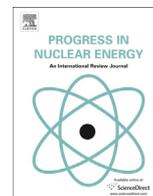




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The molten salt reactor (MSR) in generation IV: Overview and perspectives



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ABSTRACT

Molten Salt Reactors (MSR) with the fuel dissolved in the liquid salt and fluoride-salt-cooled High-temperature Reactors (FHR) have many research themes in common. This paper gives an overview of the international R&D efforts on these reactor types carried out in the framework of Generation-IV. Many countries worldwide contribute to this reactor technology, among which the European Union, France, Japan, Russia and the USA, and for the past few years China and India have also contributed. In general, the international R&D focuses on three main lines of research. The USA focuses on the FHR, which will be a nearer-term application of liquid salt as a reactor coolant, while China also focuses on solid fuel reactors as a precursor to molten salt reactors with liquid fuel and a thermal neutron spectrum. The EU, France and Russia are focusing on the development of a fast spectrum molten salt reactor capable of either breeding or transmutation of actinides from spent nuclear fuel.

Future research topics focus on liquid salt technology and materials behavior, the fuel and fuel cycle chemistry and modeling, and the numerical simulation and safety design aspects of the reactor. MSR development attracts more and more attention every year, because it is generally considered as most sustainable of the six Generation-IV designs with intrinsic safety features. Continuing joint efforts are needed to advance common molten salt reactor technologies.

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Abbreviations: CR, Conversion Ratio; EFPY, Equivalent Full Power Year; ISTC, International Science and Technical Centre, Moscow, Russia; IGC, Intergranular Cracking; LWR, Light Water Reactor; MA, Minor Actinides; MARS, Minor Actinides Recycling in Salts, Rosatom R&D project, 2010–2013, Russia; MOSART, Molten Salt Actinide Recycler Transforming system, developed at KI, Russia; MOX, Mixed Oxide fuel; MSBR, Molten Salt Breeder Reactor, developed in ORNL, USA; MSFR, Molten Salt Fast Reactor, developed in CNRS, France; MSR, Molten Salt Reactor; MSRE, Molten Salt Reactor Experiment, developed in ORNL, USA; REE, Rare Earth Elements; RW, Radioactive Waste; SNF, Spent Nuclear Fuel; TRU, Transuranic Elements; ORNL, Oak Ridge National Laboratory, USA; UOX, Uranium Oxide fuel.

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1. Introduction

Molten salt reactors (MSR) are a class of nuclear fission reactors in which the primary coolant, or even the fuel itself, is a molten salt mixture. MSRs have two primary subclasses. In the first subclass, fissile material is dissolved in the molten salt. In the second subclass, the molten salt serves as the low pressure coolant to a coated particle fueled core similar to that employed in High-Temperature Reactors (HTRs). In order to distinguish the reactor types, the solid fuel variant is typically referred to as a Fluoride salt-cooled High-temperature Reactor (FHR) (Forsberg, 2005). MSRs run at much higher temperatures (up to 700–750 °C) than light water reactors (LWR) and operate at near atmospheric pressure. Molten salts offer attractive characteristics as coolants especially their high volumetric heat capacities and high boiling points.

In liquid fueled MSRs designs, the nuclear fuel is dissolved in the molten fluoride salt coolant as fissile elements such as UF₄, PuF₃, minor actinides fluorides and/or fertile elements as ThF₄ depending on the desired application (breeder reactor, actinide burner, etc). In the core, fission occurs within the fuel salt, which then flows into an intermediary heat exchanger, where the heat is transferred to a secondary liquid-salt coolant. Molten fluoride salts were first developed because fluorine has only one stable isotope (¹⁹F) contrary to chloride based salts. Liquid fluoride solutions are familiar to the aluminum and uranium industries. All uranium used in today's reactors has been converted to and from a fluoride form in order to be enriched.

Liquid fueled MSRs are often associated with the ²³³U–²³²Th fuel cycle. The mass number of ²³²Th is six units less than that of ²³⁸U, thus many more neutron captures are required to transmute thorium to the first transuranics. Although MSRs can be fueled by any fissile material, the use of abundant thorium as fertile element enables breeding with a thermal spectrum, and is often considered as more convenient than the U–Pu fuel cycle in order to minimize the generation of highly radiotoxic transuranic elements (Hargraves and Moir, 2010). However, unlike natural uranium which contains the fissile isotope ²³⁵U, thorium does not contain any fissile isotopes and its utilization requires the aid of fissile material from the uranium cycle for initial start-up.

The potential use of a fluid-fueled reactor has been recognized for a long time. The molten-salt system has the usual benefits attributed to fluid-fuel systems over solid-fuel element systems:

- A high coefficient of thermal expansion which provides a large negative temperature coefficient of reactivity. Because the fuel is liquid, it expands when heated, thus slowing down the rate of nuclear reactions and making the reactor self-regulating,
- The possibility of continuous fission-product removal using physical (helium sparging) and pyrochemical processes. Fuel salt can be processed in an online mode or in batches in order to retrieve fission products and actinides. Actinides are then reintroduced into the fuel circuit,
- Better resource utilization by achieving higher fuel burn-up than with conventional uranium reactors using solid fuel. Transuranic elements could in principle remain in the fluid fuel of the core, be destroyed in the neutron flux, either by direct fissioning or transmutation to a fissile element until eventually they all undergo fission,
- The avoidance of the expense of transport and fabrication of new fuel elements.

MSRs have some unique characteristics offering a potentially safer, more efficient and sustainable form of nuclear power associated with an on-line fuel processing. MSR does not operate under high pressure, and is not cooled by water, making steam explosion

impossible. In case of reactor damage, the fuel salt can be drained into sub-critical storage tanks.

From the 1940s up to now, many MSR concepts have been proposed all over the world using different fuel (U, Pu, Th...) and salt compositions (chlorides, fluorides...). These research projects have tried to find the optimum solution for the fuel salt composition suitable at the same time for:

- Neutronic properties (neutron moderation, breeding ratio, fissile inventory...),
- Operating temperature (melting temperature, radiation stability, transport properties),
- Actinide and fission products solubility in the considered molten salt to guarantee a homogeneous core composition,
- Materials compatibility and salt chemistry control,
- Processing and low waste options.

