Current Czech R&D in Thorium Molten Salt Reactor (MSR) Technology and Future Directions

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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 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.

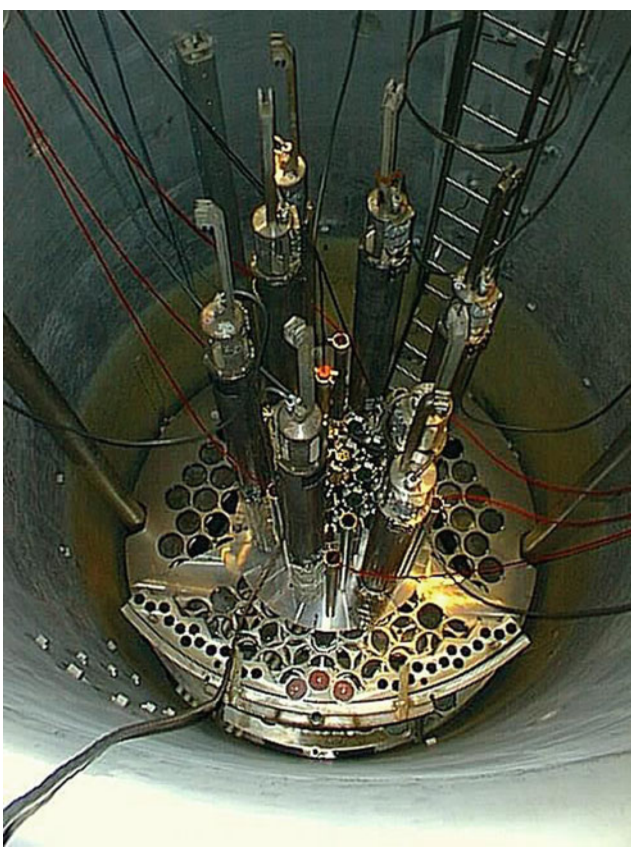

Fig. 1 View into the LR-0 reactor core with inserted salt zone

Fig. 2 Manipulation with the salt zone before insertion into reactor core

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,

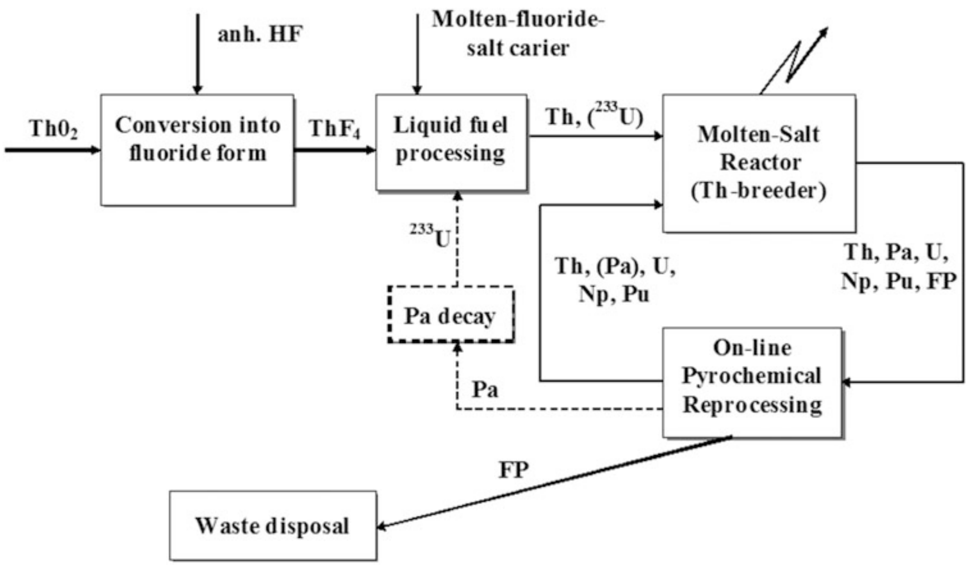

Fig. 3 Fuel cycle scheme for a MSR Th breeder. *FP* fission products; *anh* anhydrous

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.

