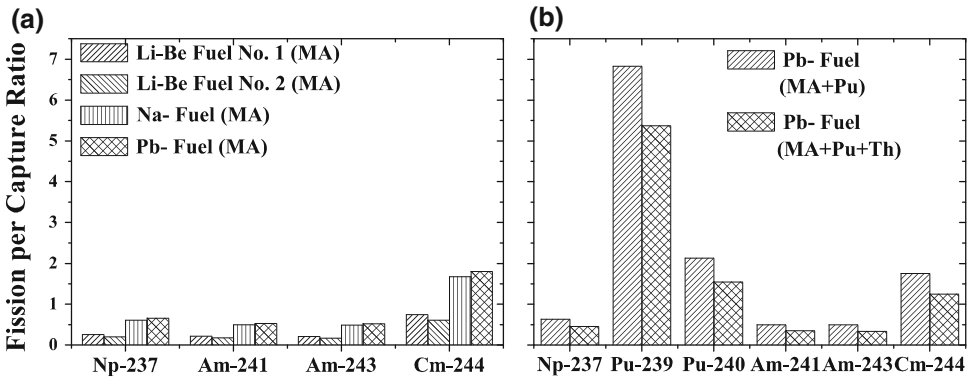

Fig. 3 **a** F/C ratio of different minor actinides when the fuels contain only MA. **b** F/C ratios of different actinides when the $PbCl_2$ based fuel is used and Pu or Pu/Th is added to the MA fuels

Table 2 The conversion ratios of Th and the amount of MA transmutation for different molten salt fuels

Fuel composition (% in mole)	Conversion ratio	MA transmutation (kg/TWd/HMt)
$^{7}LiF + BeF_2 + {}^{232}ThF_4 + (Pu + MA)F_3$ (64:18:12.6:5.4)	0.41	2.5
$^{7}LiF + BeF_2 + {}^{232}ThF_4 + (Pu + MA)F_3$ (72.8:15:8.1:4.2)	0.35	2.8
$NaCl + {}^{232}ThCl_3 + (Pu + MA)Cl_3$ (64:24.5:11.5)	0.29	4.5
$PbCl_2 + {}^{232}ThCl_3 + (Pu + MA)Cl_3$ (64:21.4:14.6)	0.19	7.1

Conversion ratios are larger and MA transmutations are smaller for $LiF + BeF_2$ based fuels, which have more thermal neutrons

Conclusion

Comparisons of different molten salt fuels based on $LiF + BeF_2$, NaCl, and $PbCl_2$ salts have been made to gauge their effect on breeding of ^{233}U from Th and on the efficiency in burning MA. When thorium fuel is used, the $LiF + BeF_2$ based fuel, which has a thermal/epithermal neutron spectrum, has a better breeding ratio of ^{233}U from Th, a higher neutron multiplication factor, and a more negative temperature coefficient. However, the NaCl or $PbCl_2$ based fuels are better for eliminating MA, because of the hard neutron spectrum. The sodium or lead chloride molten salt may deserve further investigation as a molten salt together with a thorium fuel if the issue of MA burning is considered a priority.

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Neutron Irradiation of Thorium-Based Fuels: Comparison Between Accelerator-Driven Systems and Fusion–Fission Systems

Carlos E. Velasquez, Graiciany de P. Barros, Claubia Pereira, Maria Auxiliadora F. Veloso, and Antonella L. Costa

Abstract

Accelerator-driven systems (ADS) and fusion–fission systems are investigated for long-lived fission product (LLFP) transmutation and fuel regeneration. The aim is to investigate nuclear fuel evolution and the neutronic parameters under neutron irradiation. Each system was loaded with thorium-based fuel; both systems were designed to have an initial criticality around $k_{eff} \approx 0.99$ due to safety terms. The criticality and depletion of both systems were analysed for a simulated period of 10 years. The simulations were performed in the MONTEBURNS code that links Monte Carlo N-Particle Transport Code (MCNP) to the radioactive decay burnup code ORIGEN2. The results indicate which of system produces higher fissile isotopes rate, as well as the most suitable system for achieving transmutation. The mass production from the nuclides that influence the differences in criticality are presented.

Introduction

Two ideal neutron sources from an accelerator-driven system (ADS) and a fusion–fission system are located in the central part of a subcritical system. The goal is to compare the neutronic and depletion behaviour during the burnup of the system under the irradiation of these two different neutron spectra using the same power in the core.

Methodology

Figure 1 shows the subcritical system modelled. The evaluated neutron source was placed in the central part, with dimensions of 0.9 cm radius and 38 cm height. The core is a cylinder of 6.0 m^3 filled with a hexagonal lattice loaded with a mixture of $^{232}ThO_2$ + 15 % $^{233}UO_2$. The total power during the burnup was 430 MW for both systems. The simulations were performed by using the MONTEBURNS code, which links the Monte Carlo transport code (MCNP) with the radioactive decay and burnup code ORIGEN2.

Results

Figure 2 shows the multiplication factor for the different neutron sources. The drop of k_{eff} is greater for the ADS neutron source than for the neutron fusion source. To understand this behaviour, some nuclides were evaluated. The causes of the differences in the k_{eff} behaviour are probably connected with ^{232}Th transmutation to other actinides. Figure 3 shows the ^{232}Th and ^{233}U mass variations during the burn time for both sources.

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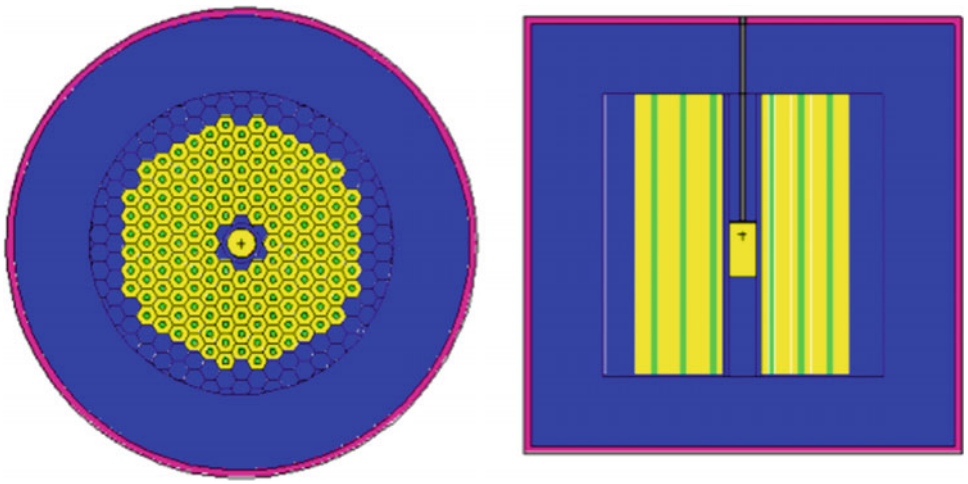

Fig. 1 Horizontal and vertical ADS cross sections

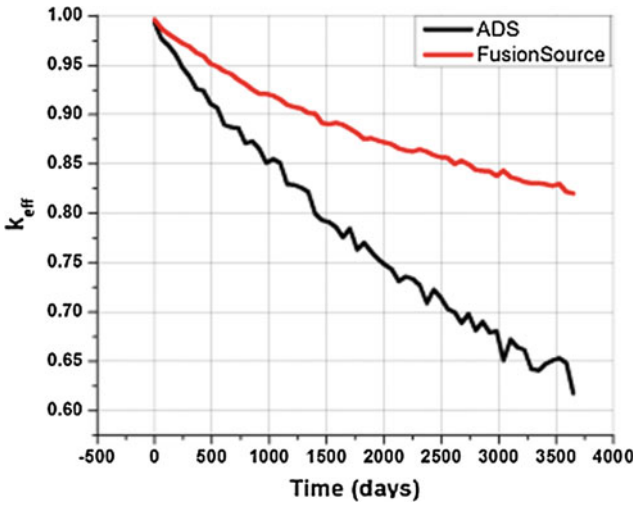

Fig. 2 k_{eff} evolution

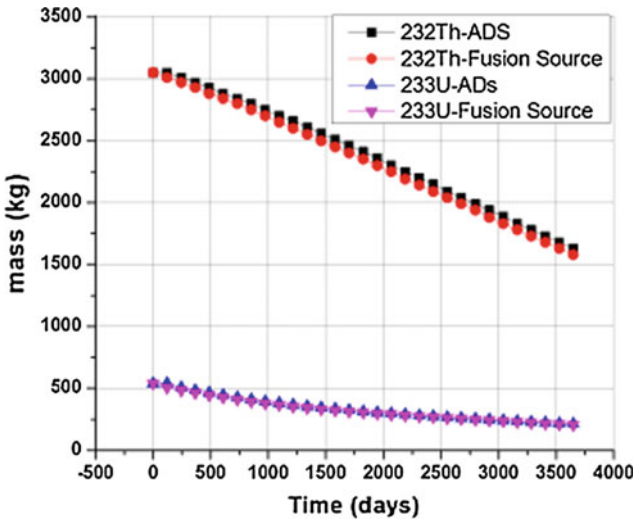

Fig. 3 Mass variation

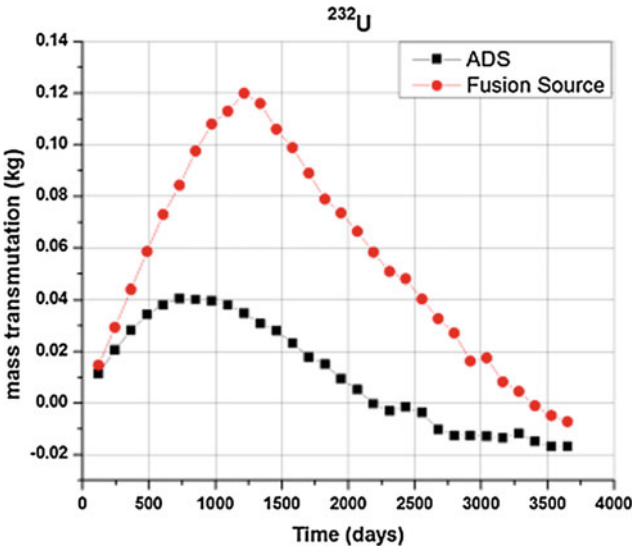

Fig. 4 ^{232}U mass variation

Figure 4 shows the amount of production and transmutation of ^{232}U during the irradiation. This difference in mass during the burn time could be a reason for the k_{eff} differences. It can be related to the production and transmutation of ^{232}U, which has a fission cross section comparable with ^{235}U, as shown in Fig. 5.

The ADS reaches a ^{232}U maximum production of 0.04 kg after 2 years, whereas the fusion source reaches a maximum of 0.12 kg after 3 years. Using the fusion source produces more ^{232}U than fissions for the first 3 years, then the role is inverted, which explains the drop in the curve after this time. Another observation is that the fission-to-capture ratio is a little bit higher for the fusion source, as shown in Fig. 6, and the ^{231}Pa production is higher for the fusion source, as shown in Fig. 7. These facts contribute to the difference in the multiplication factor (k_{eff}).

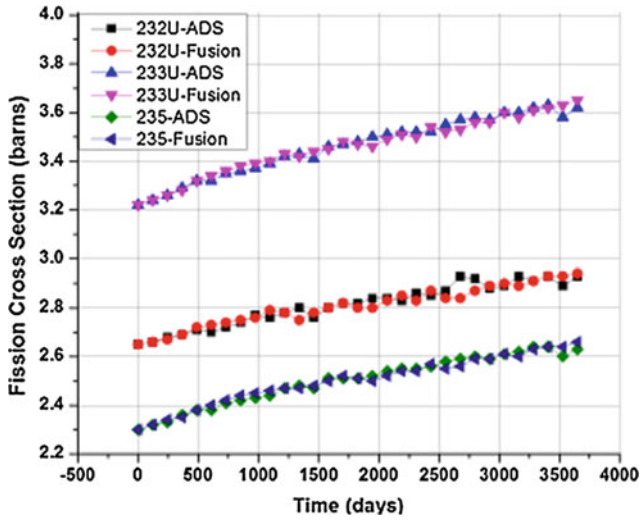

Fig. 5 Fission cross sections

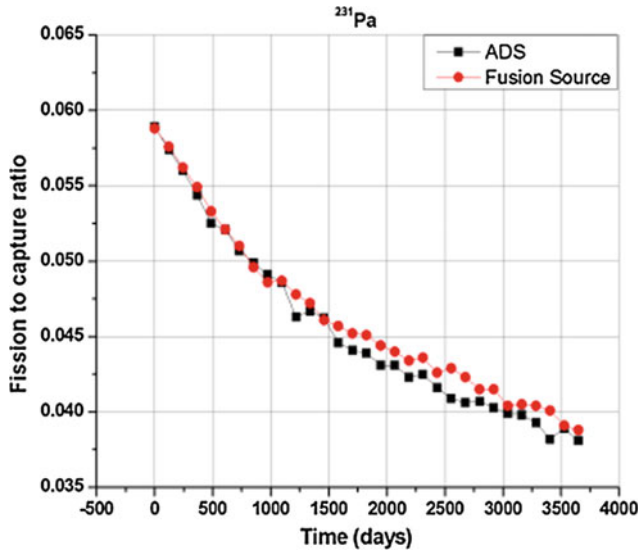

Fig. 6 Fission-to-capture ratio

Conclusion

The differences in k_{eff} are explained based on the mass production of two actinides: ^{232}U and ^{231}Pa. These differences might be explained in turn by the differences in the neutron spectra of each system, which induces variations in the production or transmutation of these actinides.

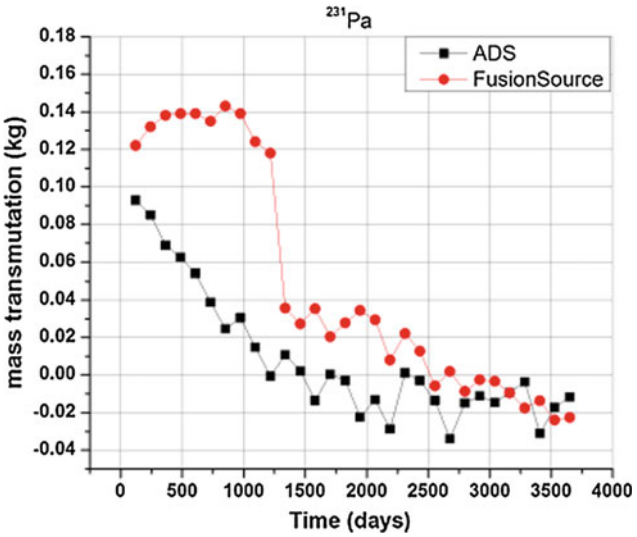

Fig. 7 ^{231}Pa mass variation

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Part X
Posters: Innovative Thorium-Reactor Concepts

Generation IV Reactor Cooling by "Gas-Lift"

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Introduction

In the case of long-term operation, heat from reactor cores could be removed by natural convection. "Gas-lift" can be used to intensify it.

"Gas-lift" is used to extract heat in two ways:

- Continuous supply of flowing gas;
- Periodic injection of gas, which is effective for gas species extraction.

In the case of a nuclear reactor, only a continuous supply of gas can be used. He or Ar can be used as the gas. The use of "gas-lift" is also possible for molten salt reactors (MSR) to clean the fuel–coolant fluorides of gaseous fission products, thereby eliminating problems with "xenon fission product poisoning" in conventional reactors. For high-temperature reactors with mechanical pumps there are material problems (the materials are not yet licensed by the American Society of Mechanical Engineers (ASME) for these temperatures and have not been tested for durability and corrosion). "Gas-lift" reactors have no moving parts. The flow in the reactor using the "Gas-lift" can be viewed on Fig. 1.

Dispersed bubble flow is the most appropriate model for calculating "gas-lift" for nuclear reactors, because it allows the maximum intensification of the natural flow and ensures operation without shocks. Determination of individual types of two-phase flow "gas-lift" in the MSR must be verified based on similarity analysis. Therefore, we have built a two-phase demonstrator (TFD) using water and air.

The problem is to determine the W_{∞} and W_g (mass flow) so that we can operate in the area of the dispersed bubble flow. For liquids, this means that the superficial velocity (liquid in the flow cross section) $v_{sl} > 6$ m/s, while the superficial gas velocity $v_{sg} < 8$ m/s (Fig. 2).

According to different authors, the parameters for the area can be distinguished.

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Description of the Experimental Device

First, we set the corresponding diagram of the experimental apparatus MSR primary circuit.

The experimental device consists of a series of construction parts that have been named after the parts of the primary circuit that they represent. The model of the whole device, including the description of its main parts, is shown in Fig. 3.

The active zone is represented here by a cylindrical metal container of 360 mm diameter and height of 300 mm. There are conical parts in the upper section to reduce pressure losses. The heat source in the model core is provided by a 2000 W heating unit. It has to be removable to enable assembly and potential exchange of the heating unit, and it is connected to the cylindrical part of the core using the flange and twelve screws. A ball valve is bolted there for filling and drainage in the bottom-most part of the core.