2. Historical experience

Extensive research into molten salt reactors started with the U.S. aircraft reactor experiment (ARE) in support of the U.S. Aircraft Nuclear Propulsion program (Bettis et al., 1957). The ARE (Fig. 1), an experimental high-temperature molten salt reactor was developed at Oak Ridge National Laboratory (ORNL). In 1954, the ARE was operated successfully for 100 h at steady-state outlet temperatures ranging up to 860 °C and at powers up to 2.5 MWt. The ARE used molten fluoride salt NaF–ZrF₄–UF₄ (53–41–6 mol%) as fuel, it was moderated by beryllium oxide (BeO) and used liquid sodium as a secondary coolant. This experiment used Inconel 600 alloy (nickel based alloy) for the metal structures and piping. The ARE showed that the fissile element UF₄ was chemically stable in the solvent and that gaseous fission products were removed essentially automatically by the pumping action of the salt circulation pump, accumulating in the pump bowl above the reactor. The fluid fuel had a very strong negative temperature coefficient, and the reactor could easily be started and stopped by changing the power demand without control rods.

The ORNL took the lead in researching the MSR through the 1960s, to investigate power reactor designs to be either simple converter reactors or breeder reactors on the Th–²³³U cycle (Rosenthal et al., 1970). Several designs were proposed using one or two fluid graphite moderated cores. In the two fluid design, a fuel salt carries the fissile ²³³U and a separate blanket salt for the fertile Th is used. The thorium blanket effectively captures the neutrons that leak from the core region leading to a high efficiency of neutron use (neutron economy), and a higher breeding ratio. ²³³U produced in the blanket, is transferred to the fuel salt by a fluorination process. Fluorination is accomplished by bubbling fluorine gas through the blanket salt, converting UF₄ to volatile UF₆. Gaseous UF₆ is converted back to UF₄ before being injected into the fuel salt. This chemical process is still considered nowadays as a powerful chemical step in the liquid fueled MSRs processing flowsheet. The main advantages of a two fluid system are a reduced initial fissile inventory and a simplification of fuel salt clean up from both protactinium (Pa removal is not necessary as it is diluted in the blanket salt) and soluble rare earth fission products from the fuel salt in absence of thorium (LeBlanc, 2010). Because thorium and rare earth fission products have similar chemical behavior, (Zagnit'ko and Ignatiev, 2012) the removal of rare earth fission products is greatly simplified without thorium in the core fluid. Rare earth fission products absorb neutrons and reduce the production of new fissile fuel, their concentration in the salt needs thus to be kept low, as they have a large cross section for neutron capture in a thermal neutron spectrum. The two fluid design offers absolute and incontrovertible advantages but rises however the

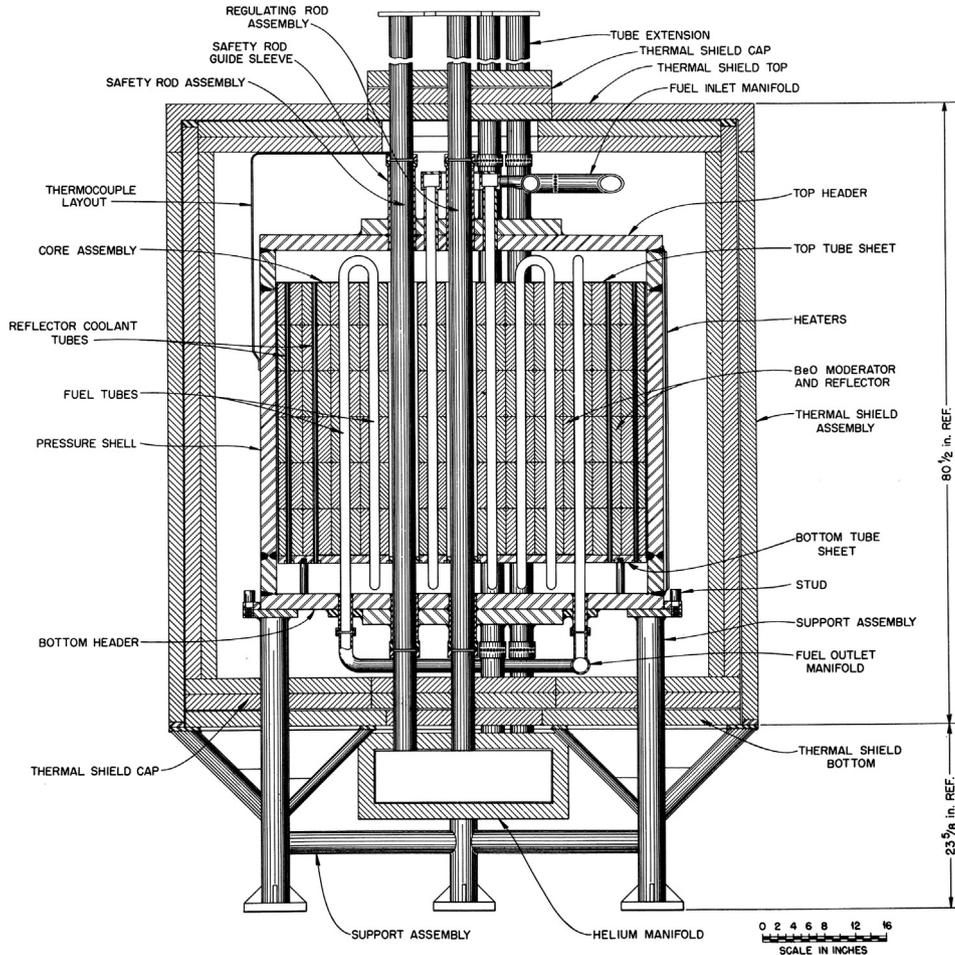


Fig. 1. ARE diagram (Bettis et al., 1957).

problem of the barrier wall (either nickel based alloy or graphite) between the core and the blanket region, that implies a more complex vessel exposed to fuel redox chemistry and radiation damages.

Vessel complexity and limited graphite lifetime (4 years) in contact with the molten fuel salt, oriented ORNL towards the construction of a simpler single-fluid reactor design: the Molten-Salt Reactor Experiment (MSRE) (MacPherson, 1985). This 8 MWth test reactor (Fig. 2) went critical in 1965 and operated with great success in a thermal neutron spectrum for 4.5 years until its shut down in December 1969. The fuel salt for the MSRE was $\text{LiF}-\text{BeF}_2-\text{ZrF}_4-\text{UF}_4$ (65–29–5–1 mol.%), moderated by pyrolytic graphite, its secondary coolant was molten $2\text{LiF}-\text{BeF}_2$ salt mixture. The MSRE operated with three different fissile fuels: ^{233}U , ^{235}U , and ^{239}Pu but the fuel salt did not contain thorium. During its operation, uranium was completely removed from the salt through fluorination by bubbling gaseous fluorine through the salt.

