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.

H. Xu (✉)
CAS Center for Excellence in TMSR Energy System, Shanghai Institute of Applied Physics, Chinese Academy of Sciences,
Shanghai, China

J.-P. Revol et al. (eds.), *Thorium Energy for the World*, DOI 10.1007/978-3-319-26542-1_6

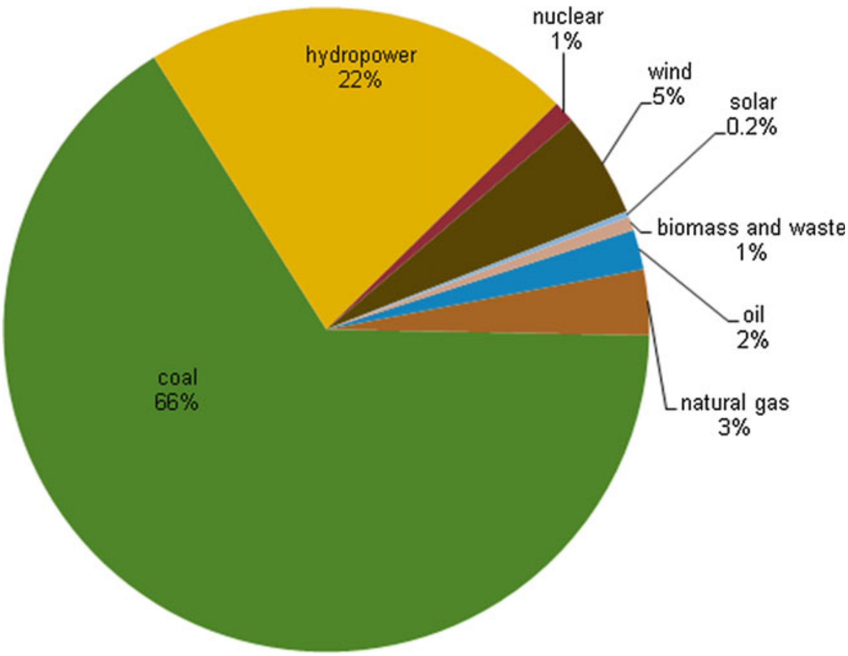


Fig. 1 China's installed electricity capacity by fuel in 2012 (installed capacity: 1,145 GW)

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.

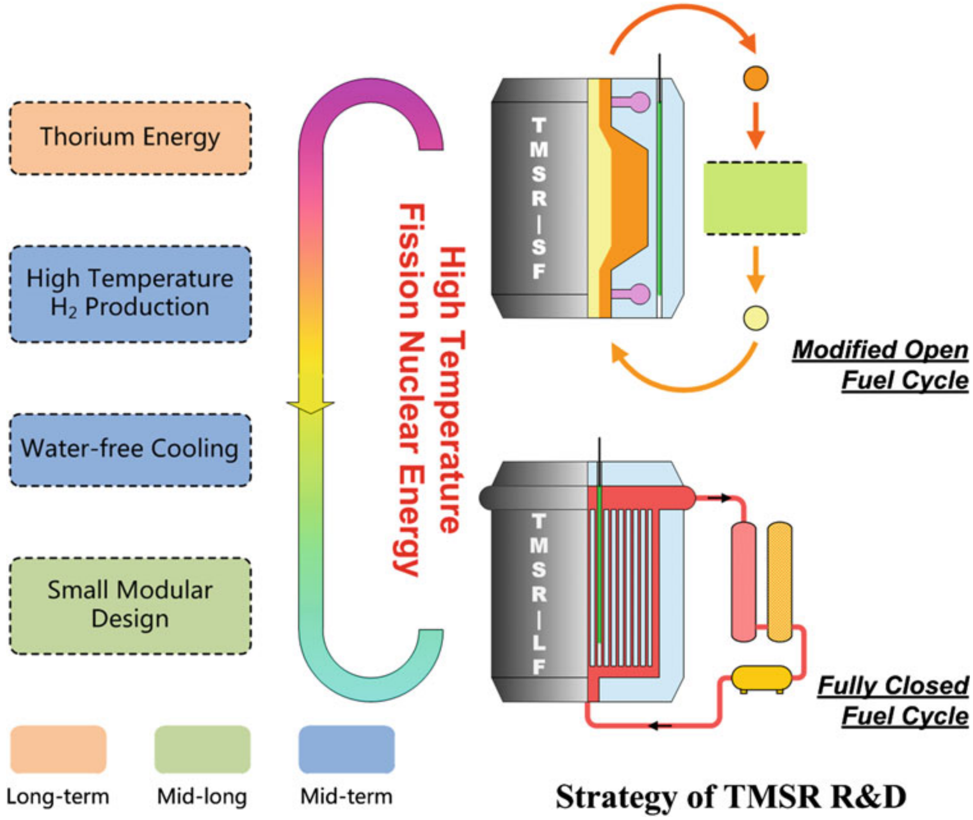
Being one of the six candidates of Generation IV reactors, molten salt reactors (MSRs) have two main subclasses. The first subclass is known as a liquid-fueled MSR (MSR-LF) [9, 10], in which nuclear fuel is dissolved in the molten fluoride salt and the salt serves both as the fuel and the coolant in 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