A short tube with a diameter of 70 mm with ten vents for the air intake is welded above the cones. One pressure pipe is welded, by inserted reduction, to every nut, which transfers the compressed air from the compressor to the area of the tractive cylinder intake. One point of the measurements is to determine the influence of the size of the supplied air on the water flow rate in the TFD water loop. Therefore, it is essential to change the diameter of individual vents for air intake easily to control the size of the created bubbles in the tractive cylinder. Changeable brass inserts are used for this purpose. The reduction—that is, actually, a drilled-through screw with one internal and one external threading—and the changeable brass insert determine the size of the air intake vent. There are a number of changeable inserts available with vent diameters of 0.5, 1 and 2 mm. If we do not use the

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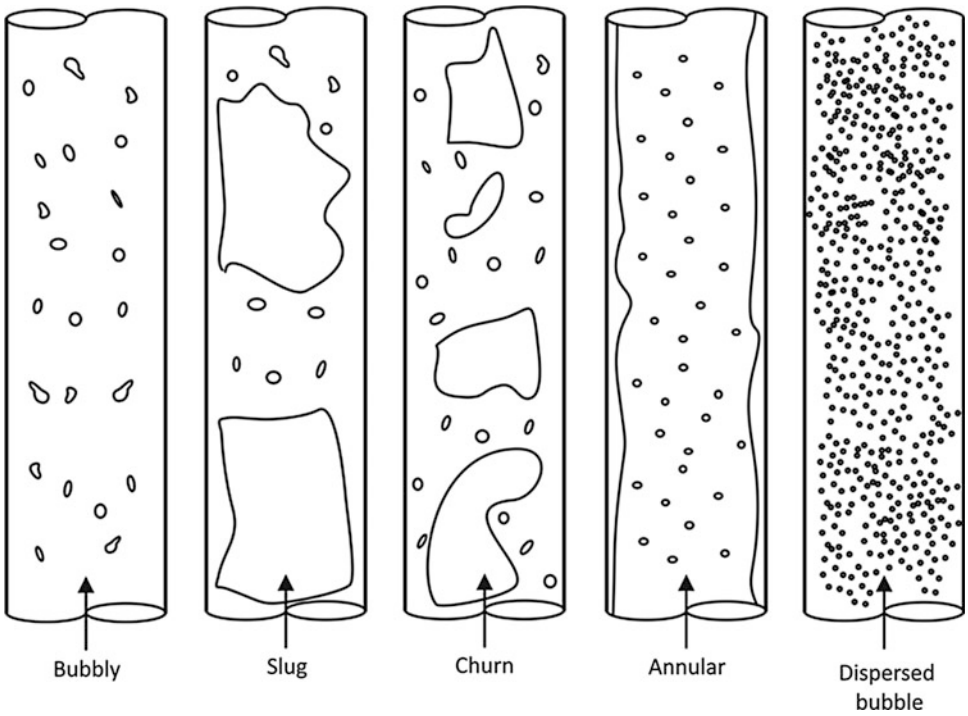

Fig. 1 Flow patterns of "Gas/Lift" [1]

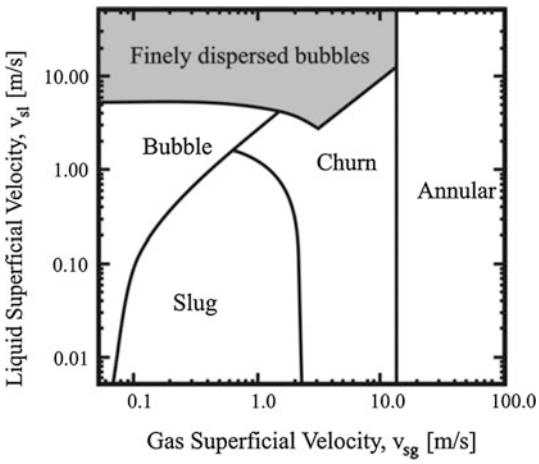

Fig. 2 Map of distribution of the flow types of a two-phase flow (Taitel) [2]

insert in the reduction, the diameter of the entry vent is 7 mm; therefore, we have four options of air intake vent size.

The tractive cylinder is made of a plexiglass pipe of a circular cross section with an inside diameter of 70 mm and a wall thickness of 5 mm. Its height is 1200 mm. It is connected (using the flange) to a metal pipe with the previously mentioned ten vents that are used for the air intake.

Conditions for the Use of "Gas-Lift"

Specification of the parameters of the core must be based on the performance requirements for the MSR. The performance of the MSR will be used either for production of electricity, or for technological purposes, or a combination. The volume of the core used in the center determines the specific power, q.

- natural convection: $q \approx 10$ [MW/m^3];
- "gas-lift": $q \approx 25$–40 [MW/m^3];
- forced convection: $q \approx 50$ [MW/m^3].

The average tensile strength of the 'chimney', D, is determined from the critical condition ($k_{\text{eff}} < 0.95$). We will consider an infinitely tall cylinder surrounded by a graphite reflector. Dimension D can be adjusted (while maintaining subcriticality) according to the flow velocity.

It is necessary to structurally arrange the primary circuit of the reactor and our TFD, so that the total pressure losses

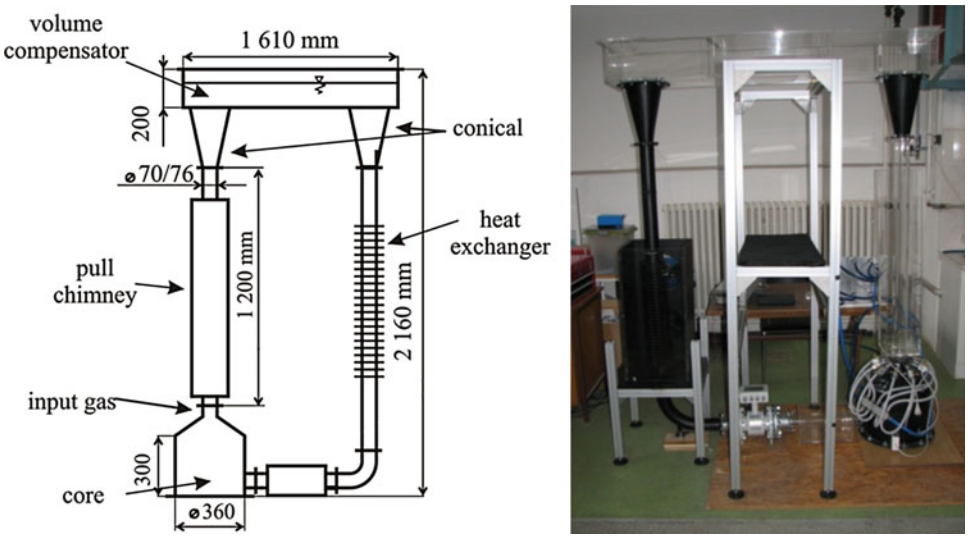

Fig. 3 Two-phase flow demonstrator, scheme (*left*) and real (*right*)

are minimal. The effectiveness of natural flow and "gas-lift" are strongly interdependent.

Conclusion

The model in the first stage is used for water and air dispersion modeling using bubble-type two-phase flow, which is advantageous in terms of the operation of nuclear reactors. Measurements on the model will be compared with the Chexal–Lellouche method [3].

The Chexal–Lellouche [4] Void Fraction Model was based on the model of Zuber and Findlay [5] and summarizes the possibilities of a two-phase flow model for cases with water vapor, water, air, and liquids used in refrigeration technology. The model describes vertical, horizontal, and angled flows.

Acknowledgments The presented research is supported by the project CZ.1.05/2.1.00/03.0108 (Sustainable Energy, SUSEN) and student project SGS-2014-070 (Increasing the efficiency, reliability and durability of power system devices 3).

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The Bumpy Road to a Technology Strategy Board Grant

Trevor Griffiths and Jasper Tomlinson

Abstract

This presentation by Trevor Griffiths and Jasper Tomlinson describes the steps that have led up to an opportunity for a micro-enterprise, Energy Process Developments Ltd, to set up in the UK in order to undertake a feasibility study for a pilot-scale nuclear molten salt reactor. The task essentially is to choose between the various options best suited to demonstrating—for the public, for decision makers, and for all interested parties—a working pilot-scale reactor. This reactor can subsequently be scaled up to an industrial prototype that will lead the way to a new industry-standard reactor for nuclear power. The ultimate fuel of choice will be thorium, both for its radiochemical advantages, and for its long-term sustainability.

The Bumpy Road to a Technology Strategy Board Grant

On the 20th June 2013, we learned of a Technology Strategy Board (TSB) competition for funding a feasibility study. We registered our intent the following day.

This particular TSB competition—Meeting the energy challenge: emerging energy technologies—is disbursing £3 million for feasibility studies. The competition had opened on 10th June with a 24th July deadline for submissions. EU rules allow the government to provide up to 75 % of the costs of such studies, leaving the bidder to provide the remaining 25 %.

The bidder had to be a small company, with or without partners. The bidder had to provide, within about a month, a credible limited liability company, a sum of £25,000 to provide the requisite 25 % for a £100,000 feasibility study, as well as a credible core group of engineers from which to develop a team capable, on a minimum budget, of conducting the study.

Most urgently, the bidder should provide answers to ten questions set by the TSB. The four fundable activities—all emerging energy technologies—could include either the integration of energy storage into a system, a novel low-carbon heat source, novel thermodynamics, or innovations that can deliver a step-change in the cost or performance of low-carbon energy. The fixed set of questions had been formulated for any or all of the fundable activities. These were not the questions we would have asked ourselves in order to clarify our intentions in respect to this fourth activity. However, each question required a response, within the exact limits of the space provided, to gain 10 points from the total of 100. We were awarded just 58.6 % for our attempt, a disastrous outcome that we are now discussing with the Department of Energy and Climate Change (DECC), who are rather well informed and are responsible for provision of whatever is needed in the energy sector.

Trevor's commitment to this initiative arises from his long involvement with molten salt chemistry. This interest was firmly established by Trevor's first post-doctoral job at the Oak Ridge National Laboratory (ORNL) in the 1960s when the molten salt reactor experiment (MSRE) was still at the heart of the Laboratory's activities. In a long career in chemistry research and teaching, Trevor has made many contributions to molten salt chemistry and to the community of colleagues involved with this particular topic. About one third of the content of a poster displayed at iThEC13 was based on a molten salt reactor presentation Trevor gave to the Molten Salt Discussion Group of the Royal Society of Chemistry.

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Jasper's determination to see a pilot-scale molten salt reactor in operation at the soonest opportunity emerged in 2008 from a meeting with two senior civil servants who had responsibilities for a (deplorable) Department for Business, Enterprise and Regulatory Reform (BERR) 2008 White Paper on Nuclear Power. Arranging this meeting had been the outcome of an exchange of letters with Gordon Brown following a fairly ludicrous close encounter at a 'focus group' gathering in Bermondsey. The civil servants languidly explained that they had no responsibility for either technology or financing in the provision of nuclear power: Industry Will Decide was the deeply irresponsible mantra, both in Brown's response to my queries and embedded in the White Paper.

Jasper was asked by Andreas Norlin to be the UK representative for the first IThEO International Conference in London (2010). Among many calls made by Jasper to promote this first IThEO event, there was one that had the effect of strengthening his determination about a pilot-scale plant agenda. In a call to the Nuclear Industries Association, Jasper suggested that they might like to alert their members to the coming conference. No, was the clear reply. Why is that then? The even-toned response: Because you are the enemy!

Immediate Disappointment

On 30th September, we were informed that we had failed to secure funding. 'Feedback'—the written comments of the independent assessors—followed at the end of October. This feedback, which includes conflicting comments from individual assessors, together with the bid submitted, can be copied to anyone interested by sending a request email to jaspertomlinson@gmail.com.

Objectives

The objectives of the proposed feasibility study were spelt out in the submission, namely:

1. Provision of a pilot-scale demonstration, working molten salt reactor to enable decision makers, the media, and the wider public to become aware of the main characteristics of alternative nuclear power plants, engineered to produce power competitive with coal and to operate as a normal extension of chemical engineering capabilities;
2. To provide a test bed for developing—for a coming industry-standard nuclear power technology—materials and designs for main components such as pumps, valves, heat exchangers, etc., and for trying out alternative configurations in respect to fuels and to uses for the thermal energy output;
3. To indicate which alternative configuration, in terms of size, type, and scope, gives the best outcome in terms of regulatory constraints, cost, and selected site, to achieve the first two objectives;
4. To provide the terms of reference for an engineering design study for the chosen alternative.

The central objective for the development of new reactor technology is provision of sustainable energy competitive with fossil fuels. In this way, a Second Nuclear Era can come into being, the first, after a lengthy trial, having proved a disappointment. Robert Hargraves, in his recent book, Thorium—Energy Cheaper than Coal, gives a useful summary of each of the available energy technologies with a standardized costing based on investment cost, operating cost, fuel cost, each modified as required to take account of load factors, etc. He emphasizes the absolute need to engineer any new technology to produce energy cheaper than, say, coal.

The very next step is to build molten salt reactors, not just to conduct more research about their viability. Ongoing research and development will continue side by side with actual implementation of this technology—the necessary investigations were mainly completed at Oak Ridge more than half a century ago.

To learn what happened next visit http://www.energyprocessdevelopments.com

Experimental Activities on Heavy Liquid Metal Thermo-Hydraulics

Julio César Pacio and Thomas Wetzel

Abstract

Heavy liquid metals, such as Pb and lead–bismuth eutectic (LBE), are fluids of interest for applications in fast reactors and accelerator-driven systems (ADS). For the development of such systems, thermal-hydraulics is a key discipline to be further investigated. The low Prandtl number of these fluids makes this problem particularly challenging and reliable experimental data is essential. In this work, an overview of the experimental activities at the Karlsruhe Liquid Metal Laboratory is presented.

Introduction

For over two decades, Karlsruhe Liquid Metal Laboratory at the Karlsruhe Institute of Technology (KIT-KALLA) has been investigating the developement of liquid metal technologies. Since the turn of the century, with the construction of large lead–bismuth eutectic (LBE) thermo-hydraulic loops, detailed experiments have been performed under reactor-like operating conditions. In this work, two sets of experiments are briefly overviewed, namely the study of spallation targets and rod-bundles cooling.

Spallation Targets for Accelerator-Driven Systems (ADS)

As a key component for ADS, and considering its extreme operating conditions, proof-of-principle and target-coolability tests, such as the two described below, are essential for the development of these systems.

- For the MEGAPIE initiative, a prototypical (nearly 1:1 scale of the lower part of the target) heated-jet test was performed at KALLA. Several flow patterns were observed for this geometry, and a region of stable operating conditions under which sufficient cooling is provided was identified [1].
- A free-surface windowless concept was investigated in the framework of the European THINS project [2]. The evolution of the target surface at different velocities was identified in the experiments, which provided a proof-of-concept as well as valuable data for the validation of numerical tools.

Heat Transfer in Fuel-Element-Representative Geometries

Due to the particular physical properties of heavy liquid metals, specific experiments for studying the core thermo-hydraulics are required. In that context, three campaigns of increased complexity were undertaken at KALLA, described below. In all cases, reactor-like conditions of heat fluxes up to 1.0 MW m^{-2} were used.

- First, a single heated rod in an annular channel was studied, including detailed measurements of temperature and velocity profiles in both the developing and fully

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developed regions [3]. This geometry gives an accurate representation of the flow behavior in the sub-channels of rod bundles with a large pitch-to-diameter ratio. Both forced- and mixed-convection regimes were investigated.

- Second, a 19-rod hexagon bundle with grid spacers was investigated in forced-convective LBE flow, as a representative geometry of reactor fuel elements. The pressure drop, heat transfer coefficients, hot-spot factors, and sub-channel center temperatures were studied for a wide range of conditions, yielding an extensive database of reliable experimental information [4].
- Third, a new test section is currently under construction, consisting of a 19-rod bundle with wire-wrap spacers. For this test section, two first-of-a-kind LBE experiments are envisaged, namely, reference heat transfer tests and analysis of the effects of blockages on the flow. Detailed temperature measurements are to be considered for obtaining a better understanding of these type of flows.

Conclusion

In summary, the experiences at KALLA with LBE flow experiments indicate that these undertakings, though challenging, are essential as proof-of-concepts as well as for validating the predictions of numerical tools.

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Combined Effect of Irradiation and Molten Fluoride Salt on Ni-Based Alloys

A.S. Bakai, K.V. Kovtun, and L.N. Davydov

Introduction

The Hastelloy-N was successfully used in the molten salt reactor (MSR) experiment [1]. Since then, corrosion tests of Ni–Mo alloys of different compositions have been performed in loops without irradiation [2, 3]. In this presentation, the results of the mechanical and corrosion tests of Ni–Mo alloys of two compositions (with and without Nb) under and without electron irradiation at T = 660 °C are briefly reviewed.