All metallic parts of the system in contact with the salt were made from the nickel-based alloy INOR-8 (later called Hastelloy-N), which was specially developed to overcome corrosion problems encountered in the ARE program with Inconel 600. This low chromium, nickel–molybdenum alloy (Hastelloy-N or Alloy N), was proved perfectly compatible with the fluoride secondary salts coolant. Hastelloy-N exposed to the fuel salt was however subject to inter-granular cracking and irradiation damage caused by (n, alpha) reactions in nickel and boron contaminants. The cause of the embrittlement was identified to be due to tellurium fission product

generated in the fuel salt. In subsequent years of the MSR program in ORNL, these issues were largely addressed by modifying the alloy make up of the Hastelloy-N (Engel et al., 1979). The control of the redox potential of the molten salt accomplished by occasional

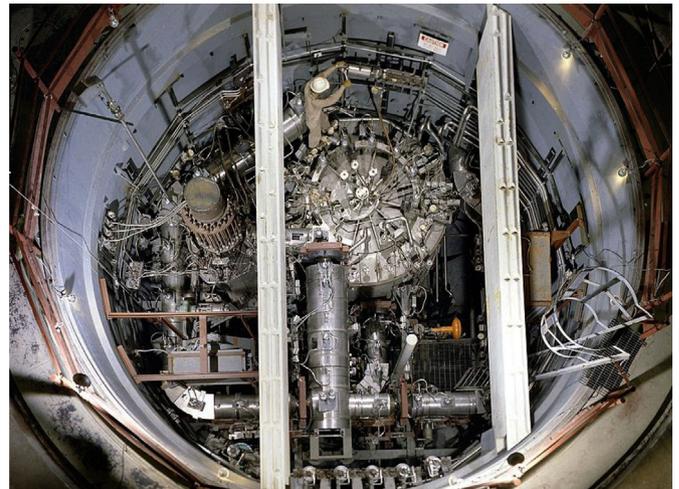


Fig. 2. MSRE.

additions of metallic beryllium was also found to be a powerful method to limit the corrosion rate for container material.

The MSRE equipment operated reliably and the radioactive liquids and gases were contained safely. The favorable experience gained from the 8 MWt MSRE test reactor led to the design of a reference breeder design with a thermal spectrum and thorium fuel cycle, the 1000 MWe Molten Salt Breeder Reactor (MSBR). The proposed MSBR design which would use LiF–BeF₂–ThF₄–UF₄ (72–16–12–0.4 mol.%) as fuel salt and NaF–NaBF₄ (8–92 mol.%) as the secondary coolant. An improved but complex processing flowsheet was intensively developed to achieve the difficult separation of thorium from rare earth fission products required for a single fluid design (McNeese, February 1971).

Despite the success of the MSRE reactor (Haubenreich and Engel, February 1970), the large research and development effort including natural and forced convection molten salt loop tests demonstrating many aspects of the reactor technology, the MSR program in ORNL was closed down in the early 1970s as the US consolidated its breeder reactor research to the liquid metal fast-breeder reactor (LMFBR) in which significantly more resources had already been invested and which were viewed as already well-advanced in 1972. The principal technical argument provided for the down selection was the larger amount of remaining development for MSRs and their competition for technical talent and facilities with LMFBRs (U.S. Atomic Energy Commission Division of Reactor Technology, Sep. 1972).

Oak Ridge did continue a modest program until the early 1980s on design and materials development, fuel chemistry and processing. They also proposed another MSR design, the Denatured Molten Salt Reactor (DMSR), developed to address proliferation concerns of the 1970s. In DMSR, continuous fuel processing was not performed (other than Kr, Xe gas removal), enriched Uranium (19.75%) was used for startup and as feed material as required to maintain criticality and ²³⁸U was added as needed to maintain denatured state. The simplified DMSR converter design with a low power density core allowing a full 30 year lifetime for graphite was called the “30 Year Once Through Design” (Engel et al., July 1980).

3. Application for MSR

The original goal of the Li, Be, Th, U/F MSR was to produce efficiently electricity in a breeder (MSBR) or converter modes (MSCR, DMSR), respectively with the fuel continuous processing or with it using only the simple helium sparging technique in the fuel circuit. Current goals include also a variety of different fuel cycle missions with or without Th–U support including burning of long lived transuranic actinides from spent fuel of current reactor fleet while producing the energy. The reactor physics of MSRs allows a flexible system, offering the potential to be operated both as a burner and a breeder, depending on the objective, and optimum resources utilization. The transmutation of TRU in MSR associated with an on-site fuel salt processing is efficient and reduces the amount of waste produced (Ignatiev et al., 2007).

MSRs are envisioned for electricity generation but, as solid and liquid fueled MSR are running at significantly higher temperatures than conventional light water-cooled nuclear reactors, they are also suitable as industrial heat sources in industries such as cement, steel, oil and chemicals. Such “Non-electric” applications could concern coal gasification, conversion of coal to olefin or diesel.

4. Structure of GIF system MSR R&D and benefit from collaboration within GIF

The decision for setting up a Provisional System Steering Committee (PSSC) for the MSR with Euratom, France, the Russian

Federation and the United States was taken by the GIF Policy Group in May 2004. In 2009 discussions were held on the mode of cooperation on MSR R&D within GIF. The Policy Group took the decision to set up a Memorandum Of Understanding (MoU) for both the MSR and LFR (Lead-Cooled Fast Reactor) systems. This MoU would provide a more flexible structure for R&D cooperation on those systems in the GIF framework for the mid-term. The MoU has been signed by CEA, on behalf of France and the JRC on behalf of Euratom in October 2010 and by Rosatom on behalf of the Russian Federation in November 2013. USA, China, Japan and South Korea are observers within the MSR-PSSC.