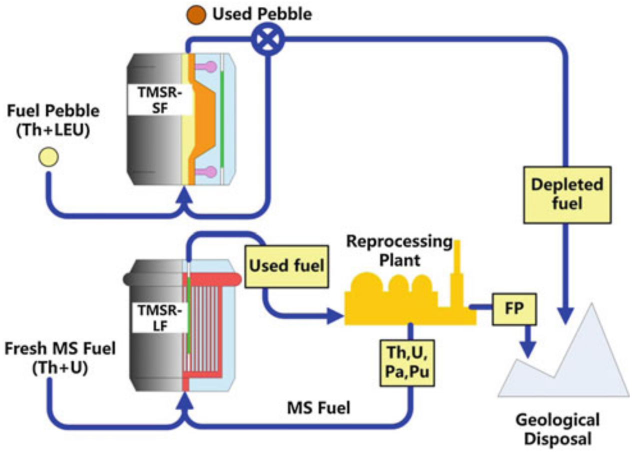


Fig. 3 Material flow of TMSRs with various fuel cycles

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.

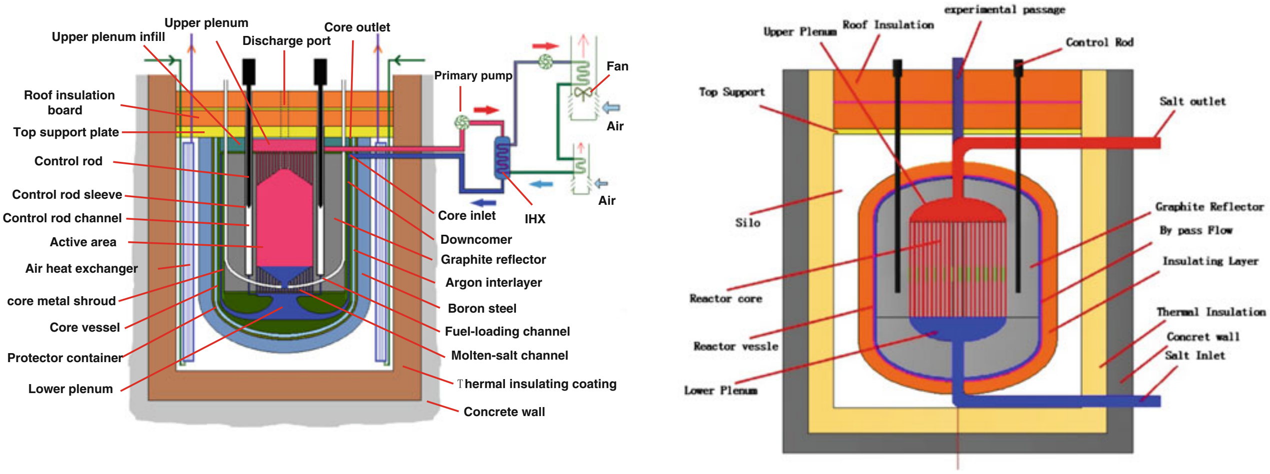


Fig. 4 Schematic structure of the TMSR-SF1 (*left*) and the TMSR-LF1 (*right*)

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

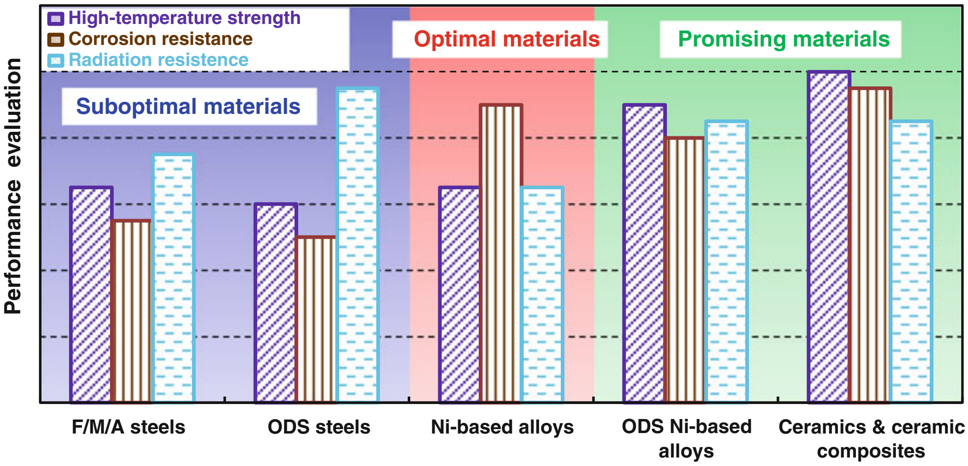
Fig. 6 Picture of the horizontal distillation facility