Materials and Electron Irradiation Test Facility (EITF)

A purified ZrF_4 (51 mol%)–NaF (49 mol%) fluoride salt mixture, which is molten at 650 °C, was used in the experiments. La was added in one experiment. Two Hastelloy alloy compositions (A and B, Table 1) were manufactured from components of high purity [4, pp. 40–47]. The alloys were homogenized at 1100 °C for 1 h and aged at 675 °C for 50 h.

The EITF was constructed and built at the Linac-10 electron linear accelerator in NSC KIPT. An electron beam of 10 MeV energy and up to 1 mA average current (up to 1 kW of power) scanned over a special assembly with 16 ampoules made of the C–C composite. Each ampoule contained samples of Ni–Mo alloys immersed in molten salt. The assembly was placed in a water-cooled chamber under an Ar atmosphere.

Mechanical Tests

Along with the standard mechanical tests [4, pp. 53–61], microindentations and nanoindentations of the materials in the vicinity of the surface and in the bulk were performed [4, pp. 67–73]. The yield strength, $\sigma_{0.2}$, and ultimate tensile stress, σ_B, depend on the deposited energy, E_{dep}. Macroscopic mechanical properties change according to the influence of the molten fluoride salts and irradiation. These changes are caused by microstructural alterations, primarily by alterations of the grain boundaries and dislocation system, as well as by heterogeneities in the composition induced by radiation. The alloy A becomes softer due to irradiation. The plasticity of the irradiated alloy (15 %) is considerably larger than that of the non-irradiated specimen (9 %), but remains close to its plasticity in the initial state (14 %). Dependence of $\sigma_{0.2}$ on E_{dep} is less pronounced at 650 °C. Unlike alloy A, for alloy B the $\sigma_{0.2}$ dependence on E_{dep} is small at T = 20 °C and appears to be considerable at T = 650 °C.

Microstructural and Composition Investigations

The microstructure was characterized by high resolution transition electron microscopy (TEM) with a Philips CM30 microscope operating at 300 kV. Macroscopic precipitation develops in both alloys after 700 h irradiation in molten salt at T = 650 °C. Alloy B irradiated in molten salt shows much larger grain boundary precipitation compared with that of alloy A [4, pp. 82–88]. Not only density, but also the size of the precipitates at the grain boundaries of alloy A are considerably less than in alloy B. In alloy A, precipitates are composed mainly of Ni–Ti–Si. The grain boundaries of alloy B are laden with precipitates of 2–7 μm in size and many of them include a high content of Y (up to 30 at.%).

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Table 1 Composition (wt%) of the investigated Hastelloy type Ni–Mo alloys

Alloy	Ni	Mo	Cr	Fe	Al	Mn	Nb	Ti	Si	Y	C
A	78.15	11.70	6.70	1.50	0.83	0.50	–	0.47	0.15	–	–
B	78.10	11.70	6.20	1.50	0.83	0.50	0.50	0.47	0.15	0.05	–

Corrosion Tests

The corrosion of alloys A and B in the molten ZrF_4–NaF mixture without and under irradiation has been studied by cyclic voltammetry, X-ray analysis, scanning electron microscopy, and metallography [4, pp. 62–73]. Without irradiation, the corrosion rate saturates at a rather low level $\sim 2 \times 10^{-4}$ mm/year. As a result of irradiation, the corrosion rates increase considerably. The corrosion rates of both alloys are ~ 0.2 mm/y at E_{dep} = 6192 eV/at. At a much smaller deposited energy, E_{dep} = 121 eV/at, the corrosion rate of alloy A is half that at the higher E_{dep} value, but alloy B has a corrosion rate two orders of magnitude smaller. The addition of Nb and Y not only depresses the inter-granular corrosion, but also makes the corrosion rate sensitive to the deposited energy. The addition of La (10.8 wt%) in the salt changes the corrosion rate and nanohardness of alloy A [4, pp. 62–66] considerably.

Conclusion

EITF is an efficient device for corrosion tests of MSR construction materials thanks to electron irradiation capable of reproducing realistic irradiation conditions. Ni–Mo alloys A and B have acceptable corrosion resistance in molten salt at 650 °C. After electron irradiation in EITF for 700 h, the estimated corrosion rate was about 0.1 mm/year. The corrosion mode and resistance is rather sensitive to Nb and Y concentrations, and presumably to other dopants. Compositional optimization is needed to minimize the corrosion rate. Direct reactor tests of the alloys are desirable to highlight the usefulness of test devices such as EITF.

Acknowledgments This research was partially supported by STCU within Project #294. These results have been partially published in a special issue on the physics of radiation effects and radiation materials science [4].

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Part XI
Posters: Thorium-Fuel Cycle and Transmutation

High-Conversion Th-U-233 Fuel Cycle for Current Generation in PWRs

D. Baldova and E. Fridman

Abstract

The main goal of this study is to evaluate the operational feasibility of the high-conversion (HC) Th–U-233 pressurized water reactor (PWR) core through performing 3D fully coupled neutronic and thermal-hydraulic (T-H) analysis. The proposed HC core model consists of 193 typical 17 × 17 PWR fuel assemblies. Each fuel assembly is subdivided into two regions designated as seed and blanket. The central seed region, which has high U-233 content, serves as a neutron supplier for the peripheral blanket region. The blanket region consists mostly of Th-232 and acts as a U-233 breeder.

Introduction

This paper presents the results of the 3D full core neutronic and thermal-hydraulic (T-H) analysis of a pressurized water reactor (PWR) core fully loaded with high conversion Th–U-233 seed–blanket (SB) fuel assemblies (Fig. 1). The 3D full core calculations were performed with the DYN3D core [1], employing the two-group cross sections (XS) generated by the code HELIOS [2]. The XS were generated as functions of burnup, fuel and moderator temperatures, and soluble boron levels. The full core analysis was done for different HC PWR core cases that varied in the initial seed enrichment, in the average power density level, and in the inlet coolant temperature (T_{in}). All the HC PWR core cases were analyzed by assuming three-bath fuel management. As an alternative to the conversion ratio, the instant-to-initial fissile inventory ratio (FIR) was evaluated. The T-H behavior was analyzed through an evaluation of the limiting T-H parameters including the minimum departure from nucleate boiling ratio (MDNBR) and fuel central line temperature (T_{CL}). It is required that the maximum T_{CL} should be 200 °C below the melting point of the Th–U oxide, which is 3100 °C. The local heat flux should be lower than the critical heat flux by 1.3, which is the MDNBR value typically used in the PWR thermal analysis.

Results

The results presented in Table 1 suggest that the HC Th–U-233 PWR core at power density levels of 70 and 65 W/cm^3 comply with the safety limits: both could potentially be operated. However, the FIR in both cases is lower than unity. The HC Th–U-233 PWR core operated at a power density of 60 W/cm^3 and with the higher inlet coolant temperature of 275 °C meets both the safety requirements and self-sustainability (i.e., FIR equal to unity). The HC Th–U-233 core with the lowest power density of 55 W/cm^3 could be potentially operated, too. However, it can be seen that the reduction in the power density to 60 W/cm^3 is sufficient to

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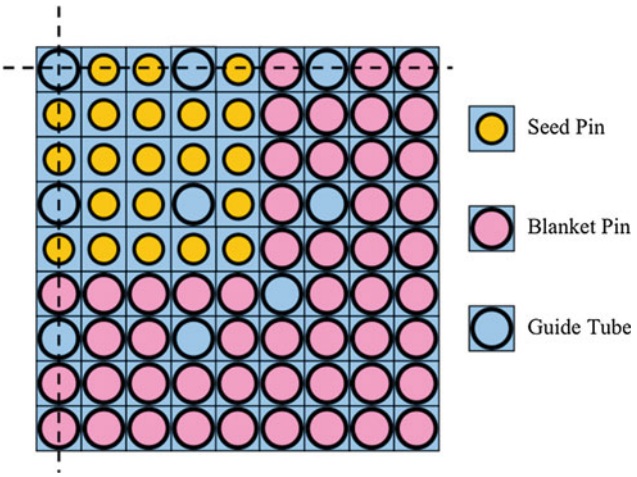

Fig. 1 1/4th of the 17 × 17 SB fuel assembly

Table 1 Results of 3D full core analysis at BOC

Power density (W/cm^3)	T_{in} (°C)	FIR at discharge	MDNBR	Max. T_{CL} (°C)
70	265	0.98	1.46	2110
65	265	0.99	1.54	1957
60	275	1.00	1.52	1870
55	289	1.01	1.51	1774

ensure the safe operation and to achieve a self-sustainable fuel cycle. The results are presented only for the Beginning of Cycle (BOC) because it is the most restrictive point as in the SB fuel assembly most of the power is initially generated in seed. Therefore, the power peak between the seed and the blanket is the highest at the BOC.

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Thorium and Transuranic (TRU) Advanced Fuel Cycle: An Option for Brazilian Nuclear Plants

Fabiana B.A. Monteiro, Carlos E. Velasquez, Rochkhudson B. de Faria, Diego M. de Almeida, Ângela Fortini, Claubia Pereira, Maria Auxiliadora F. Veloso, and Antonella L. Costa

Abstract

A typical pressurised water reactor (PWR) fuel element containing TRU–Th fuel cycle was simulated. The study analysed the behaviour of thorium insertion spiked with reprocessed fuel by considering different enrichments that varied from 5.5 to 7.0 %. The reprocessed fuels were obtained by using the ORIGEN 2.1 code from a burnt PWR standard fuel (33,000 MWd/tHM burned), with 3.1 % of initial enrichment, which has remained in the cooling pool for five years. The k_{eff}, hardening spectrum, and the fuel evolution during burnup were evaluated. This study was performed by using the SCALE 6.0 code.

Introduction

A study of thorium reprocessed fuel insertion into a pressurised water reactor (PWR) fuel element was analysed by considering different transuranic (TRU) enrichments that varied from 5.5 to 7.0 %.

Methodology

The reprocessed fuels were obtained by using the ORIGEN 2.1 code from a burnt PWR standard fuel (33,000 MWd/tHM burned), with 3.1 % of initial enrichment, which has remained in the cooling pool for five years. The burnt fuel was submitted to a UREX+ reprocessing technique, which recovers 99.95 % of uranium, 71.0 % of neptunium, 99.5 % of plutonium, 98.0 % of americium, and 79.0 % of curium. The recovered elements were spiked with ^{232}Th until the desired fissile quantity was obtained. A TRU–Th fuel element with different enrichments was evaluated and compared with a typical UO_2 (3.2 %) fuel, by using SCALE.6.0, through KENO-VI and TRITON control modules.

The fuel element is shown in Fig. 1.

Results

The k_{eff} values are shown in Fig. 2. For all fuels, the initial k_{eff} value is lower for TRU–Th than for UO_2 fuel. Comparing UO_2 standard fuel with greater fissile quantity material TRU–Th, the behaviour of both materials is similar at the beginning of the burnup, but at the end, the k_{eff} of TRU–Th indicates that the possibility of an extended burnup exists, unlike that of UO_2. Other neutronic and thermal-hydraulics parameters must be analysed to verify this possibility.

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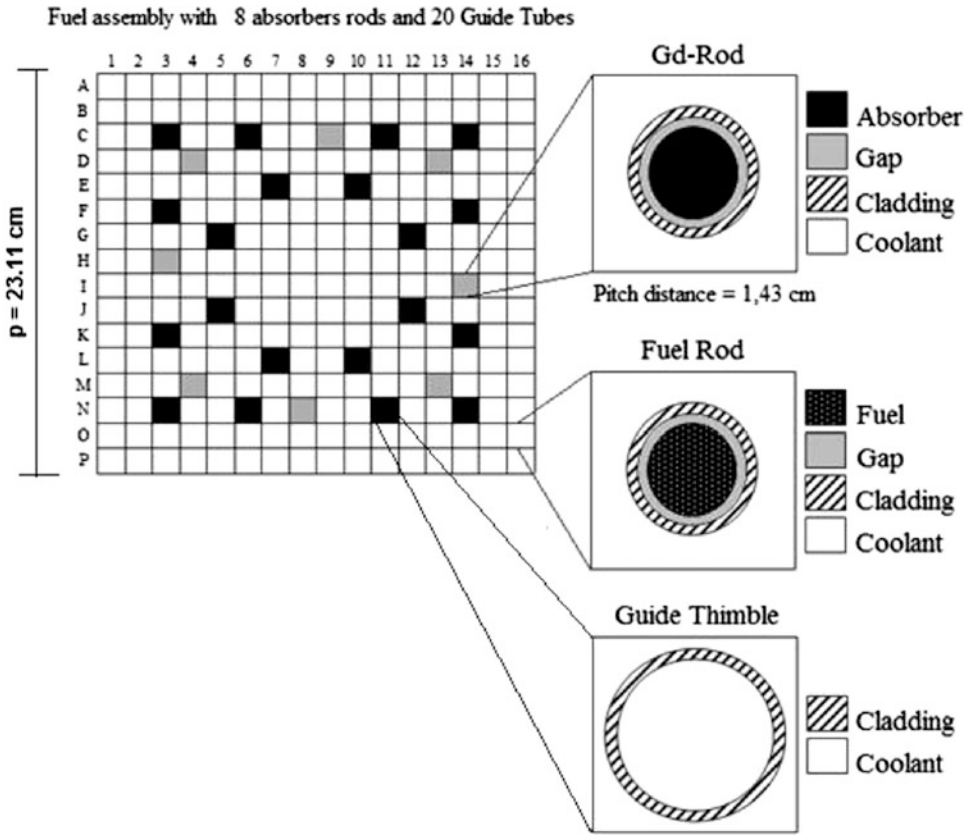

Fig. 1 The fuel element

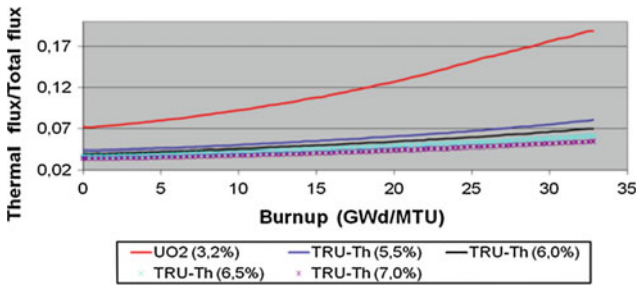

Fig. 3 Hardening spectrum during burnup

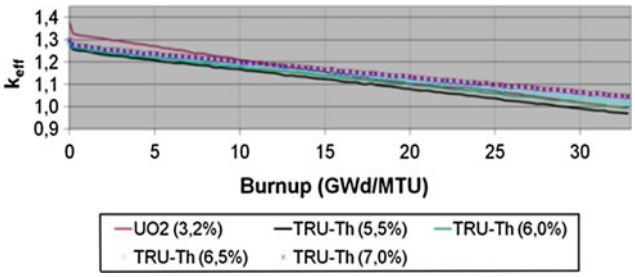

Fig. 2 k_{eff} during the burnup

Figure 3 presents the hardening spectrum for each analysed fuel, with the different enrichments for the reprocessed fuels. The spectrum of the fuel with TRU is more hardened due to the high thermal neutron absorption cross sections (Fig. 4).

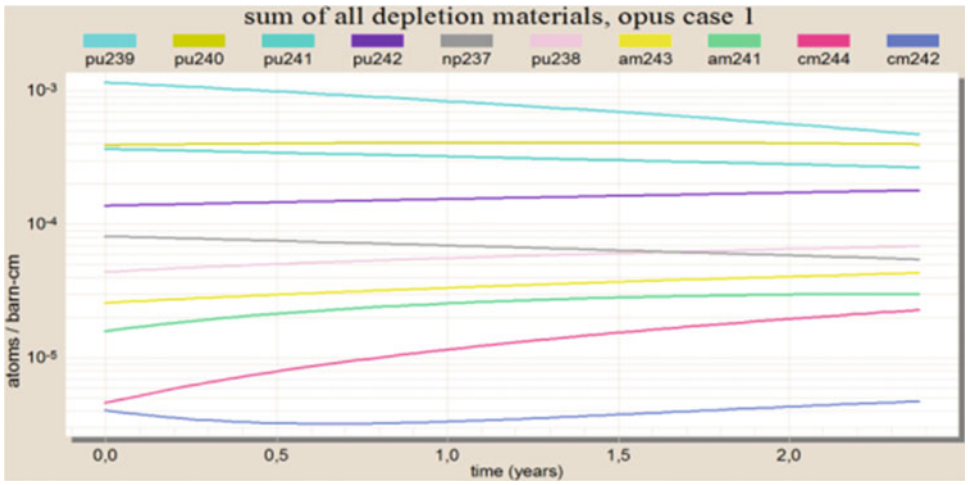

Fig. 4 TRU–Th (7.0 %) evolution during burnup

Conclusion

In a first analysis, the utilisation of advanced fuels based on thorium with transuranic elements in a fuel element of a typical PWR indicates a possibility of an extended burnup. The good results justify extending the study to evaluate core and other neutronic parameters to verify the possibility of using TRU–Th fuels in PWR.