In the beginning MSRs were mainly considered as graphite-moderated reactors with a thermal neutron spectrum. Since 2005 liquid fueled MSR R&D has focused on fast-spectrum MSR options combining the generic assets of fast neutron reactors (extended resource utilization, waste minimization) with those related to molten salt fluorides both as fluid fuel and coolant (low pressure and high boiling temperature, optical transparency). Recent MSR developments in Russian Federation on the 1000 MWe Molten Salt Actinide Recycler & Transmuter (MOSART) and in France on the 1400MWe thorium Molten Salt Fast Reactor (MSFR) address the concept of large power units with an intermediate/fast neutron spectrum in the core (Rubiolo et al, January 2013; Ignatiev et al, 2014; Heuer et al., 2014). The fast neutron spectrum molten salt reactors open promising possibilities to exploit the ²³²Th–²³³U cycle and can also contribute, in the transmuter mode, to significantly diminishing the radiotoxic inventory from current-reactor used fuel in particular by lowering the masses of transuranic elements (TRU).

The United States, which participates in the SSC as an observer mainly works on FHRs as a nearer term reactor class whose technology developments are supportive of MSFRs. The Chinese Academy of Science through its Shanghai Institute of Applied Physics (SINAP) is actively pursuing since 2011 both liquid fueled MSRs (Thorium Molten Salt Reactor: TMSR) and FHR developments.

Although the different MSRs concepts interests are focused on different baseline concepts (MSFR, MOSART, TMSR and FHR), large commonalities in basic R&D areas exist and the Generation IV framework is useful to optimize the R&D effort. The following topics have been identified to overcome the remaining technological challenges:

- Materials and Components
- Liquid salt chemistry and properties
- Fuel and fuel cycle
- System design and operation
- Safety and safety system for innovative liquid fuels
- System integration and assessment

5. Status of new national projects

- Europe

The new interest in MSRs is a consequence of changing goals and changing technologies. There is a renewed interest in breeder reactors and reactors for burning of TRU's from spent fuel. For the latter option, MSRs have the unique advantage that there is no fuel fabrication step — an extremely complex task for fuel elements with higher actinides. Several national or international MSRs concepts were conceived and new research programs were launched. The performances and design parameters of MSRs concepts have been extensively revisited in Europe in the EURATOM 5th, 6th and 7th Framework Programs (MOST, ALISIA and EVOL projects). These EURATOM projects were carried out keeping a tight link with the

complementary International Science & Technology Centre projects (ISTC#1606 and #3749) in Russian Federation on MSRs including fuel salt properties, materials compatibility and liquid salts chemistry issues. From 2011 to 2013 these efforts were also supported by Rosatom in the frame of the MARS (Minor Actinides Recycling in Salts) project.

The state-of-the-art review of MSRs technology performed in the MOST project basically confirmed the potential of MSRs as breeders or burners and re-evaluated the issues to be further addressed by R&D (Vergnes and Lecarpentier, 2002; Renault and Delpech, 2005). The results have emphasized the potential of fast or “non-moderated” (graphite free), concepts for both breeding and burning (Mathieu et al., 2006; Mathieu et al., 2009). Fast-spectrum MSRs concepts, both breeders or burners, have large negative temperature and void reactivity coefficients, a unique safety characteristic not found in solid-fuel fast reactors. Traditional thermal-spectrum MSRs require relatively rapid processing of the molten salt if they are to be breeder reactors. The fast-spectrum MSRs have a higher breeding ratio that enables the reactor to be a breeder reactor with longer cycle times to remove soluble fission products. In the MSBR design, a 4 m³/day reprocessing rate was mandatory to insure a sufficient breeding ratio while the processing rate can be reduced down to 40 L/day in the MSFR concept (Delpech et al., 2009). Pyrochemical salt processing is mandatory to avoid the accumulation in the fuel salt of large quantities of lanthanides and zirconium that could be detrimental to several properties such as actinide trifluorides solubility.

These studies led to the design of two fast spectrum concepts: MOSART and MSFR (Fig. 3). The first concept aims to be used as an efficient burner of TRU waste from spent LWR fuel with MA/TRU ratio up to 0.45 without any uranium and thorium support (Ignatiev et al., 2012). The second concept has a good breeding capability when using the thorium fuel cycle but high power densities would be required to avoid excessive fissile inventories (300 MW/m³).

A promising single fluid configuration for the 2400 MWt MOSART is the homogeneous cylindrical core (3.6 m high and 3.4 m in diameter) with 0.2 m graphite reflector filled with 100% of LiF–BeF₂ salt mixture. It is feasible to design a critical homogeneous core fueled only by TRU trifluorides from UOX or MOX LWR used fuel while equilibrium concentration for trifluorides of actinides (0.4 mol% for Li, Be/F core, with the rare earth removal cycle 300

EFPD) is truly below solubility limit (~2 mol%) at minimal fuel salt temperature in primary circuit 600–620 °C. Maximum temperature in the fuel circuit does not exceed 720 °C.

Recently (Ignatiev et al., 2012), the flexibility of single fluid MOSART concept fuel cycle is underlined, particularly, possibility of its operation in self-sustainable mode using different loadings and make up. Single fluid 2400 MWt Li, Be/F MOSART core containing initial loading 2 mol% of ThF₄ and 1.2 mol % of TRUF₃, with the LnF₃ removal cycle of 300 EFPD after 12 years of slow increase of Th content in the solvent can operate without any TRUF₃ make-up, relying only on Th support as a self-sustainable system. The maximum concentration of TRU during this transition does not exceed 1.7 mol %. At equilibrium the molar fraction of fertile material in the fuel salt is near 6% and it is enough to support the system with CR = 1 within 50 yrs reactor lifetime.

The reference MSFR is a 3000 MWt reactor with a total fuel salt volume of 18 m³ (Mathieu et al., 2009), operated at a maximum fuel salt temperature of 750 °C. The fuel salt is composed of lithium fluoride and thorium fluoride and the proportion of heavy nuclei is fixed at 22.5 mol%. The preliminary design of the primary circuit of the MSFR is a single compact cylinder (2.25 m high × 2.25 m diameter) where the nuclear reactions occur within the liquid fluoride fuel salt acting also as the coolant. The fuel salt flows in the central part of the core freely from the bottom to the top without any solid moderator. The return path of the salt is divided into 16 sets of pumps and heat exchangers located around the core. Bubbles are injected in the fuel salt circulation after the exchangers and separated from the liquid at the core outlet. The fuel salt runs through the total cycle in 3–4 s. The total fuel salt volume is distributed half in the core and half in the external fuel circuit (salt collectors, salt-bubble separators, fuel heat exchangers, pumps, salt injectors and pipes). The lower neutronic reflector is connected to a drain system enabling the reactor core to be drained for planned shut downs or in case of incidents that lead to a temperature increase in the core. Thus the entire fuel inventory can be passively drained by gravity into subcritical, passively cooled tanks. MSFR configurations corresponding to various starting modes of the reactor are all characterized by excellent safety coefficients and have the same very good deployment capabilities.