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

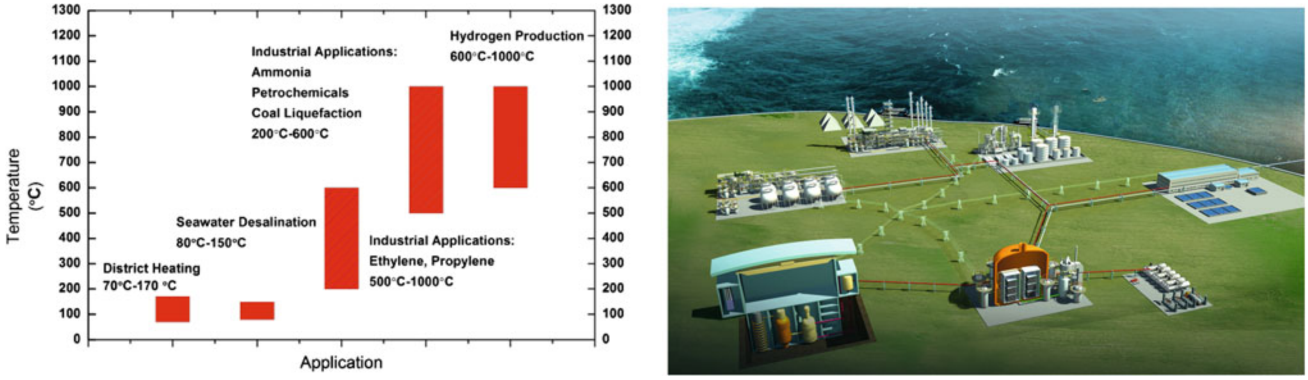


Fig. 9 *Left* Applications of nuclear heat in different temperature ranges. *Right* A sketch for a nuclear hybrid energy system based on TMSR

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.

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.

Fig. 10 kW integrated SOEC evaluation platform

Acknowledgments This work is supported by the Thorium Molten Salt Reactor Nuclear Energy System under the Strategic Pioneer Sci. and Tech. Project of the Chinese Academy of Sciences under contract no. XDA02000000

References

1. US Energy Information Administration report
2. IAEA: Power Reactor Information system
3. World Nuclear Association (WNA): Nuclear Century Outlook Data
4. David, S.: Future scenarios for fission-based reactors. Nucl. Phys. A **751**, 429–441 (2005)
5. Kaimal, K.N.G., et al.: Effect of irradiation and dopant concentration on the migration of xenon in uranium dioxide. Metall. Mater. Proc. **1**, 293–300 (1990)
6. Matzke, H.J.: Diffusion processes in nuclear fuels. In: Agarwala, R. P. (ed.) Diffusion Processes in Nuclear Materials, pp. 9–69. North Holland, Amsterdam (1992)
7. Shiba, K.: Diffusion processes in thoria and thorium-based oxides with emphasis on fission fragments irradiation effects. In: Agarwala, R.P. (ed.) Diffusion Processes in Nuclear Materials, pp. 71–97. North Holland, Amsterdam (1992)

8. Anantharaman, K.: Utilization of thorium in reactors. J. Nucl. Mater. **383**, 119–121 (2008)
9. Robertson, R.C.: Conceptual design study of a single-fluid molten-salt breeder reactor, ORNL-4541 (1971)
10. Heuer, D., et al.: Towards the thorium fuel cycle with molten salt fast reactors. Ann. Nucl. Energy **64**, 421–429 (2014)
11. Scarlat, R.O., Peterson, P.F.: The current status of FHR technology and its overlap with HIF target chamber concepts. Nucl. Instrum. Meth. A **733**, 57–64 (2014)
12. Xu, H.J., et al.: Some physical issues of the thorium molten salt reactor nuclear energy system. Nucl. Phys. News **24**(2), 24–30 (2014)
13. Murty, K.L., Charit, I.: Structural materials for gen-IV nuclear reactors: challenges and opportunities. J. Nucl. Mater. **383**, 189–195 (2008)
14. Olson, L.C., Ambrosek, J.W., Sridharan, K., Anderson, M.H., Allen, T.R.: Materials corrosion in molten LiF–NaF–KF salt. J. Fluor. Chem. **130**, 67–73 (2009)
15. Non-electric applications of nuclear power: seawater desalination, hydrogen production and other industrial application. In: Proceedings of an International Conference, on Non-electrical Applications of Nuclear Power, Oarai, Japan (2007)
16. Yan, X.L., Hino, R.: Nuclear Hydrogen Production Handbook. CRC Press, Boca Raton (2011)