Acknowledgements The authors are grateful to CNEN (Brazil), FAPEMIG (Brazil), CAPES (Brazil), and CNPq (Brazil) for the support.

Reference

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Evaluation of Fuel Cycles Based on U–Th and Pu–Th Mixtures in a Very High Temperature Hybrid System

L. García, A. Muñoz, L. Rodríguez, J. Rosales, C. García, F. Rosales, and C. Brayner

Abstract

Thorium-based fuel cycles are well suited to producing long-term nuclear energy with low radiotoxic waste, in addition to having intrinsic resistance to the proliferation of nuclear weapons material. In this study, fuel cycles based on U–Th and Pu–Th mixtures were evaluated in a hybrid system concept consisting of a pebble-bed very high temperature reactor (VHTR) and two accelerator-driven systems (ADS) of the same type, with the aim of obtaining a deep burn of the fuel. The cycles were divided into two stages: the first taking place in the VHTR and the second in the two ADSs using the fuel discharged from the VHTR. A continuous refueling strategy was considered, in order to extend the burn time and reach a deeper burn of the fuel. Several parameters describing the fuel behavior and the minor actinides (MA) inventory were compared for the analyzed cycles.

Parameters

The study focused on the comparison of two systems: the first a VHTR and the second an ADS. Fuel elements were: 6 cm diameter graphite pebbles containing TRISO particles. The coolant was gas (helium). The thermal power: in the VHTR variant was 200 MW; and in the ADS: 60 MW each [1, 2]. The studied fuel cycles based on thorium as fertile material were:

- Cycle 1: U + Th-232 with 8 % U-235 enrichment.
- Cycle 2: Pu-239 + Th-232. Pu-239 represents 8 % of the fresh fuel.
- Cycle 3: U-233 + Th-232. U-233 represents 6 % of the fresh fuel.

Initially, fuel (2211 kg) is loaded in the VHTR and burnt for 830 days. Then, the fuel is extracted from the VHRT and half of the fuel is burnt in each ADS for 1240, 400, or 210 days for the cycles 1, 2, and 3, respectively. The principal tool for particle transport and fuel burnup calculations was the MCNPX code, version 2.6e, library ENDF/B VI.2.

Results

See Table 1.

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Table 1 Parameters of the spent fuel discharged from the system (VHTR + ADS)

Cycle	Breeding coefficient	Consumption of the initial fissile isotope (%)	Final mass of Pu isotopes (kg)	Final mass of MA (kg)
1	0.98	U-235: 95	26.2	6.1
2	0.47	Pu-239: 99	17.3	4.7
3	0.71	U-233: 69	0.015	0.03

Conclusions

- Cycle 1 presented the highest breeding coefficient, which enables longer fuel cycles to be performed.
- Cycle 2 drastically reduced the initial mass of Pu-239.
- Cycle 3 presented the lowest build-up of Pu isotopes and MA.

Acknowledgments L. Rodríguez thanks Daniel Milián Pérez, from InSTEC, for his help in carrying out this work.

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Thermal Modeling of Thorium Sphere-Pac Fuel in an Annular Pin Design

C. Cozzo and M.A. Pouchon

Abstract

Particulate fuel is a promising concept to produce minor actinide (MA)-doped fuels for transmutation. This study assesses the performance of the fuel by simulating heat generation and temperature distribution within an arrangement of spherical ThO_2 particles. An analytical model also addresses the temperature-dependent change in the necking area between the spheres. To increase the heat transfer, an internally cooled pin concept is suggested. Its simulation promises good performance.

Introduction

Thorium-based fuels are promising substitutes for the classical uranium matrix. Although thorium can be used as a fertile material in thermal reactors [1], the present study focuses on ThO_2 in a fast reactor [2], which offers optimal [3] transmutation of minor actinides (MA). As aqueous techniques are favorable for the production of MA-doped fuels, the sphere-pac concept [4] is proposed. The performance of three fuel arrangements in an sodium-cooled fast reactor (SFR) are compared: (a) pellets, (b) sphere-pac fuel, and (c) sphere-pac fuel with a central cooling tube [5], which increases the heat transfer. Also, the annular pin is easily filled with spherical particles. The characteristics of the fuel in-pile were calculated with the SPACON code [6].

Model, Results, and Conclusions

The conditions used in the simulations are those of the European sodium-cooled fast reactor [5]. The coolant temperature is set at 450 °C, and the cladding material is steel AISI 316. The code was updated with the physical properties of ThO_2 such as the thermal conductivity [7] and

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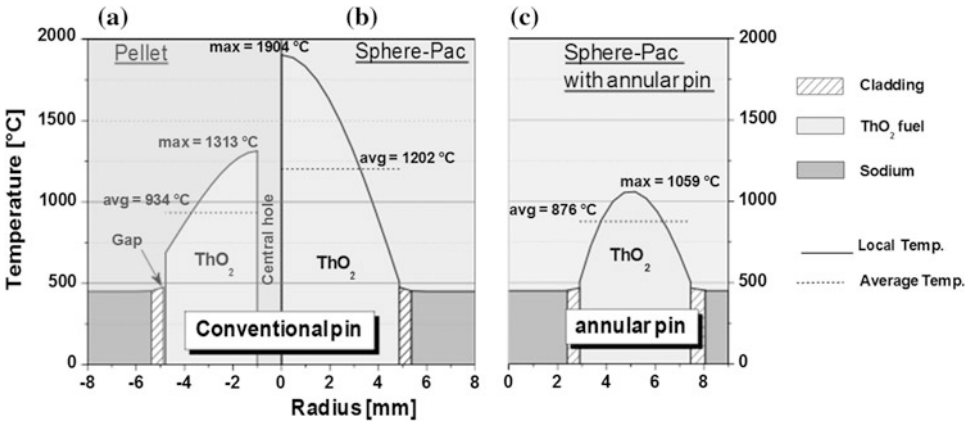

Fig. 1 Calculated temperature profiles for the three cases

expansion [8] and the Poisson coefficient [9]. The hardness experimental value [10] of ThO_2 at room temperature with a temperature dependence is similar to that measured for UO_2 [11]. The linear power is 300 W/cm for the conventional pin; for the annular one, it is calculated so that the power/section ratio is set constant: 300 × (147.9/793.6) = 603.3 W/cm.

The maximum temperatures for conventional pins are calculated (Fig. 1) to be 1904 °C (average 1202 °C) and 1313 °C (average 934 °C) for the sphere-pac and pellet, respectively. However, for the annular pin design, the maximal temperature of the sphere-pac only reaches 1059 °C (average 876 °C). Therefore, regarding the in-pile fuel temperature, transmutation of the minor actinides in an SFR with thorium oxide based fuel is optimal using spherical particles in an annular pin design.

Acknowledgments The Swiss CCEM (PINE and MeAWaT) and the European commission (contracts no. 295664-FP7-PELGRIMM & no. 295825-FP7-ASGARD) are thanked for partial financial contribution.

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Theoretical Modelling of ThO_2 Grain Boundaries Using a Novel Interatomic Potential

Ashley Shields and Nora H. de Leeuw

Abstract

Nuclear power generation is an important way to satisfy rising global energy needs without increasing dependence on coal and petroleum. However, conventional nuclear fuels, such as uranium and plutonium dioxides, raise several safety concerns. Thorium dioxide is a potentially safer alternative for nuclear reactors and, as such, is the subject of renewed research interest. Owing to the hazards of conducting experimental work on radioactive substances, a robust theoretical understanding of ThO_2 and uranium-doped ThO_2 fuel is needed. We have developed a new Th–O interatomic potential, which we have used to model ThO_2, U-doped thoria surfaces and grain boundaries, and which we will use to investigate the effect of defects on these materials.

Introduction

ThO_2 requires the contribution of a neutron source, such as uranium or plutonium, to generate a sustainable fission reaction. Owing to the hazards of conducting experimental work on radioactive substances, a robust theoretical understanding of this doped ThO_2 fuel is needed. We have developed a new Th–O interatomic potential, which we have used to model ThO_2, U-doped thoria surfaces and grain boundaries, and which we will employ further to investigate the effect of defects on these structures.

Methodology: Fitting a New Interatomic Potential

A new Buckingham potential was derived for ThO_2 to use in future modelling work, including modelling grain boundaries. The Th–O potential was fitted to available experimental data such as the bulk modulus, elastic constants, and lattice parameters in GULP 3.4 [1] (Fig. 1). The transferable Catlow O···O interaction was used [2], unchanged.

Methodology: Modelling Grain Boundaries

Starting from a relaxed bulk crystal of ThO_2, the (111) flat surface was created using Metadise 3.06. From the flat surface, a stepped surface was cut, then reflected across the cell, and the energy was minimized. Finally, the cell was grown to show the grain boundary. A number of grain boundary structures, consisting of different terrace areas and step heights, were created to reflect the microstructure of the ThO_2 material (Fig. 2).

The boundary energy relates the energy of the grain boundary with the energy of an equivalent number of atoms in the bulk material, which is, therefore, a measure of the

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ThO_2	Experimental Value[3,4]	Calculated Value	% Error in Calculated Value
Lattice Parameter (Å)	5.601	5.6001	0.00178
Bulk Modulus (GPa)	198	206.4	4.24
Shear Modulus (GPa)	98	94.4	-4.08
Elastic Constant C_{11} (GPa)	367	389.5	6.14
Elastic Constant C_{12} (GPa)	106	114.9	8.36
Elastic Constant C_{44} (GPa)	79.7	65.8	-16.72

Fig. 1 Agreement of fit [3, 4]

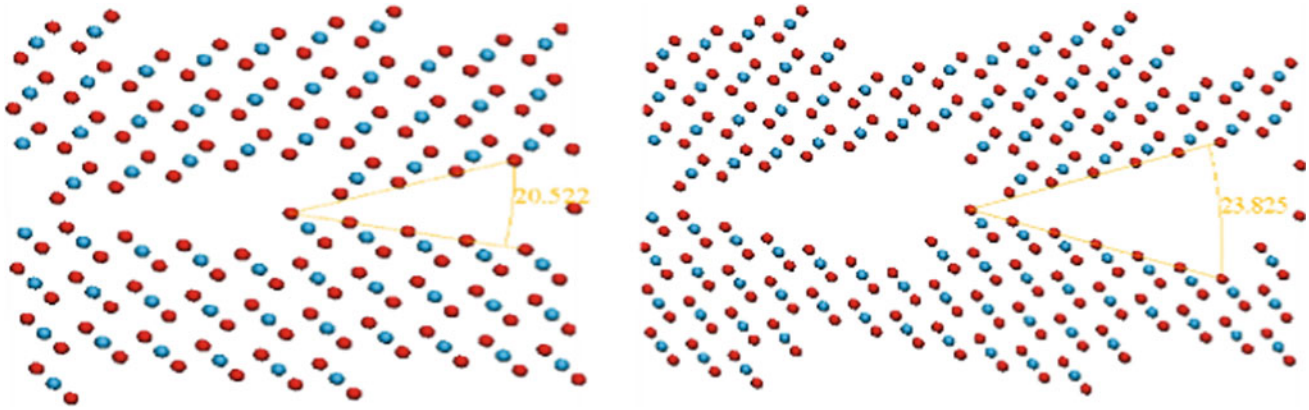

Fig. 2 Two of the six ThO_2 grain boundaries modelled in this work

Terrace Length (Å)	Angle (°)	Area (Å²)	Boundary Energy (J/m²)
22	21	258.73	2.66
22	30	307.42	3.42
30	17	365.90	2.03
30	24	411.49	2.69
40	13	473.77	1.40
40	19	517.47	2.24

Fig. 3 Boundary energies

stability of the grain boundary relative to the bulk crystal (Fig. 3).

Current and Future Research

We are currently carrying out classical molecular dynamics calculations of the pure thoria grain boundaries created above. We have also created models of 18 U-doped ThO_2 grain boundaries, which will better simulate real world use as a nuclear fuel, and we will investigate the migration of water, fission product gases, and other defects across both the pure and U-doped grain boundaries.

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A Novel Approach for Preferential Recovery of ^{90}Sr from Irradiated ThO_2

Chirag K. Vyas, Pranav M. Joshirao, Rakesh Shukla, Jong Seo Chai, and Vijay K. Manchanda

Abstract

Thorium is likely to play a pivotal role in the sustained use of nuclear energy as uranium reserves dwindle in the next few decades. This outlook is a result of thorium's abundant supply (3–4 times that of uranium), favorable neutronic and physico-chemical properties (as a fuel material), intrinsic proliferation resistance (owing to the presence of ^{232}U along with ^{235}U), the lower content of minor actinides in the spent fuel, and better long-term behavior of its waste in geological repositories. Its high η factor makes ^{233}U the best fissile isotope, of all existing fissile isotopes, for thermal neutrons, but it produces significant amounts of ^{90}Sr. If this ^{90}Sr can be recovered, it can be used as a valuable radionuclide for ^{90}Y—a potential radionuclide for therapeutic applications, such as treating lung cancer, melanoma, and renal cell carcinoma.

In lieu of a written contribution by the authors, the actual poster presented at ThEC13 is included.

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Thorium Energy Conference THEC13

October 27-31, 2013, Globe of Science and Innovation, CERN, Geneva, Switzerland

A novel approach for preferential recovery of ^{90}Sr from irradiated ThO_2

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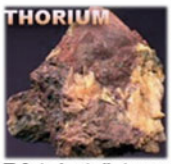
Introduction



Due to its abundant supply (3-4 times of uranium) favorable neutronic and physico-chemical properties (as a fuel material), intrinsically proliferation resistance (due to the presence of ^{232}U along with ^{233}U), lower content of minor actinides in spent fuel and better long term behaviour in geological repository, thorium is likely to play a pivotal role for the sustenance of nuclear energy once the uranium reserves dwindle after few decades. High eta factor makes ^{233}U as the best fissile isotope of all existing fissile isotopes for thermal neutrons. ^{90}Sr is a valuable radionuclide for ^{90}Y (potential radionuclide for therapeutic applications, such as lung cancer, melanoma, and renal cell carcinoma).

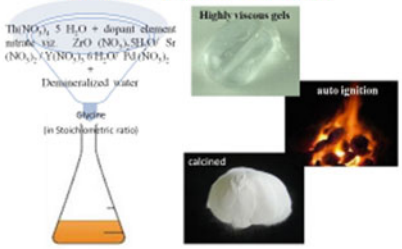
ThO_2 is chemically inert and requires strong HNO_3 and complexing agent like F^- for dissolution. **An attempt is made in the present work to carry out preferential leaching of strontium from mixed (Th/Sr) oxide using dilute HNO_3 / $HClO_4$.** ThO_2 (sintered at about 1250°C) doped with 0.25% Sr was employed for the dissolution studies in the present work. Purification of Sr was carried out using Sr-selective extraction chromatographic material. Recovery of ^{90}Sr from spent nuclear fuel at dissolution stage may facilitate safer fuel reprocessing and eliminate partially the need for interim storage of conditioned high level waste

% Cumulative Yield of ^{90}Sr for various fissile/ fissionable nuclides with thermal and fast neutrons

Fissile/ Fissionable nuclide	Thermal	Fast
^{232}Th	-	7.32
^{233}U	6.65	6.39
^{238}U	-	3.11
^{235}U	5.37	5.22
^{239}Pu	2.01	2.03
^{241}Pu	1.51	1.50

Experimental

Preparation of Sintered and Doped ThO_2



Dissolution Studies

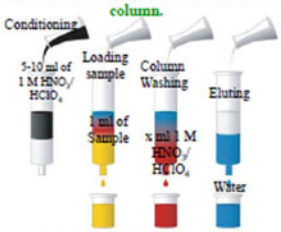
Th doped with 0.25 % Sr/ Zr/ Pd/ Y + 10/25 ml of x M of HNO_3 / HCl/ $HClO_4$ acid

Refluxation for specific interval of time (5 min. to 4 hours)

The solution was double centrifuged and 500 µl of supernatant was diluted to 10 ml using H_2O.

The diluted solution was analyzed by ICP-OES

Separation of Sr from Th and other Fission Products4,4'(5')-di-t-butylcyclohexano 18-crown-6, 2 ml column.



Counted using Perkin Elmer (1480 WIZARD) Gamma Counter.

K_d Determination

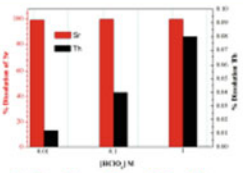
Dry the activity [^{137}Cs (I), ^{85}Sr (II), ^{152}Eu (III)]; dissolve the dried activity in the specified volume of desired molarity of $HClO_4$/ HNO_3.

Count 1 mL of above solution using NaI(Tl) counter. Add 200mg of Sr selective resin to 2mL of the activity with the specified acid and equilibrate it for 1 hour in shaking incubator at constant temperature (25 ± 0.1°C)

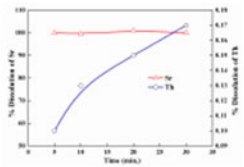
Separate the resin and liquid using 0.45µm PTFE syringe filter. Count 1 mL of separated liquid using NaI (Tl) detector.