The European partners have carried out theoretical and experimental studies to verify the feasibility of the MOSART and MSFR systems to reduce long-lived waste toxicity and produce electricity

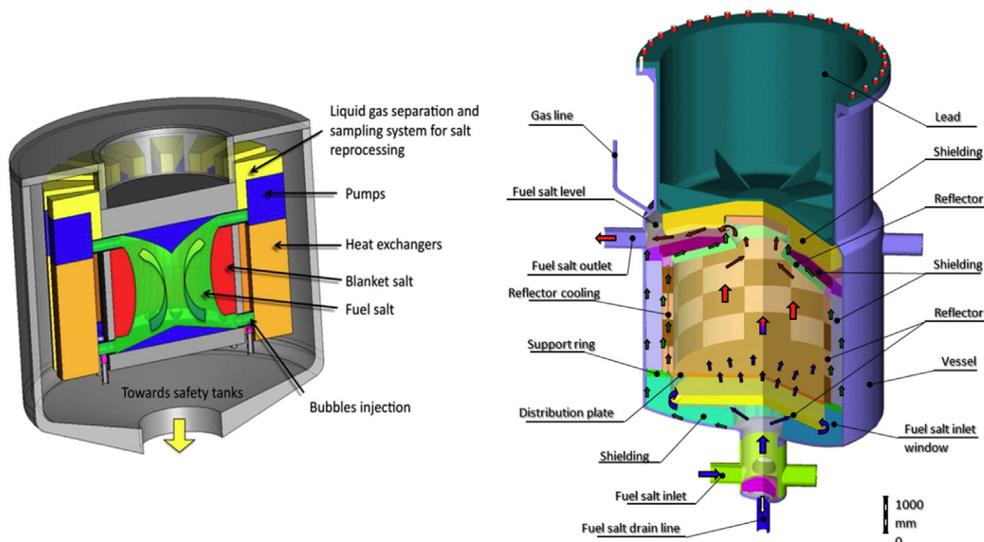


Fig. 3. MSFR and MOSART concepts.

simultaneously. The common objective of the MOSART and MSFR projects is to develop a conceptual design for intermediate/fast-spectrum MSRs with an effective system configuration – resulting from physical, chemical and material studies – for the reactor core, the reprocessing unit and waste conditioning (Merle-Lucotte et al., 2013). The conceptual design activities are intended to increase the confidence that fast-spectrum MSR 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).

Partially distinct and independent program, which is focused on the thermal spectrum MSR system with LiF–BeF₂ carrier salt, has been conducted in the Czech Republic from 2004 under the national project SPHINX. The effort has been focused mainly to the development and verification of selected fuel cycle technologies, development of structural material for the fluoride molten salt media and to theoretical and experimental studies of MSR physics (Uhlír and Juříček, 2013).

Existing development of the thorium – uranium fuel cycle technology covers both the verification of MSR liquid fuel processing technology (LiF–BeF₂–UF₄ and LiF–BeF₂–ThF₄) and the laboratory investigation of on-line reprocessing techniques. Here the effort has been focused mainly on the investigation of electrochemical actinide and fission products separation from LiF–BeF₂, LiF–NaF–KF and LiF–CaF₂ carriers (Uhlír et al., 2012).

The SPHINX project supported also the development of structural material for MSR technology. New nickel alloy called MONICR (Ni–Mo–Cr type alloy) was developed by SKODA Nuclear Machinery in the cooperation with COMTES FHT. Experimental production of tubes and sheets from MONICR was realized in 2008 and basic corrosion and irradiation tests were done in 2010–2011. Further development of MONICR alloy is realized by COMTES FHT.

The investigation of the MSR physics was focused on the experimental measurements of fluoride salts neutronics in the LR-0 and LVR-15 reactors of the Nuclear Research Institute Řež (UJV) and the Research Centre Řež (Fig. 4). Basic principles of the method of measurement were successfully verified and selected neutronic data of LiF–NaF, LiF–BeF₂, LiF–NaF–UF₄, LiF–NaF–ThF₄, LiF–BeF₂–UF₄ and LiF–BeF₂–ThF₄ salt mixtures were obtained by irradiation of instrumented BLANKA probes inserted in the central part of the LR-0 reactor core where the standard VVER fuel

assemblies serve as the neutron driver. Initial measurements were negatively affected by the use of natural lithium in LiF. The bilateral agreement, concluded between the US – Department of Energy and the Ministry of Industry and Trade of the Czech Republic in 2012, enabled the US to provide 75 kg of FLIBE coolant salt highly enriched in ⁷Li to the Czech Republic. The salt shipped from Oak Ridge National Laboratory in May 2013 will be used in Research Centre Řež for ongoing joint investigation of FHR and MSR neutronics.

A parallel program on the thermal spectrum MSR ran at TU-Delft in which the graphite-salt lattice was optimized with regard to temperature feedback. Self-breeding can be achieved in a range of lattices which provide sufficient negative temperature feedback. The graphite life span is 10–20 years with a power density between 5 and 10 MW/m³. Transient calculations demonstrated the inherent safety characteristics of the reactor design (Nagy, 2012).

- US

Solid fueled FHRs (Fig. 5) are a nearer term molten salt reactor option. FHRs by definition feature low-pressure liquid fluoride salt cooling, coated particle fuel, a high-temperature power cycle, and fully passive decay heat rejection. FHRs have the potential to economically and reliably produce large quantities of electricity and high-temperature process heat while maintaining full passive safety.

Leveraging the inherent reactor class characteristics avoids the need for expensive, redundant safety structures and systems and is central to making the economic case for FHRs. Additionally, as a HTR class, FHRs can efficiently generate electricity and provide the energy for high-temperature industrial processes (notably including the production of hydrocarbon fuel). Moreover, high-temperature operation increases FHR compatibility with dry cooling.