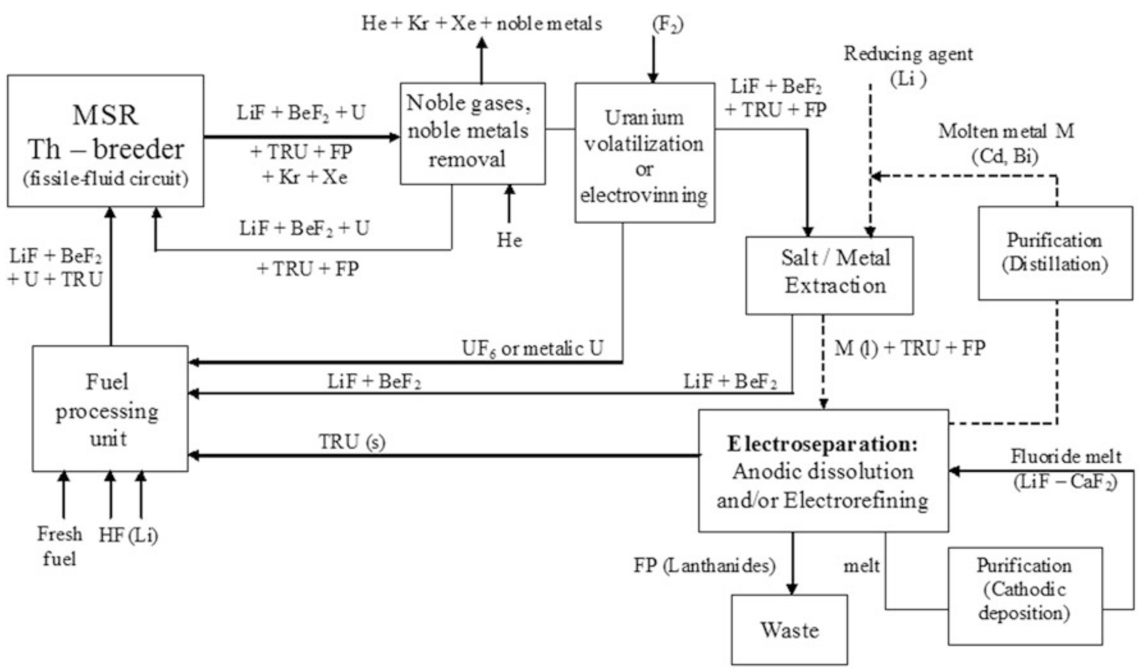

Fig. 4 Conceptual flowsheet for MSR Th breeder fissile-fluid circuit on-line reprocessing technology. *TRU* transuranium elements; *FP* fission products

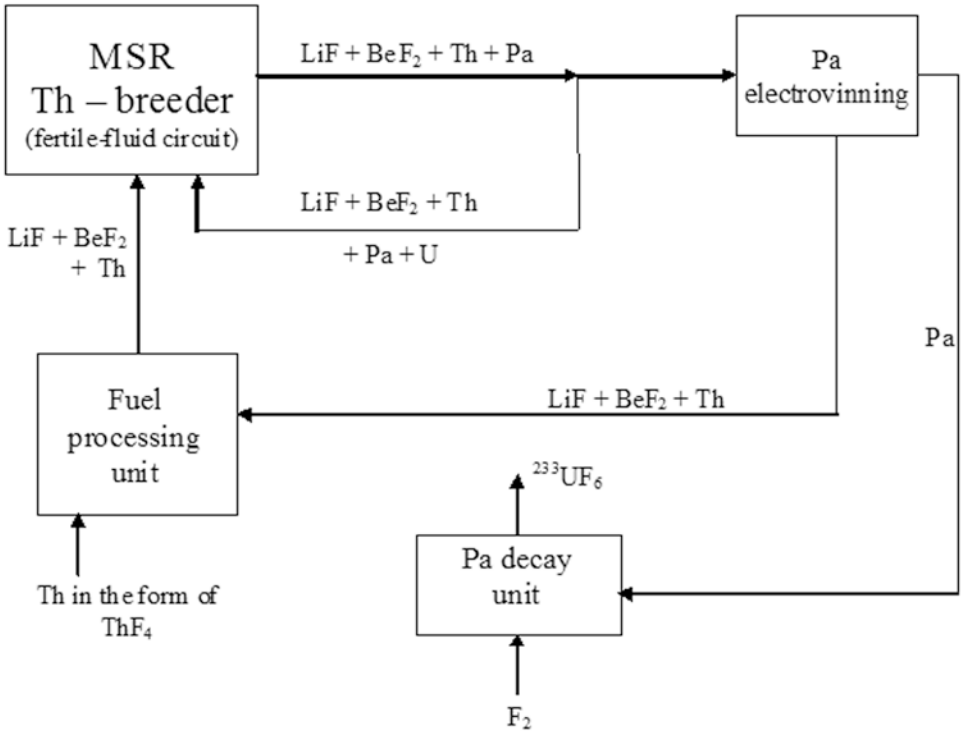

Fig. 5 Conceptual flowsheet for MSR Th breeder fertile-fluid circuit on-line reprocessing technology. *TRU* transuranium elements; *FP* fission products

- 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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