Experiments with Th and Zr were carried out in similar way except the analysis was carried out by ICP-OES.

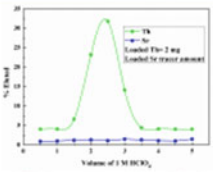
Results and Conclusion



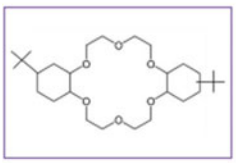
% Dissolution of Sr and Th of 50 mg calcined $(Th/Sr)O_2$ in 10 mL of $HClO_4$ medium after 10 min. of refluxation



% Dissolution of Sr and Th in 50 mg calcined $(Th/Sr)O_2$ in 1 M HNO_3 medium as a function of time



Elution pattern of Th and Sr in 1 M $HClO_4$ medium



Structure of 4,4'(5') Ditertiary butyl cyclohexano 18 crown 6

%Dissolution of Th and different fission products

Acid	Sr	Zr	Pd	Y	Th
1 M HNO_3	~100	28	36	8	0.13
1 M $HClO_4$	~100	3.2	18	1.6	0.08

Distribution coefficient data (cm^3/gm) for Sr(II), Th(IV) and Zr(IV)

Metal ion	1 M HNO_3	S.F. w.r.t. Sr(II)	1 M $HClO_4$	S.F. w.r.t. Sr(II)
Sr(II)	8.75 ± 0.45	-	22.6 ± 1.36	-
Th(IV)	0.18 ± 0.01	48.6	0.44 ± 0.03	51.3
Zr(IV)	1.60 ± 0.08	5.46	2.27 ± 0.18	9.95

This work provides a novel approach to mitigate the radiotoxicity of the discharged Th based fuel caused by ^{90}Sr and suggests a new chemical separation procedure to purify ^{90}Sr from Th and other fission products using dilute perchloric acid . Purified ^{90}Sr has many applications including parent radionuclide for ^{90}Y, a promising nuclide for radiopharmaceuticals and compact power heat source .

CERN EPFL ECOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE iThEC International Thorium Energy Committee ThEC THORIUM ENERGY CONFERENCE 2013 GENEVA ITHEO PAUL SCHERRER INSTITUT PSI

Part XII
Posters: Thorium Reactor Physics

Preliminary Applications of ANET Code for the Investigation of the Hybrid Soliton Reactor Concept

N. Catsaros, B. Gaveau, M.-T. Jaekel, J. Maillard, G. Maurel, P. Savva, J. Silva, M. Varvayanni, and T. Xenofontos

Abstract

The Monte Carlo code ANET is under development based on the high energy physics code GEANT3.21. It can simulate ADS systems, accounting for the high-energy proton beam interaction with spallation target, neutron trajectories, and the reaction rate distribution in the core. At the same time, it is structured to account for core materials modification due to nuclear reactions and to radioactive decay. Preliminary ANET applications for the investigation of the hybrid reactor concept are presented here.

The ANET Code and the Hybrid Soliton Reactor (HSR) Concept

ANET has three main loops: on events, particle tracking, and microscopic time step. The reactor parameters (geometry, inventory, temperature, neutron source) can change during computation. The HSR concept is described by a long cylindrical vessel, Φ, which contains the fertile and fissile materials as well as transuranic waste to be destroyed (Fig. 1).

Results and Concluding Remarks

Preliminary ANET results for a conceptual HSR loaded with $^{238}U/^{239}Pu$ show that after an initial decrease, the fissile material (^{239}Pu) increases regularly showing breeding capability (Fig. 2). Clear breeding is shown for beam power ~ 3 MW. A regular k_{eff} decrease, which persists once

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breeding is established, is seen. It can be concluded that the preliminary analysis indicates that a thermal HSR could be conceived to reach breeding performances when operating at low and medium power.

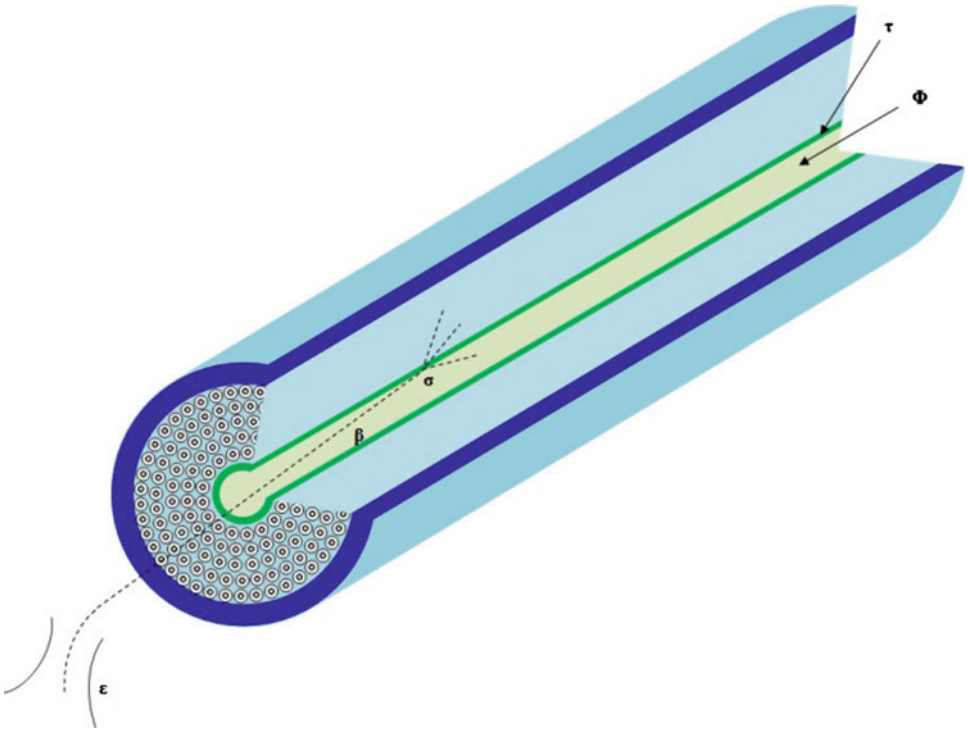

Fig. 1 Fuel pins in a hexagonal geometry; empty central tube with a moving neutron source (σ); proton beam (β) from an accelerator; hit target (τ) produces neutrons

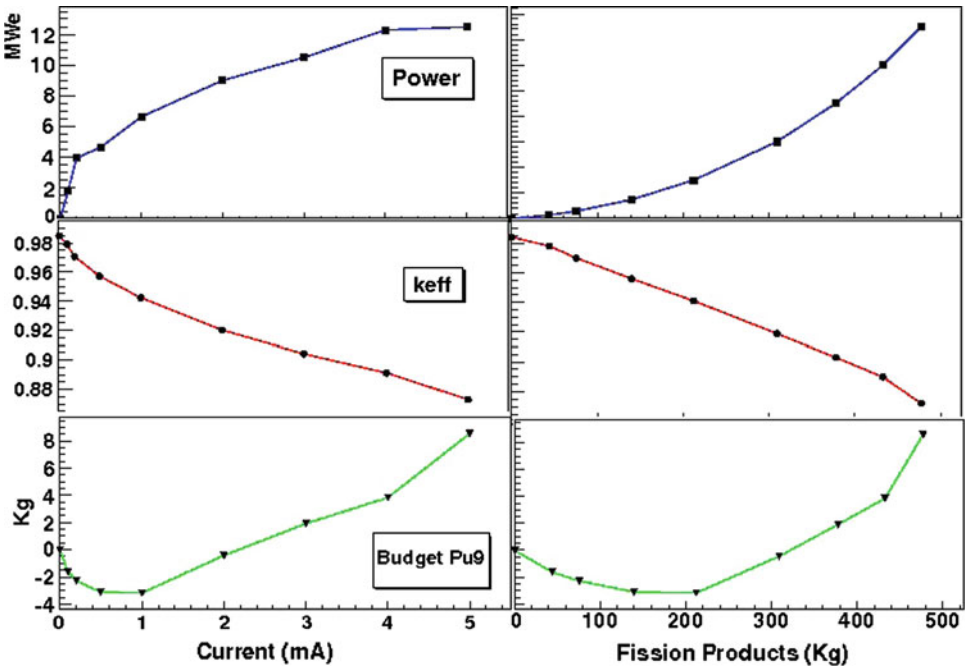

Fig. 2 Preliminary results from a conceptual HSR configuration

The Proto-Earth Georeactor: A Thorium Reactor?

Claude Degueldre and Carlo Fiorina

Georeactors have been suspected to occur in natural uranium deposits [1]. They have, for example, been found in the Earth's crust in Oklo, Gabon [2]. The feasibility of one or more massive nuclear fission reactors deeper inside the Earth was also proposed. The existence nowadays, or in the past, of such georeactors has been suggested by experimental evidence, such as: (1) a higher $^{3}He/^{4}He$ ratio in various basalts samples versus air [3]; (2) different Xe isotope ratios from various geosystem samples compared with air [4]; and (3) translation of the North Magnetic Pole toward Siberia [5].

The ERANOS 2.2n core-physics code [6] has been used in this study to evaluate the neutronic features of such potential georeactors and their evolution over time. ERANOS2.2n is a state-of-the art core physics code developed at CEA (France) for the analysis of fast neutron spectrum nuclear reactors.

The calculations performed show that the uranium composition 4.5 Ga ago ($\sim$25 % ^{235}U) and 3 Ga ago ($\sim$7 % ^{235}U) provide a neutron multiplication factor of $k_{\infty} \approx 1.4$ and $k_{\infty} \approx 1$, respectively. This implies that the geosystem may have reached criticality in this range of time. Possible activity of a georeactor today would be possible in the case of its specific power being relatively high ($\sim$1 mW kg^{-1}). This would allow the transmutation of ^{238}U into ^{239}Pu, and eventually into ^{235}U, to be quick enough to overcome ^{235}U decay and consumption. Figure 1 shows the ^{235}U enrichment in natural U in the case of zero power (inactive georeactor) or in case that a georeactor had been operating with a 1 mW kg^{-1} specific power.

The role of thorium would have been essentially that of a neutron absorber. In fact, ^{232}Th generates the fissile material ^{233}U when irradiated, but the half-life of the latter (1.6×10^5 years) is extremely short in geological terms, thus making its buildup negligible. As an example, Fig. 2 shows the evolution over time of the multiplication factor and of various isotopes for a $\sim$5 mW kg^{-1} georeactor that started as a pure uranium mixture, but has been affected by gradual

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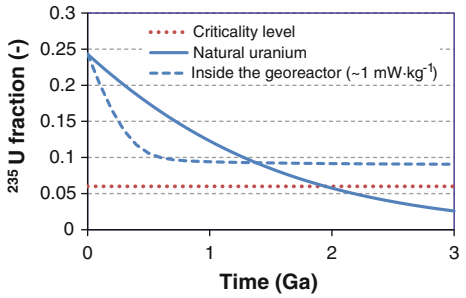

Fig. 1 Evolution over time of the ^{235}U fraction in natural uranium and for a $\sim$1 mW kg^{-1} georeactor only composed of uranium

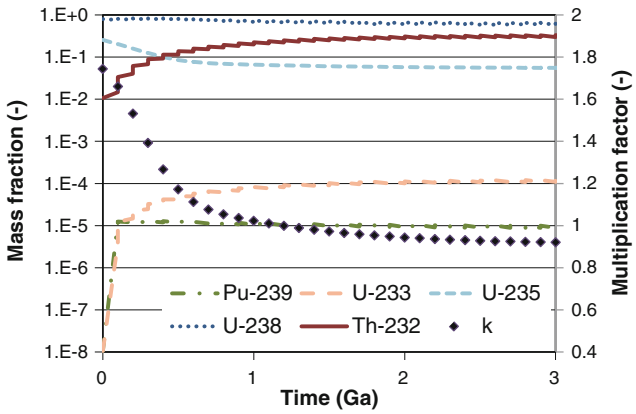

Fig. 2 Evolution over time of multiplication factor and of various isotopes for a $\sim$5 mW kg^{-1} georeactor that started as a pure uranium mixture, but has been affected by gradual thorium contamination

thorium contamination. In spite of this high U enrichment, the increasing thorium concentration would switch off the georeactor after approximately 1 Ga.

Even if the hypothesis of a georeactor operation in both the proto-Earth period and nowadays could be acceptable, this would be difficult to prove. A future possibility would be assessing and reconstructing the system by antineutrino detection and tomography through the Earth.

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Part XIII
Posters: Accelerator-Driven Systems

GEM*STAR Multipurpose Applications

R.P. Johnson, G. Flanagan, F. Marhauser, C.D. Bowman, and R.B. Vogelaar

Abstract

In the GEM*STAR accelerator-driven subcritical reactor [1] that we plan to build, the accelerator allows subcritical operation (no Chernobyls), the molten salt fuel allows volatiles to be continuously removed (no Fukushimas), and the fuel does not need to be enriched or reprocessed (to minimize weapons proliferation concerns). The molten salt fuel and the relaxed availability requirements of process heat applications mean that the required accelerator technology is available now. The thermal spectrum, graphite-moderated GEM*STAR reactor can be used without design modification with different fuels for process heat or electricity generation. These fuels include thorium, spent nuclear fuel, weapons-grade Pu-239 or U-233, and natural un-enriched or depleted uranium. We report progress on the technical design report, including the internal spallation target, as well as progress in achieving support to burn 34 tonnes of weapons-grade plutonium, in accordance with the year 2000s Plutonium Management and Disposition Agreement between the US and Russia.

In lieu of a written contribution by the authors, the actual poster presented at ThEC13 is included.

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GEM*STAR Multipurpose Applications

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Abstract

In the GEM*STAR [1] accelerator-driven subcritical reactor that we plan to build, the accelerator allows subcritical operation (no Chernobyls), the molten salt fuel allows volatiles to be continuously removed (no Fukushimas), and the fuel does not need to be enriched or reprocessed (to minimize weapons proliferation concerns). The molten salt fuel and the relaxed availability requirements of process heat applications imply that the required accelerator technology is available now. The thermal-spectrum, graphite moderated GEM*STAR reactor can be used without design modification with different fuels for process heat or electricity generation. These fuels include thorium, spent nuclear fuel, weapons-grade Pu239 or U233, and natural un-enriched or depleted uranium. The first application most likely to be funded is to burn 34 tonnes of weapons-grade plutonium according to the year2000 Plutonium Management and Disposition Agreement between the US and Russia.

[1] Charles D. Bowman R. Bruce Vogelaar, Edward G. Bilpuch, Calvin R. Howell, Anton P. Tonchev, Werner Tornow, R.L. Walter, "GEM*STAR: The Alternative Reactor Technology Comprising Graphite, Molten Salt, and Accelerators," Handbook of Nuclear Engineering, Springer Science+Business Media LLC (2010).

Introduction

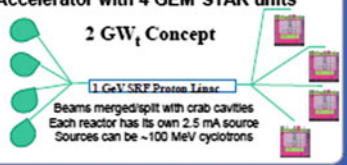
Muons, Inc. and ADNA Corp. are developing GEM*STAR (Green Energy Multiplier * Subcritical Technology Alternative Reactor) to serve multiple roles. The same design can be used to convert fertile Th232 or U238 into fissile U233 or Pu239 respectively, and to control the fission process itself. By concentrating on process heat to convert natural gas to diesel fuel, the demands on the accelerator are reduced and the potential profits relative to electricity production are increased. An attractive first application is the burning of excess weapons-grade plutonium slated to be destroyed by the 2000 U.S.-Russian Plutonium Management and Disposition Agreement.

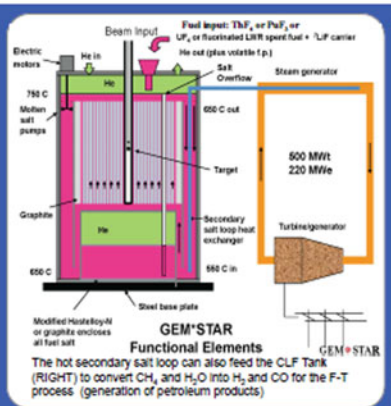
GEM*STAR involves interfacing known technologies, where:
1) A Superconducting RF (SRF) LINAC produces a powerful proton beam
2) that strikes a spallation target to generate neutrons to drive
3) a GEM*STAR Accelerator-Driven Subcritical Reactor (ADSR) fueled with molten salt eutectic mixtures of U, Th, Pu, and Li fluorides
4) to heat a secondary salt loop to convert natural gas and carbon into feedstock for the Fischer-Tropsch generation of petroleum products.