The US is broadly pursuing advanced technologies to improve the affordability of nuclear power. Several of the crosscutting technology developments support FHRs. A key continuing task is the irradiation testing of coated particle fuel, which supports both gas- and salt-cooled reactors (Baldwin et al., 2012). Also advanced ceramic matrix composite development and testing continues in support of light water reactors, advanced reactors, and fusion energy systems (Katoh et al., 2013). Activities also continue within the ASME on introducing ceramic composites into the Boiler and Pressure Vessel Code for high temperature reactor use. The US has also recently initiated development of a licensing framework for advanced nuclear reactor technologies (Letter from Glennra, July 9, 2013). The American Nuclear Society has also begun development of an FHR design-safety standard (ANS 20.1). The US is also

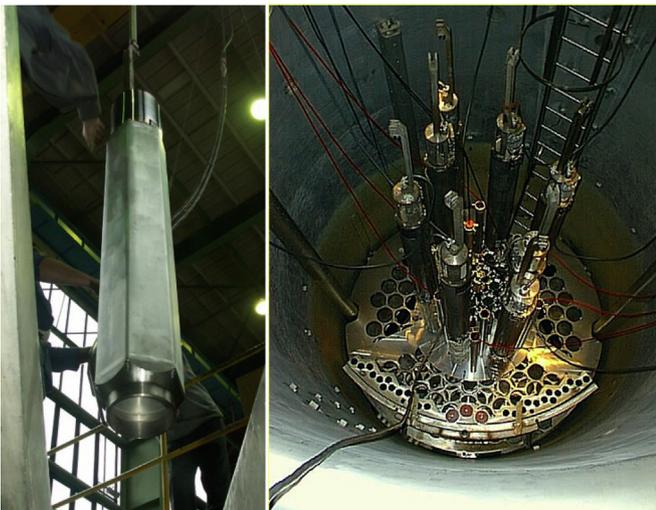


Fig. 4. BLANKA probe in LR-0 reactor for measurement of salt neutronics.

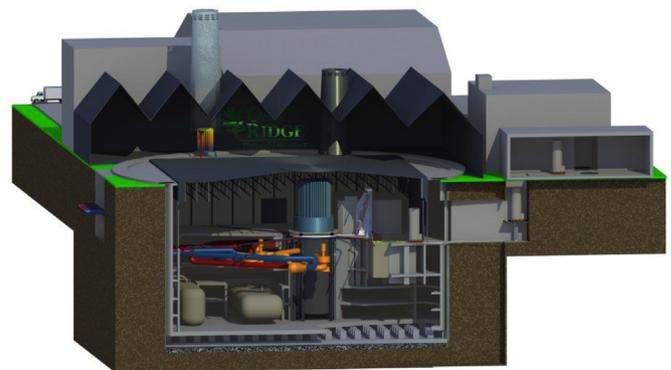


Fig. 5. Cut away view of a preconceptual design for a central station type FHR.

pursuing development of a high temperature fission chamber that would be useful for FHR start-up monitoring and is investigating in-vessel optical monitoring technology, which is especially useful for reactors with single-phase transparent coolants (Anheier et al, August 2013).

Directly supporting FHRs, the US has recently completed a forced convection liquid salt test loop (Yoder et al., 2014). US researchers have also recently completed both a preliminary mechanical, structural, and neutronic evaluation of a central station type FHR (Varma et al., September 2012) and have created an initial technology development roadmap for FHRs (Holcomb et al., September 2013). The US is also supporting FHR development through university research. A team from the Massachusetts Institute of Technology, the University of California at Berkeley, and the University of Wisconsin is, currently performing an integrated research project entitled “High-Temperature Salt-Cooled Reactor For Power and Process Heat”. The Georgia Institute of Technology and the University of Tennessee are teaming up to investigate FHR core design options, and the Ohio State University is developing a liquid salt test loop and FHR hydraulic models. Also, John’s Hopkins University is developing carbide coatings for nickel-based alloys to enable liquid fluoride valves.

• China

In January 2011, Chinese Academy of Science launched the “Thorium Molten Salt Reactor Nuclear Energy System” (TMSR, see Fig. 6) project aiming at developing solid and liquid fueled MSR which strive for realizing effective Thorium energy utilization and hydrogen production by nuclear energy within 20–30 years. In

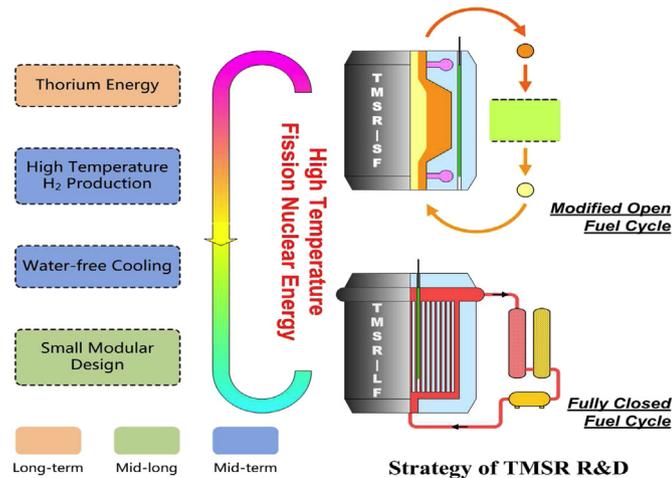


Fig. 6. Strategy of TMSR R&D.

- 1) FUJI-U3 (self-sustaining by ²³³U as fissile)
- 2) FUJI-Pu (using Pu as initial fissile)
- 3) FUJI for transmutation of Minor Actinides
- 4) Super-FUJI (1000MWe large sized plant)
- 5) Mini-FUJI (pilot plant)

August 2013, TMSR project became one of the candidates of the national energy major application-technology research & demonstration projects of the Chinese National Energy Administration. The near-term goals of TMSR Project till 2017 are to build two test reactors: a solid fueled one (TMSR-SF, FHR type) and a liquid fueled (TMSR-LF, MSR type). The project includes also the development of a Pyro-Process Complex.

• Japan

Japan has a 30 years history on MSR study after ORNL has stopped its major activity in 1970s. Furukawa and his group proposed a “Thorium Molten Salt Nuclear Energy Synergetic System (THORIMS-NES)” in 1980s. THORIMS-NES is a combination of fission power reactor of Molten Salt Reactor (MSR-FUJI), and Accelerator Molten Salt Breeder (AMSB) for production of fissile ²³³U (Furukawa et al., 2008). Besides the design study, several molten salt loops were manufactured by his group.

Conceptual design of FUJI has started in late 1980s based on the ORNL studies. FUJI is size-flexible, but typical value is 100–200 MWe. A reactor vessel and pipes are made of Hastelloy N. There is a graphite moderator within a reactor vessel, but the graphite moderator does not require replacement during a reactors’ lifetime of 30 years. Fuel salt is based on FLiBe with U/Pu/Th fluoride. Continuous chemical processing of fuel salt is not performed in FUJI except removal of gaseous fission products. Several designs were studied for various applications of FUJI as follows (Yoshioka et al., 2013) (Fig. 7).

After the severe accidents of the Fukushima plants on March 11 2011, a plant safety issue and a fuel cycle issue became strongly recognized.

As for the first issue on safety, MSR-FUJI has very high safety, and severe accidents are essentially impossible. Some of major accidents were analyzed so far, and safety criteria and guidelines for accident analysis were proposed (Varma et al., September 2012). Also, it was confirmed that MSR is still safe against Fukushima type accidents (Varma et al., September 2012).

As for the second issue of the back-end problem, using Pu in FBR is stagnant and consumption of Pu is in public discussion. MSR is essentially appropriate as a Pu-burner. Besides the above FUJI study, FLiNaK based MSR was investigated, which was inspired by Russian proposal (Mitachi, 2013). Another recent proposal is a liquid fuel module, called Reactor in Reactor (RinR). RinR is to place fuel assemblies of LWRs in order to provide burner device of Pu/MA. A preliminary design was proposed (International Symposium, 2013).

As for the academic activities of the Atomic Energy Society of Japan, two specialists committees started in 2013; one is “Application of Molten Salt Technology to Nuclear Engineering”, and another is “Thorium Fuel Working Group” which studies thorium for solid and liquid fuel. In University of Tokyo and Fukui University,

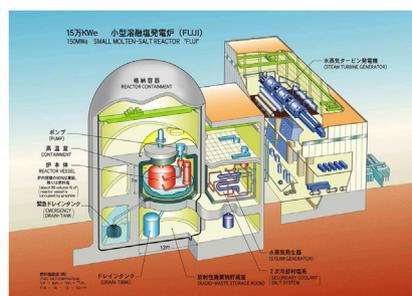


Fig. 7. Bird's-eye view of MSR-FUJI.

a study on accident performance of fluoride salt fuel started in 2013 by the support of an electric power company (Varma et al., September 2012).

The International Thorium Molten-Salt Forum is promoting MSR and related Th cycle since 2008 by researchers/engineers/citizens among 13 countries (<http://msr21.fc2web.com/e>).

6. Open technical issues and future R&D priorities

Specific scientific and technological challenges remain for the development of liquid fueled MSRs and the safety approach has yet to be established (Brovchenko, 2013). Liquid fueled MSR viability assessment should address essential R&D issues in particular in the following areas:

6.1. Salts and materials

A large body of literature exists on the corrosion of metallic alloys by molten fluorides salt. Much of this work was done in the past at ORNL and involved both thermal and forced convection loops. The major impurities that must be removed to prevent severe corrosion of the container metal are moisture/oxide contaminants. During the US MSR program, considerable effort was devoted to salt purification by HF/H₂ sparging of the molten salt, which is described in numerous ORNL reports. A great deal of effort has still to be devoted to develop analytical and purification methods able to:

- Identify the oxygen-containing species (oxide type, hydroxyl...),
- Purify the molten-salt mixture,
- Determine accurately the oxygen content in salt melts.

The structural materials retained for the MSR container were special Ni–Mo alloys with a low concentration of Cr. The composition of the alloy was optimized by ORNL researchers for corrosion resistance (both in a low oxygen gas atmosphere and in molten fluorides), irradiation resistance and high temperature mechanical properties. Less severe corrosion attack (<10 μm/year) was seen for the Hastelloy N in contact with the MSRE fuel LiF–BeF₂–ZrF₄–UF₄ salt at temperatures up to 704 °C for 3 years (26,000 h). The most surprising observation was the almost complete absence of corrosion for Hastelloy N during the 3-year period to the MSRE coolant LiF–BeF₂ salt (Ignatiev and Surenkov, 2012).

As a nearer term option, the key materials development challenges for solid fuel Li, Be/F cooled FHRs focus on demonstrating adequate performance for licensing rather than establishing fundamental materials performance. A substantial body of knowledge exists for Alloy N, the leading candidate material for FHR test reactors. Alloy N, however, is not currently approved as a material for high-temperature NPPs. Thus, a materials qualification effort would be required for use of Alloy N in any part of the containment boundary or to perform any other safety function. Developing a safety case for limited term Alloy N use at temperatures less than 704 °C may be possible based upon existing data from the earlier MSR program and/or limited term supplemental qualification testing. The maximum allowable stress for Alloy N decreases rapidly above 600 °C, becoming too low for practical use above 700 °C. Compared to the leading candidate materials that are usually considered for high-temperature nuclear reactor construction, Alloy N has significantly lower high-temperature strength. However, developing and qualifying improved performance alloys for longer term high stress applications will require substantial investment over a number of years. ORNL has recently begun the development of successor alloys to Alloy N. Samples of the new alloys show promise for application in high stress applications at higher temperatures

along with good fluoride salt corrosion resistance. As a first step, early phase, long-term advanced salt-compatible alloy property measurements will need to be performed. Thus, improved alloys can be developed and made available for commercial FHR deployments while a limited-term safety case for use of Alloy N at test reactors can be developed in parallel.

Material integrity is directly linked to the instrumentation and control of liquid salt because the corrosion of the structural materials by the molten salt strongly depends on the redox potential of the salt. It has been demonstrated that the salt redox potential is a key parameter in the corrosion phenomena of structural materials of MSRs. The chemical corrosion can be controlled by a redox buffer which controls the potential of the fuel salt. The redox buffer considered for MSR is the redox system U(IV)/U(III). This potential has to be measured on line in the reactor core because the potential increases with operation time due to the fission reactions (Delpech et al., 2010). Addition of a reducing agent leads to a decrease of the fuel salt potential. For FHR the redox buffer couple is not selected yet.