GEM*STAR Advantages for the Nuclear Industry

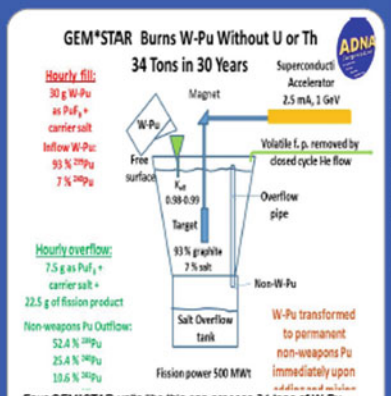
- Subcritical operation - The LINAC provides extra neutrons for fission
- Volatile radioactive elements are continuously purged to reduce potential accident release sources by a factor of a million
- Molten salt fuel (MSF) can include natural thorium, natural uranium, depleted uranium, spent fuel from Light Water Reactors, or Excess weapons-grade plutonium (W-Pu).
- Nuclear proliferation issues reduced – The MSF does not need to be
  - enriched or
  - reprocessed to remove fission products or higher actinides
- MSF technology was demonstrated at the ORNL Molten Salt Reactor Experiment (MSRE)
- MSF technology operates at high Temperature, needed for process heat and electrical power generation efficiency

Accelerator with 4 GEM*STAR units





The hot secondary salt loop can also feed the CLF Tank (RIGHT) to convert CH_4 and H_2O into H_2 and CO for the F-T process (generation of petroleum products)



Four GEM*STAR units like this can process 34 tons of W-Pu, producing 80 billion gallons of diesel at <$2/g al to provide the DOD with most of what it needs for 30 y.

Benefits of GEM*STAR for Weapons-grade Pu Disposition

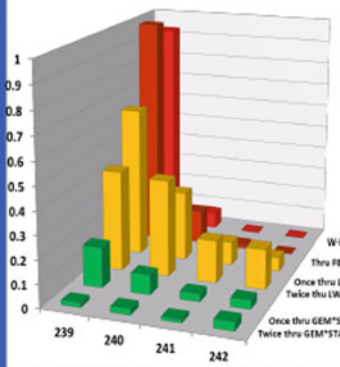
- **Burned W-Pu never useful for weapons**
- **Burned W-Pu never decays back to weapons useful material**
- **Conversion to non-W-Pu in minutes**
- **Pu isotopic mixture can be reduced from 34 tons to 0.2 tons if desired**
- **Also converts Commercial Pu (C-Pu) to-non-weapons-useful material**
- **Never requires a critical mass - no control rods**
- **No reprocessing or enrichment required**
- **No conversion to MOX; simple conversion of Pu metal and PuO_2 to PuF_3**
- **Fission energy converted to diesel and sold as green fuel to DOD**
- **No stored large volatile fission product inventory as in LWRs (Fukushima)**
- **Liquid fuel moved by He pressure; no radiation exposure to humans**
- **Operates at atmospheric pressure - No pressure vessel**
- **Passive recovery from a loss of coolant accident (LOCA)**

First Application: Utilization and Destruction of Weapons-grade Pu

The figure to the left and the one below it show conceptual pictures of a GEM*STAR unit and its application to use and destroy W-Pu. A 2.5 MWb accelerator generates 500 MWt, reducing 30 g of W-Pu to 7.5 g per hour and adding the neutron generating isotopes Pu240 and Pu242. The figure below shows the isotopes of the original W-Pu (back row-red) compared to the outputs of GEM*STAR (green), the Russian BN800 Fast Breeder (dark red), and the US MOX plan using Light Water Reactors (yellow). By passing the fuel through the reactor a second time, additional benefits can be seen. However, in the case of MOX, the fuel has to be chemically reprocessed to remove fission products. GEM*STAR does not require reprocessing for a second pass.

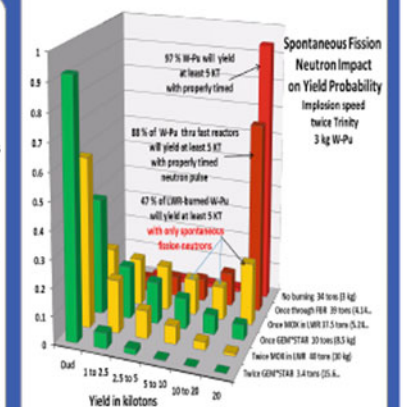
GEM*STAR does better because of the effectiveness of the thermal neutron spectrum both to produce Pu240 and Pu242, and to be less sensitive to fission products than MOX-LWR. Pu242 is even more effective than Pu240 to cause premature explosions in that it is a neutron emitter with a lifetime longer than Pu239. The bottom figure below shows that even the small remnant after two passes in GEM*STAR is useless for weapons.

Isotope remnants after treatment



Isotopes 240 and 242 are neutron emitters that turn weapons-grade plutonium into commercial grade. W-Pu has less than 8% Pu240.

Utility of remnants for weapons



The effectiveness of utilizing and destroying W-Pu is a compelling case for GEM*STAR to be the first accelerator-driven subcritical reactor. Th and SNF are next.

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A Provisional Study of ADS Within the Turkic Accelerator Complex Project

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Abstract

The Turkic Accelerator Complex (TAC) project has been developed with the support of the Turkish State Planning Organization through collaboration of ten Turkish universities. The complex is planned to have four main facilities: Super-Charm Factory, free-electron laser (FEL), synchrotron radiation (SR) source, and GeV energy, mA current proton linear accelerator (linac). As Turkey has essential thorium reserves, the ADS has become a very attractive emerging energy technology for our country.

Introduction

One of the most important applications of accelerator technology is the emerging accelerator-driven system (ADS), in other words "green" nuclear technology, which could provide a solution for mankind's energy problems without destroying our environment [1, 2]. In this respect, the proton accelerator part of the Turkic Accelerator Complex (TAC) project [3, 4] is of particular importance for Turkey.

Thorium in Turkey

According to a joint report by the OECD Nuclear Energy Agency and the Inter-national Atomic Energy Agency [5], Turkey has 880,000 tonnes of thorium reserves compared with 7300 tonnes of uranium reserves. The first number, which reflects thorium deposits in monazite minerals in the Eskisehir-Sivrihisar region, does not include thorium reserves in other regions (Isparta, Malatya, etc.), which have not been investigated in detail. For example, Canakli 1 and Canakli 2 deposits in the Isparta-Aksu region are estimated at 19,760 tonnes of ThO_2 in thorite minerals [6]. Thorium reserves could provide for the energy needs of Turkey for thousands of years, whereas the uranium reserves aren't enough even for one year.

The TAC Proton Accelerator

Keeping in mind ADS applications, we are concentrating on a high intensity (>1 mA) proton linear accelerator (linac) in the GeV energy region. The low and medium energy parts of proposed proton accelerator will consist of normal conducting cavity structures. After the ion source, there will be RFQ (radiofrequency quadrupole), DTL (drift tube linac) and CCDTL (coupled-cavity drift tube linac) accelerator structures. There is a low energy beam transport channel (LEBT) between the ion source and the RFQ. Its task is to

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match the ion beam to the RFQ by using the solenoid and steering magnets. For the high energy part, use of normal conducting CCL (coupled-cavity linac) cavities are planned. Beam simulations over the whole line have been performed and it has been shown that the beam will be stable up to a 60 mA beam current. Beam simulations for a superconducting version of the high energy part are under development.

Project PROMETHEUS

The PROMETHEUS Project (Design and Construction of a Radio Frequency Quadrupole at TAEK-SANAEM) is ongoing for the design and development of a four-vane radiofrequency quadrupole (RFQ), together with its H^+ ion source, a low energy beam transport (LEBT) line, and diagnostics section [7]. The main goal of the project is to achieve the acceleration of low energy ions up to 1.5 MeV by an RFQ (352 MHz) shorter than 2 m. A plasma ion source is being developed to produce a 20 keV, 1 mA H^+ beam. Simulation results shows that a beam transmission of 99 % could be achieved at 1.7 m downstream, reaching an energy of 1.5 MeV. The first accelerated protons are expected to be available by the end of 2015.

Conclusion

In the framework of the TAC project, we are working on the design of a proton linac for medium scale (250 MW) power generation, which corresponds to 1 GeV energy and 2.5 mA current proton beams. In our opinion, the lower current will simplify reliability and availability aspects. Keeping in mind the essential thorium reserves, Turkey should prepare a corresponding state program and join with emerging international collaborations as soon as possible.

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ADS Research Activities at Sungkyunkwan University

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Introduction

Thorium-based accelerator-driven system (ADS) technology for future nuclear energy is being developed at Sungkyunkwan University. Our research activities include research on computational modeling, partitioning and transmutation, detector development, and life cycle assessment analysis. A brief overview is given here.

Research Topics

Nuclear Data Library (TNudy)

The ENDF format for nuclear data was designed many years ago [1]. We propose a new prototype of a nuclear data manipulation package (TNudy) based on the ROOT system [2]. The object-oriented C++ framework of the ROOT system has been the de facto standard in high energy and nuclear physics experiments worldwide for the last ten years. Starting from the ENDF format, the data will be stored in a machine portable binary format. Root files are internally structured with direct access to their different sections and are automatically compressed upon writing and uncompressed upon reading, thereby minimizing the disk occupancy. The ROOT framework includes the so-called Virtual Monte Carlo system, which allows us to run different major transport Monte Carlo (FLUKA, Geant4, Geant3) with a common geometry modeler, which is also part of the ROOT system. Various routines have been developed for ENDF sub-libraries, data visualization, and user-friendly application. Extending these routines and verification of the libraries are under progress [3].

Transmutation by Adiabatic Resonance Crossing Method

A novel method of transmutation by adiabatic resonance crossing (TARC) was pioneered by C. Rubbia et al. at CERN in the 1990s [4]. Transmutation of isotopes can be effective not only for transmutation of long-lived fission products, but also for the production of medical radioisotopes. Based on this concept, we have done a design study and heat transfer analysis to determine the main physical parameters of the TARC system by considering the neutron production, energy deposition, and other related data. We assume the use of 2 kW proton beams of 50 MeV with a 40 μA current, which can be delivered by the MC-50 Cyclotron at KIRAMS (Korea Institute of Radiological And Medical Sciences). A TARC setup has been designed, including a neutron converter target and a heat removal system, by using the Monte Carlo particle transport code FLUKA [5]. A thermal analysis was performed for cooling the Be target by using the ANSYS-CFX package [6]. An elaborate design of a heat removal system is made, given the relatively high power deposited over a localized region of the target.

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Radiochemistry

Transmutation of long-lived radionuclides is one of the major objectives of the ADS program. ^{237}Np, ^{239,240,242,244}Pu, ^{241,243}Am, and ^{245}Cm are the principal alpha-emitting long-lived radionuclides. These radionuclides can be obtained in the pure state only through aqueous routes. In this context, apart from other extractants, diglycolamides (TODGA) have been investigated extensively in leading laboratories for actinide partitioning [7]. Studies are in progress on the separation of minor actinides using a TODGA-based extraction chromatography resin, a novel nanocomposite material, polystyrene–TODGA (PS–TODGA), and carbon nanotube–TODGA derivatives.

Fission products such as ^{137}Cs, ^{90}Sr, and ^{99}Mo find extensive applications in industry and in nuclear medicine. ^{232}Th used in thorium-based ADS is a potential source of these valuable radionuclides. Novel separation procedures are under development for the separation and purification of these fission products from Th(IV) and other activation/fission products [8].

Development of Detectors

In the wake of the Fukushima accident, there is a need to efficiently monitor the release of alpha-emitting radionuclides caused by core meltdown or loss of coolant in a spent fuel pond. As a consequence of the exposure of diethylene-glycol bis(allylcarbonate) (CR-39) to alpha radiation (emitted from ^{232}Th), radiation damage causes modification of its physicochemical properties, including absorbance, surface roughness, and reflectance. These property modifications can be correlated with the chemically etched track density, which is conventionally followed for the quantification of alpha-radiation off-line. A CR-39-based fiber optic reflectance sensor has been developed with the aim to monitor on-line the presence of alpha-emitting radionuclides (in or around a nuclear facility) [9, 10]. Also, new mixtures of solvents are under development to speed up the etching procedure of CR-39 [11]. Other advanced materials such as graphene and DNA structures are under investigation as radiation sensors [12, 13].

MICROMEGAS detectors have been developed for the detection and tracking of charged and neutral particles in many applications, such as nuclear physics, high-energy physics, and medical applications. The control parameters of MICROMEGAS include materials, the type of gases, the mixing ratio of gases, and the geometry. The characteristics and the performance of MICROMEGAS are determined by optimizing the parameters through varying conditions for the construction and operation. GARFIELD simulations of MICROMEGAS were conducted to optimize the characteristics and performance with respect to the nature of the gas mixture, dimensions of the gas chamber, applied voltages of drift, and amplification regions. With the results of the simulation study, a MICROMEGAS gaseous detector was built and characterized by using standard X-ray sources (^{55}Fe, ^{109}Cd) at 1 atm and room temperature. We measured the voltage gains against the applied voltage range of each region for different combinations of gases and compared them with the simulation results [14].

Life Cycle Assessment

We have studied a system model for the life cycle assessment (LCA) of pyro-processing for a closed nuclear fuel cycle. The impact covers the complete life cycle of the nuclear process, including material mining, milling, conversion, and fabrication of nuclear fuels as well as material collection, construction, operation, and disposal of nuclear waste at a power plant. Previous studies have analyzed the life cycle impact of the nuclear power process, but these studies dealt with a once-through cycle only and excluded the reprocessing phase. To focus the technology development of next generation nuclear technology to be more sustainable, the environmental burden of the closed fuel cycle should be considered. In this work, we address the system boundary of pyro-processing (as one of the reprocessing technologies) for the LCA analysis and estimate its associated life cycle impacts [15]. The study is a pilot research of applying LCA to a nuclear technology. As a next step, LCA would be applied to an ADS system to estimate its environmental performance.

Summary

Research activities have been focused on computational modeling, partitioning and transmutation, detector development, and life cycle assessment analysis. A new nuclear data library based on the ENDF and ROOT system has been developed. Production of radioisotopes and transmutation has been studied based on the concept of TARC. Thermodynamic analyses of a multi-channel helium-cooled device have been performed with the computational fluid dynamics (CFD) code CFX. Partitioning studies have been conducted using ‘green’ extractants. Preferential recovery of ^{90}Sr from irradiated ^{232}Th has been studied. We have conducted so-called life cycle assessment analyses for estimating the cost of

conventional and modified fuel cycles of nuclear power. MICROMEGAS detectors have been developed to detect photons, charged particles, and neutrons. Also, an on-line alpha detector has been developed by using solid-state nuclear track detector CR-39 and fibre optic sensors.

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Design of a Compact Transportable Neutron Source in TIARA/EU/FP7

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Abstract

Neutron sources are increasingly used in physics laboratories and also have potential applications to accelerator-driven systems (ADS). Improving yields is the key; this entails improving the density of the neutron flux and compactness of the neutron source. Under the auspices of the EU-FP7 project TIARA, "Test Infrastructure and Accelerator Research Area", efforts are currently underway at CERN to design a small compact transportable neutron source with a dense neutron field that could be installed in many facilities and laboratories around the world.

Retained Design

The overall aim of the FP7 project TIARA is to structure and optimize the R&D effort in the field of accelerator science and technology in Europe. In particular, the task entitled TIHPAC (Test Infrastructure for High Energy Power Accelerator Components) serves to encourage the application of target technology to as wide applications as possible. After a review of the existing applications, a need was identified for a flexible compact neutron source allowing testing of materials samples in a compact flexible configuration and allowing its shipment to different laboratories.

The overall design seeks to provide a flexible facility for material testing under p + n irradiation, LM corrosion, and constant/cyclical stress. The resulting proposed facility has been named T-MIF, an acronym for Test Material Irradiation Facility. In this compact design, liquid metal is used as the target material to provide spallation neutrons on the one hand, and as a coolant for evacuating the beam heat deposition on the other. The main components are represented in Fig. 1. The different types of shielding are represented as an overall transparent envelope for a better visibility of the components.

Specific Design Details

The genesis of the design of the target originates with the Eurisol program (Fig. 2 left), a project aimed at developing high-power neutron targets for the investigation of rare isotopes. The design has been adapted (Fig. 2 right) to allow the placement of material samples in the beam impact area to obtain the highest possible dpa for material testing, currently 5–10 dpa per annum.

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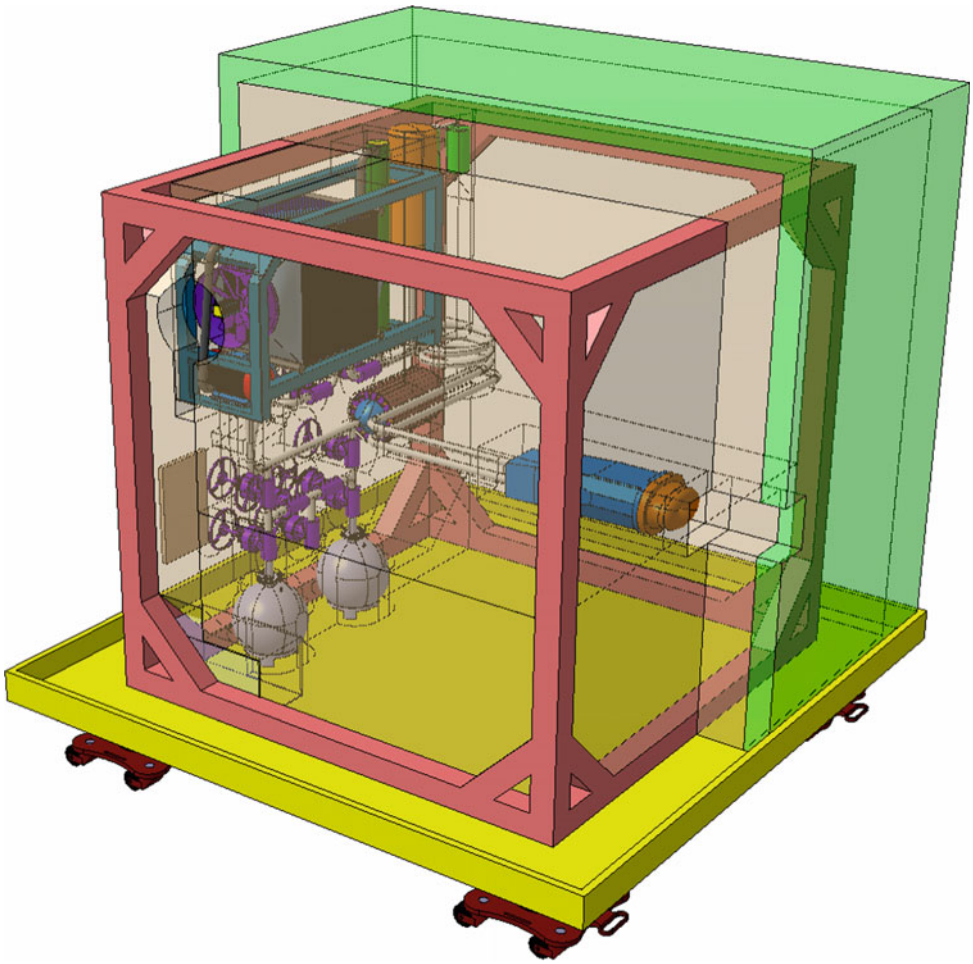

Fig. 1 Overall design of the material testing station

Fig. 2 Tested Eurisol target (*left*) and derived T-MIF target concept (*right*)

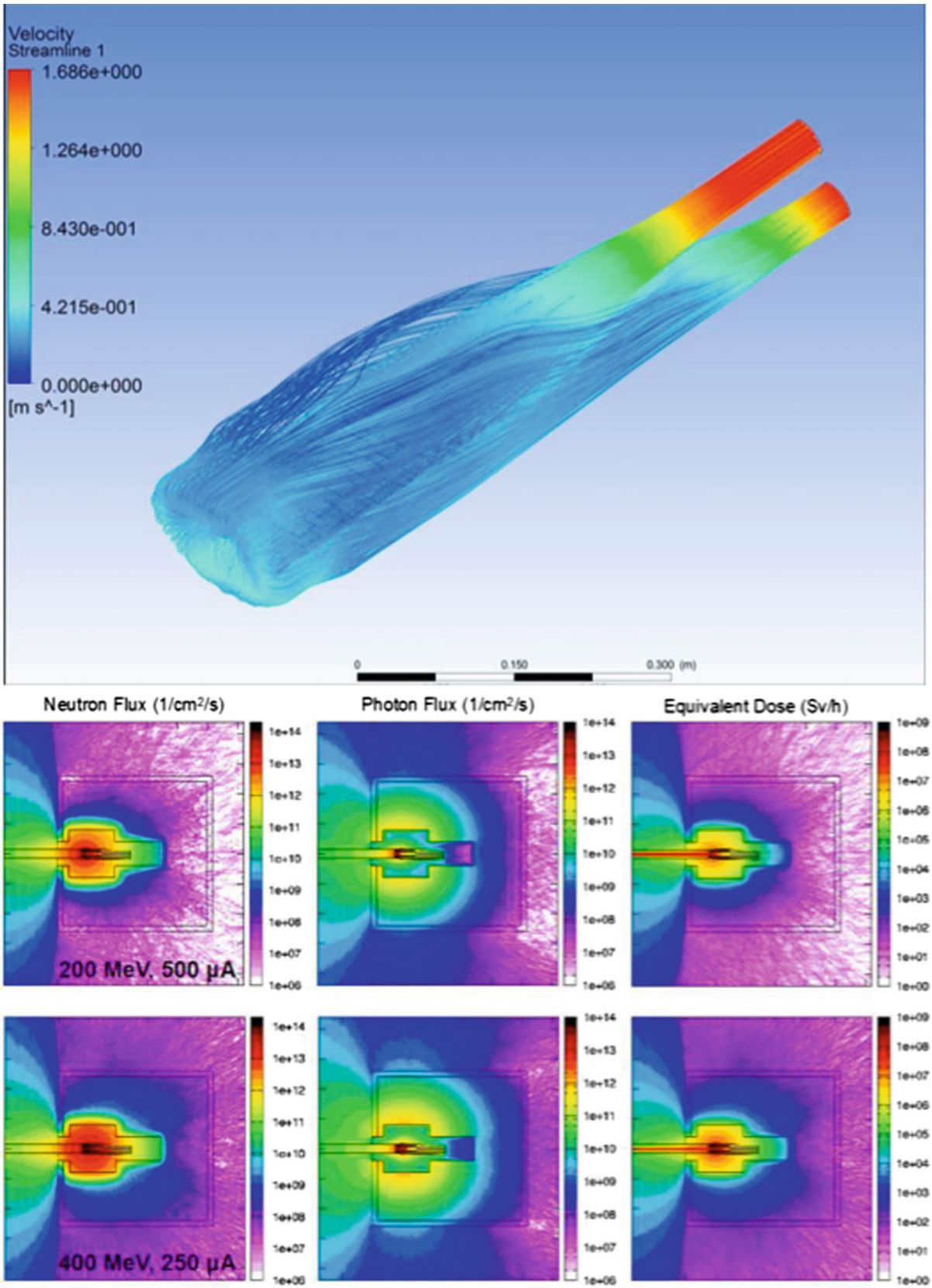

Fig. 3 Flow in target (*top*) and neutron flux through facility (*bottom*)

A number of analyses were conducted to verify the validity of the design, including computational fluid dynamics (Fig. 3 top), neutronics (Fig. 3 bottom), and finite element analysis for specific critical elements such as the beam window.

A 800 MeV/u, 16 MW Cyclotron Complex

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Abstract

This paper addresses an attempt to start investigating the use of the superconducting ring cyclotron (SRC) developed for the DAEδALUS experiment, for ADS-reactor/transmuter applications.

A cyclotron complex capable of 8 MW continuous-wave (CW) proton beam power delivery is being designed [1], consisting of an injector cyclotron (IC) able to accelerate a 5 electrical milli-Ampere H_2^+ beam to 60 MeV/u, a superconducting ring cyclotron (SRC) booster able to accelerate the beam to 800 MeV/u, and stripping extraction to deliver a 10 mA proton beam.

The use of an H_2^+ beam has two advantages: (1) beam extraction from the SRC can be accomplished by stripping foil, avoiding the need for a clean separation between turns, which is mandatory for high-efficiency proton beam extraction; (2) space charge effects are significantly reduced due to the value of $q/A = 0.5$ (with q the charge, A the atomic mass number). At higher beam currents the space charge reduces the focusing effects and broadens the beam size. A measure of its strength is the so-called "generalized perveance", which, for particles with the same speed, is proportional to qI/m (with I the beam current and m the particle mass). Thus, the space charge effects for 5 mA of H_2^+ beam are equivalent to a 2.5 mA proton beam with the same speed, whereas a 10 mA proton beam current is extracted from the SRC. According to simulations performed with the OPAL code [2], the 5 mA H_2^+ beam accelerated along the SRC is expected to undergo similar space-charge effects as the 2.4 mA proton beam accelerated currently at the Paul Scherrer Institute (PSI).

Similarly, beam acceleration through the IC will have characteristics similar to the acceleration of PSI's "Injector 2" (protons, 0.87–72 MeV). Although the use of a separated-sector cyclotron could be a robust and safe solution, to save money and space, a compact cyclotron design is considered. The central region and the characteristics of the injected beam are quite similar to commercial cyclotrons used for radioisotope production [3]. These are routinely able to accelerate up to 1 mA and have gone up to 2 mA for H^- beams. If we compare the space charge effects of a 2 mA H^- beam injected at 25–30 keV with the space charge effects of a 5 mA H_2^+ beam injected at 35 keV/u, the generalized perveance values are very similar. As the central region of a compact cyclotron is very small, the crucial point is in the injection and acceleration of 5 mA H_2^+ along the first few laps (turns). An experimental program is in progress to demonstrate the feasibility and reliability of this central region.

The beam dynamics through the injector cyclotron have been simulated by using the OPAL code. The magnetic field boundary near the extraction region is designed to produce a crossing of the resonance $Q_r = 1$, which produces a precession of the orbit motion. The combination of the large orbit radius, about 2 m, together with the high energy gain per turn (2 MeV/turn, 4 RF double-gap cavities operated at 250 kV), and added to the effect of the orbit precession, produce an inter-turn separation of 20 mm. In this condition it is realistic to consider extracting the beam with an efficiency of about 99.98 %. The protection of the electrostatic

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deflector and of its septum is simplified by the possibility to remove the beam halo by using stripper foil located upstream of the deflector device.

An additional crucial item is the injection of the beam into the SRC. The final part of the injection path has been studied, details of which can be found in a poster contribution for these proceedings [4].

The SRC is similar in size to the SRC at RIKEN [5]. Space-charge-dominated beam dynamics simulations using OPAL have been performed for an earlier, eight-sector geometry, and indicate acceptable transmission and low beam losses. Engineering magnet design studies regarding a six-sector configuration [6] have been conducted on the present design.

The code OPERA was used to simulate the magnetic field of the cyclotron and the beam dynamics features were investigated by using the OPAL code. In the SRC, the beam loss at extraction is relaxed by using stripping extraction, which has an efficiency very close to 100 %, even though the beam orbits at the extraction radius are not fully separated. The two main problems that we investigated are: (1) the vertical focusing at higher energies, which is usually weak in a superconducting cyclotron, and (2) the relatively large energy spread of the extracted proton beam, due to the multi-turn extraction. Distribution of the H_2^+ beam in the vertical and radial planes, at a fixed azimuthal position, for 0 and 5 mA were computed. Beam halos extend vertically ±20 mm, although they clear the vacuum chamber, which has a 70 mm gap. It is quite evident from the simulation that, in the SRC, space charge and neighboring bunch effects introduce vertical beam halo, but these effects have a negligible impact on beam losses.

Among other things, the new, six-sector design allows more room for installation of the PSI-like radiofrequency (RF) cavities and reduces the cost compared with the earlier eight-sector design [7]. The Technology and Engineering Division (T&ED) of the MIT Plasma Science and Fusion group developed the magnet conceptual design of the six-sector SRC to provide a realistic solution both for the coils and for the cryostat. The latter satisfies the cryogenic request and, at the same time, is robust enough to balance the enormous magnetic forces. This new configuration allows the installation of four PSI-like RF cavities, able to provide 1 MV acceleration voltage, so minimizing the length of the acceleration path and reducing beam–gas interaction losses. The injection inflector occupies a valley, whereas a magnet pair in the upstream valley allows proper adjustment of injection [4]. Further optimization of the boundary of the iron pole is in progress to achieve an isochronism better than 10^{-3} and a phase shift lower than 20°. This procedure consists of shimming the boundary of the cyclotron pole sectors carefully in both the design and construction phases.

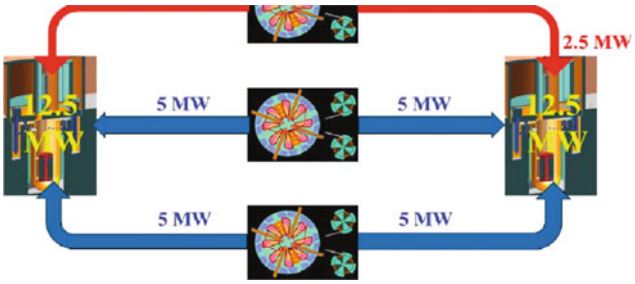

Fig. 1 Possible arrangement of a pair (of a series) of 12.5 MW beam power subcritical units, based on an SRC and its two 60 MeV/u injectors. In case of failure in a cyclotron, it is possible to increase the beam power from the others and maintain a total 12.5 MW per unit

The cyclotron complex described will be able to supply a maximum beam current of 10 mA. The beam current limit is due to the losses inside the injector cyclotron and is to the upper limit of the H_2^+ source. To deliver higher power, we plan to use two injector cyclotrons operated with a RF frequency half the frequency of the SRC. In this way, it is feasible to merge the two beams into a single 10 mA beam, which is subsequently injected into the booster, so delivering about 20 mA, 16 MW power proton beam. In this operation condition, one has to address the beam–gas losses. A vacuum below 10^{-8} Torr is necessary to minimize this effect. The RF cavities will be equipped with vacuum pumps that, according to the experience of PSI, can achieve a vacuum of 2.5×10^{-6} Torr. To achieve a vacuum that is two orders of magnitude better, we plan to install Non Evaporable Getter (NEG) pumping in the empty space between the RF cavities and the sector magnets.

In the ADS application, a multiple-cyclotron scheme allows maintenance without powering down the plant. A solution is to account for four reactors driven by a five cyclotrons accelerator complex (Fig. 1). In the present case of a six-sector cyclotron design, the power is limited by the vacuum, an upper limit is near 12–14 MW. If the energy is increased, for instance up to 1.5 GeV assuming nine sectors instead, the power limit could increased to 15–16 MW.

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A 4 MW High-Power Spallation Source for an ADS Demonstrator

K. Samec and Y. Kadi

Abstract

A concept for a high-power spallation source suitable for an accelerator-driven system (ADS) demonstrator is proposed, based on the 1 MW MEGAPIE liquid metal target tested under irradiation and the thermal-hydraulic test of the 4 MW EURISOL liquid metal target. The new design features an elongated geometry suited to producing high-density neutron fluxes in a subcritical core. The design has been calculated to absorb 4 MW of proton beam power and can be scaled up to 10 MW, sufficient to power a full-scale industrial prototype of an ADS demonstrator.

Introduction

Defense in depth is achieved in the design according to [1]; the occurrence of an accident is minimized, its progress limited, and the effects thereof held well below legal limits. Lessons learnt from the high-power liquid metal targets have been integrated in the concept; MEGAPIE, tested under a proton beam, and EURISOL, tested hydraulically at liquid metal velocities of up to 6 m/s [2]. Standard nuclear practices such as multiple barriers and redundant safety systems were adopted.

Main Features of the Design

The MEGAPIE configuration has been improved upon by implementing an overhead proton beam, which does not penetrate the containment; an external heat exchanger and an external electromagnetic pump, which are, thus, physically separated from the liquid metal circulating in the spallation source. From EURISOL, a 4 MW liquid metal, the cusp-shaped beam window was retained, which can withstand a 4 MW beam and, hence, achieve neutron flux densities over 10^{15} (n/cm^2/s) (Fig. 1).

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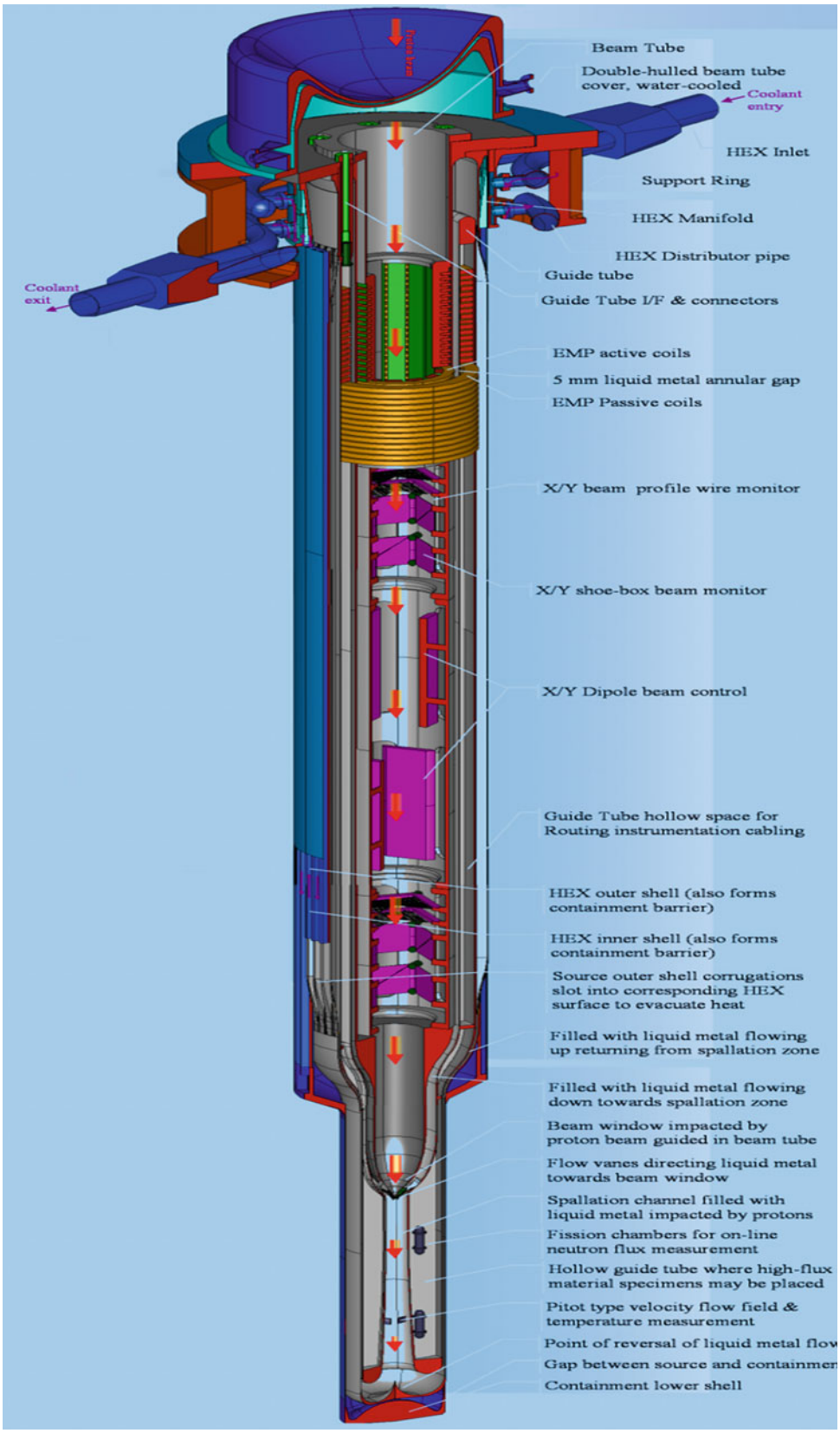

Fig. 1 Proposed design of the high-power spallation source for an ADS [1]

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Subcriticality Monitoring for the Accelerator-Driven Thorium Reactor (ADTR™) Control

R. Ashworth

Measuring the reactivity of the accelerator-driven thorium reactor (ADTR) core accurately ensures safe, optimal performance of the reactor. The preferred method is to momentarily drop the accelerator beam current. The result of this reduction is to see a fast, step-like, reduction in core neutron flux followed by a more gradual reduction to equilibrium. This delayed neutron effect is complemented by core temperature fluctuations to enhance the measurement accuracy of core reactivity and k_{eff}. The method is applicable to other accelerator-driven system (ADS) reactors.

The ADTR [1] has a design target to maintain a margin to prompt criticality equal to that enjoyed by a typical U-235-fuelled pressurized water reactor (PWR). This resulted in an optional setting of $k_{\text{eff}} = 0.995$ to maintain safe operation while achieving a subcritical amplification gain of ~ 500, hence, maximising efficiency. The prompt margin can be defined as the reactivity, ρ, minus the delayed neutron fraction, β, of the fissile fuel.

$$\rho = 1 - 1/k_{\text{eff}} \tag{1}$$

Table 1 shows that the target margin to prompt criticality is met for the full operating life of the fuel cycle, starting with a plutonium fissile starter and ending with U-233 fuel, when using $k_{\text{eff}} = 0.995$. Regular monitoring of the core reactivity will be required.

Maintaining the subcritical margin by accurate monitoring and control is crucial. Control utilising absorber rods is discussed in [1], and accuracy of measurement is equally important. Accelerator beam current step fluctuation is the chosen measurement method. Figure 1 shows the reactor core neutron flux behaviour following a step reduction in beam current. The neutron flux undergoes a prompt drop from the initial flux of $R0$ to $R1$ at time $t = 0$. The flux then follows a decay curve the form of which is dependent on the fuel temperature profile following the reduction in beam current and power. Five different temperature profiles are investigated, typically $\Delta T = 160$ °C represents the ADTR design load at the beginning of a fuel cycle and 240 °C at the end of the cycle. Reactivity can be derived from this flux behaviour as follows:

$$\rho = \frac{\beta_{\text{eff}}(1 - X)}{1 - X - \Delta i_{\text{p}}/i_{\text{p}}} \tag{2}$$

where $X = \frac{R1}{R0}$, i_{p} = beam current and Δi_{p} = step change in beam current [2].

The parameter that represents the greatest uncertainty in the reactivity calculation of (2) is the value of $R1$. From Fig. 1, determining the value for $R1$ is highly dependent on ΔT; indeed, at 0 °C it is the point where the vertical line blends into the exponential decay curve, whereas at 400 °C, it is a point where the flux behaviour reverses, stops dropping, and then begins to rise again.

The proposed method to determine $R1$ is to deduce a straight line from the flux behaviour during the first few seconds following the step change and to extrapolate to the intersection with the vertical at time $t = 0$. Assuming sufficient neutron counts are recorded to generate the straight line, for $\Delta T > 150$ °C, this method should give an $R1$ accuracy of better than ±2 units from Fig. 1. Translating this to reactivity, we can see that a reading of 208 units for $R1$ with U-233 gives $\rho = -5.3 \times 10^{-3}$ and an error of ±10 % for the calculation of reactivity. When operating at $k_{\text{eff}} = 0.995$,

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Table 1 Prompt margins at $k_{eff} = 0.995$

Fissile isotope	β	ρ	Prompt margin
U-235 (PWR)	6.7×10^{-3}	0.0	-6.7×10^{-3}
U-233 (ADTR)	2.9×10^{-3}	-5.0×10^{-3}	-7.9×10^{-3}
Pu-239 (ADTR)	2.2×10^{-3}	-5.0×10^{-3}	-7.2×10^{-3}

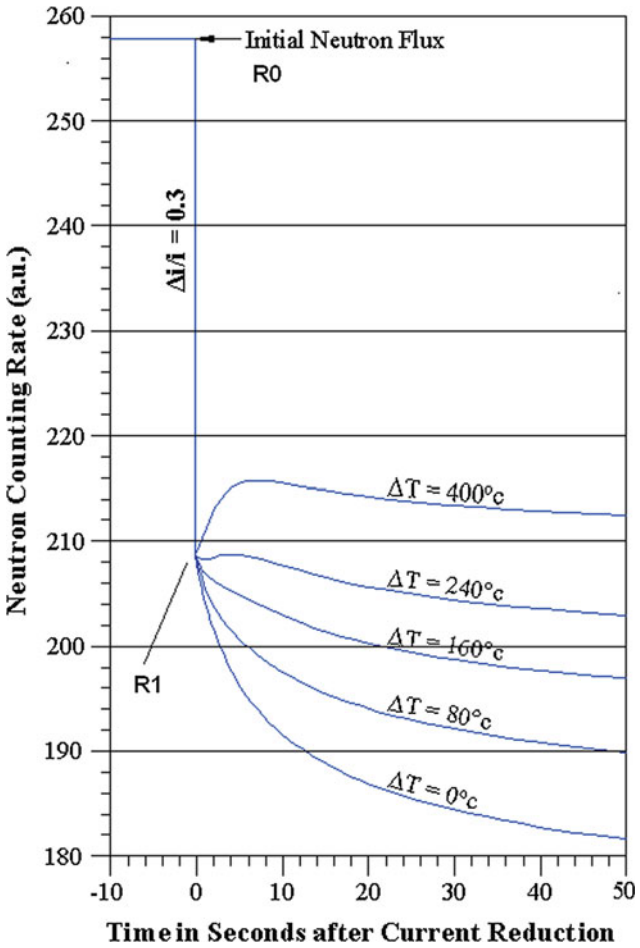

Fig. 1 Neutron flux behaviour following step reduction in the proton beam current [2]

we have a 10 % margin over a U-235 PWR, Table 1. Hence, using the method described here, we are able to meet our operational safety targets, while maintaining a healthy energy amplifier gain of around 500.

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Utilization Potential of Thorium in CANDU Reactors and in Fusion–Fission (Hybrid) Reactors

Sümer Şahin and Hacı Mehmet Şahin

Abstract

World thorium reserves are approximately three times more abundant than the natural uranium reserves. Turkey is rich is thorium resources. Hence, thorium remains a potential energy source for future energy strategies in Turkey.

Although thorium is actually not used as nuclear fuel in conventional reactors, it has very promising potentials for utilization in CANDU reactors as a mixed fuel with non-conventional nuclear materials.

Nuclear power plants are producing nuclear waste materials in substantial quantities in the form of minor actinides (MA). MA can be defined as the most hazardous radioactive waste products because of their long-term, high-level radioactivity. A significant fraction of reactor waste MA consists of diverse plutonium isotopes, which represents serious public and political concern with respect to misuse of this plutonium and also accidental release of highly radiotoxic material into the environment.

Large quantities of reactor grade (RG) plutonium have been accumulated in the course of nuclear electricity generation over the past 50 years in civilian reactors as nuclear waste. This became a serious concern already in 1970s, where accumulation of approximately 1700 tons of reactor grade plutonium by the year 2000 was postulated [1–3]. In a later report, it was cited, "In August 1996, there were 439 nuclear power plants in 32 countries. The world's power reactors consume the equivalent of 60,000 tons of uranium each year. The total amount of plutonium produced worldwide is about 1270 tons at present (mostly unseparated), and is accumulating at 70 tons per year. It is estimated that civilian plutonium separation programs will produce 190,000 kg of plutonium during the 1990s" [4]. European sources report "At present, the total park of nuclear power plants in the European Union ($\sim$125 GW of nominal power) produces approximately 2500 tons of spent fuel annually (oxygen of the oxide not being accounted for in this figure), containing about 25 tons of plutonium and 3.5 tons of the minor actinides (MA), neptunium, americium, and curium, and 3 tons of long-lived fission products" [5–7].

Plutonium and heavy water moderators can give a good combination with respect to neutron economy so that mixed fuel made of ThO_2/RG-PuO_2 or ThC/RG-PuC would lead to very high burnup grades. In that case, it is advisable to use advanced nuclear fuel technologies rather than conventional fuel pellets. The TRISO type fuel has demonstrated by Peach Bottom Reactor experiments to withstand very high fuel burnup levels. In this work, the utilization of TRISO fuel made of mixed Th/RG-Pu and Th/MA in CANDU reactors with the following scenarios has been investigated:

1. In the first scenario, five different mixed fuel have been selected for CANDU reactors composed of: 4:96 % RG-PuO_2/ThO_2; 6:94 % RG-PuO_2/ThO_2; 10:90 % RG-PuO_2/ThO_2; 20:80 % RG-PuO_2/ThO_2; and 30:70 % RG-PuO_2/ThO_2. Each fuel is uniformly added into each fuel rod in a fuel channel. The corresponding operation lifetimes have been found to be approximately 0.65, 1.1, 1.9, 3.5, and 4.8 years and with burnups of about 30,000, 60,000, 100,000, 200,000, and 290,000 MWd/tonne, respectively. Higher initial plutonium charge leads to higher burnups.

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2. In the second scenario, a mixed fuel composition has been varied in radial direction: 100 % RG-PuO_2; 80:20 % RG-PuO_2/ThO_2; 60:40 % RG-PuO_2/ThO_2; and 40:60 % RG-PuO_2/ThO_2. The fuels were placed in the first, second, third, and fourth rows, respectively, for power flattening in the CANDU fuel bundle. In this case, reactor operation lifetime increases to >8 years and burnup to 580,000 MWd/tonne.
3. In the third scenario, three carbide mixed fuels have been selected: 10:90 % RG-PuC/ThC; 30:70 % RG-PuC/ThC; and 50:50 % RG-PuC/ThC. The corresponding operation lifetimes are approximately 2.7, 8.4, and 15 years with burnups of about 72,000, 222,000, and 366,000 MWd/tonne, respectively.
4. In the fourth scenario, a mixed fuel composed of 95:5 % ThO_2/minor actinides, MAO_2, has been selected. This leads to full power operation for a period of 10 years. After the second year, the CANDU reactor begins to operate practically as a thorium burner. Also in this case, very high burnup could be achieved without fuel change or renewal (up to 200,000 MWd/tonne).

These scenarios have demonstrated the possibility of large-scale thorium utilization, based on the well-established present-day CANDU reactor technology.

In critical fission reactors with thermal neutron spectra, two neutrons are needed to fission thorium as the threshold fission energy lies at $\sim$1.4 MeV. On the other hand, (D, T) fusion reactions produce highly energetic neutrons at 14.1 MeV, for which thorium has a quasi-comparable fission cross section with uranium so that one fusion neutron will be needed to the release fission energy out of the neutron. Calculations on a laser fusion driven fusion–fission (hybrid) reactor have led to fissile burnups exceeding 400,000 MWd/
tonne. In addition to fission energy production in situ, such a reactor would produce $\sim$160 kg ^{233}U per year.

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Stop Designing Reactors!

John Laurie

Leo Szilard was a physicist. On 12 September 1933, he had a remarkable idea, which would become known as the nuclear chain reaction. Enrico Fermi was also a physicist. He headed a team that designed and built the world's first nuclear reactor, achieving criticality on 2 December 1942. The staggering intellectual achievements of these and many other physicists brought humanity into the nuclear age.

The Manhattan Project laid the foundations for the design philosophy for the first nuclear era. The physicists had worked out and mathematically proven the fascinating new science of neutronics, so they were the ones tasked with designing the first nuclear bombs. Armies of people from other scientific and technical disciplines labored to provide the materials to build them. The bombs worked, the Second World War was brought to an end, and the physicists were heroes.

This apparently successful design philosophy was then applied to the peaceful use of nuclear fission for the generation of power. The physicists would design the reactor, and then everyone else would design and build a nuclear energy system around it.

In the first nuclear era, fission is physics.
The champion of the first nuclear era is the Reactor Physicist.

The nuclear energy system includes all activities, equipment, disciplines, and resources associated with the generation of nuclear power. For example: mining, milling, transport, enrichment, fuel fabrication, facility fabrication, irradiation, power conversion, maintenance, retreatment, geological isolation—basically everything from digging up the fuel to putting the waste back into the ground.

It was thought that the huge energy density of uranium—around 1 million times that of fossil fuels—would be enough to ensure that nuclear power became the dominant means of power generation within a few decades, even if the reactor design that was chosen for roll out resulted in an energy system that wasn't actually very efficient, or safe, or clean, or sustainable, or cheap…

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In many countries, nuclear energy systems have been imposed on the population by central governments. The electrical power generated has brought huge benefits, but public criticisms of nuclear energy systems have been largely ignored, communication has been very poor, and mistrust has grown, fueled by environmental groups and fossil fuel lobbies.

Today, 435 nuclear power plants provide around 11 % of the world's electricity, but progress has stalled at a time when, more than ever before, the world needs abundant energy that is free from greenhouse gas emissions.

The failure of the first nuclear era is its design philosophy.

In a world that is largely and increasingly democratic and market driven, putting the technology first and the customer second is a flawed strategy. People want to consume energy that is **reliable, cheap, safe, sustainable, and clean**. It is essential to start with a deep and fundamental understanding of those customer requirements before even starting to think about how to fulfill them.

For over 60 years, we have been designing the wrong thing. To begin the second nuclear era, a paradigm change is required. Instead of designing reactors, we need to be thinking primarily about **the design of the nuclear energy system as a whole**. Obviously, those systems will have a nuclear reactor as a very important component, but the detailed design of the reactor should not be pursued unless the overall system gives very substantial improvements in **all areas of customer requirements**. To do otherwise is a waste of scarce scientific and technical resources.

The ThEC13 conference at CERN discussed three approaches for extracting energy from thorium (Table 1).

Concerning accelerator-driven systems, there was considerable discussion of potential benefits through design of reactors that are safe, sustainable, and clean. But by ignoring cost and reliability, physicists working on these reactor designs seem to be perpetuating the failures of the first nuclear era.

In a nuclear energy system, many different disciplines are involved. Designing an effective system therefore requires a multidisciplinary approach. Important synergies are possible through close collaboration between experts.

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Table 1 Three approaches for extracting energy from thorium

Possible systems for extracting energy from thorium, compared with generation 3 uranium pressurized water reactors (PWRs)	Solid thorium fuel in PWRs	Liquid fuel	Accelerator-driven systems
Reliable	😐	☺	☹
Cheap	😐	☺?	☹
Safe	😐	☺	☺
Sustainable	☺	☺	☺
Clean	☺	☺	☺

> In the second nuclear era, fission will be re-positioned at the interface between physics and chemistry. The champion of the second nuclear era will be the Nuclear Energy System Architect.

This message is going to be rather unpopular with physicists, who have, after all, been at the center of massive advances for humanity in the first nuclear era. But fighting climate change and energy poverty in the 21st century may well **depend** on whether the physics community can be humble enough to **share** nuclear fission with other disciplines.