The adoption of fast spectrum MSR designs and the possible application of MSR as industrial heat sources have introduced new challenges such as higher salt operating temperatures (up to 750 °C), metallic barriers and reflectors able to function in a strong neutron flux. The compatibility of salts with structural materials for fuel and coolant circuits, as well as fuel processing material development needs thus to be addressed.

Special attention will have to be paid to the measure and control of the U(IV)/U(III) ratio in order to reach the desired corrosion resistance of Ni based alloys. The instrumentation and control of liquid salt must be developed with in-situ measurements at a lab-scale in order to:

- Measure and control the redox potential of the salt and to limit the corrosion,
- Measure the composition of the salt containing fissile and fertile elements.

New materials are developed and tested in France (Ni–W–Cr alloys, EM-721) and the Russian Federation (Ni–Mo–Cr alloys) (Brovchenko, 2013; Ignatiev and Surenkov, 2013). In the molten Li, Be, U/F and Li, Be, Th, U/F salts with [U(IV)]/[U(III)] ratio ≤ 100 up to temperatures 750 °C the HN80MTY alloy with 1% added aluminum is the most resistant to tellurium intergranular cracking of Ni-base alloys under study (including Hastelloy-N, MONICR and EM-721 alloys). The metallurgy and in-service properties of these alloys need to be investigated in further details regarding irradiation resistance and industrialization.

6.2. The next steps needed to develop these alloys must involve

- Irradiation, corrosion, tellurium exposure, mechanical property and fabrication tests to finalize the composition for scale up,
- Procurement of large commercial heats of the reference alloy;
- Mechanical property and corrosion tests of at least 10,000 h duration,
- Development of design methods and rules needed to design a reactor (breeder or burner) to be built of the modified alloy.

Both FHRs and MSRs will make extensive use of continuous fiber composites (CFCs) for reactor vessel internal components. Some of these components will be large and have complex geometry (e.g., the lower core support plate in FHR or fuel salt distribution plate in MOSART). CFCs are being evaluated for in-vessel structural applications in other reactor classes (e.g., using silicon carbide–silicon carbide CFCs as channel boxes at boiling water reactors to minimize

the core zirconium content); however, a significantly larger role for CFCs is envisioned at FHRs due to their ability to maintain their structural characteristics at high-temperatures.

Tritium control is an important issue for both MSR and FHRs because tritium is the only radionuclide that under normal operating conditions, without failed fuel, has the potential for significant release. Tritium is generated primarily by the interaction of neutrons with lithium and beryllium in the primary coolant. FHRs and MSRs will produce significantly more tritium LWRs, but less than heavy-water-moderated reactors. However, tritium control for both designs is complicated because at high temperatures tritium permeates available structural alloys. The large contact surface area and thin walls of heat exchanger tubes means that heat exchangers will be the primary release pathway. Tritium trapping in the secondary NaF–NaBF₄ coolant was proposed and experimentally proved in molten salt loop by ORNL for the MSBR design. The primary tritium release prevention mechanism planned for first-generation FHRs is chemical capture to form yttrium tritide within double-walled heat exchangers.

6.3. Fuel and fuel cycle

Experimental investigations of physico-chemical properties of actinide fluorides containing salts are carried out to elucidate the influence of the salt composition on thermal and physical properties of the MSR fuel (melting temperature, solubility of actinides and vapor pressure). Experimental results, received within ISTC#3749 and Rosatom MARS projects, on key physical & chemical properties of MOSART and MSFR fuel salts which determine main core parameters (e.g., inlet/outlet temperatures, critical loadings and temperature reactivity coefficient of the fuel salt, etc) are described in details elsewhere. An extensive thermodynamic database of various fluoride systems has been developed in the Institute for Transuranium Elements (EC/JRC/ITU, Germany) since 2002 through the assessment of phase diagrams by both modeling and experimental studies (Beneš and Konings, 2012, 2009). This effort culminated in the full thermodynamic description of the MSFR system which was used to optimize the initial fuel composition, done within the frame of the EVOL project. As part of the general understanding of the fluoride salt behavior, a systematic investigation of the heat capacity of the key systems coupled with phase diagram investigations are performed at ITU.

These data are now used in multi-physics numerical codes (neutronics and thermo-hydraulics) for the simulations of the fuel salt heat and flow distribution in the reactor cavity (reactor reactivity, fuel irradiation, reactivity feedback coefficients, reactor core wall temperature...).

The different steps of the reprocessing process (except fluorination for which a large experimental feedback from US research work is found in the literature, (Briggs, 1966)) are currently tested in the Russian Federation and France laboratories. The work includes the measurement of actinide and fission product distribution coefficients to evaluate the efficiency of the reductive extraction process using two different liquid pool of bismuth containing lithium as developed by ORNL. The other steps of the reprocessing are also tested in French laboratories such as the back extraction of lanthanides in a chloride or/and fluoride molten salt and the precipitation of lanthanide oxides using bubbling of Ar–H₂O gaseous mixture (Laplace et al., 2011). The activity coefficients of lanthanides and actinides were estimated using several literature sources leading to the construction of a database available for LiF–ThF₄ molten salt (Delpech, 2013).

A molten salt loop (Forced Fluoride Flow for Experimental Research project, France) is developed to investigate helium sparging as a trapping method for non-soluble fission products

(noble metals) dispersed in the flowing liquid as well as gaseous fission products. Implementation of instrumentation (temperature, level and flow rate measurement) on the whole experimental set up is under progress.

The coupling of neutronic and reprocessing simulation codes in an integrated numerical tool is also used to calculate the extraction efficiencies of fission products, their location in the whole system (reactor and processing unit) and to predict radioprotection issues (Doligez et al., 2014; Fiorina et al., 2013; Aufiero et al., 2013a). Studies are however limited by the uncertainties on the design and knowledge of the complete chemical reprocessing scheme.

The thermodynamic properties of all phases considered in a multi-component system such as the fuel salt have to be assessed in order to collect new data which are necessary for developments of MSR designs, reprocessing scheme and simulation codes. Physical-chemical behavior of fuel salts and in particular coupling between neutronic, thermal-hydraulic and chemistry need to be thoroughly investigated.

Acquisition of fundamental data for the extraction processes is still needed especially for the actinide-rare earth fission products separation. The extraction of rare earth elements has to be done because of the low solubility of these trifluoride elements and their neutronic captures that decrease the reactivity balance. This critical step of the pyrochemical process have to be examined both from a thermodynamic and kinetic point of view. The different steps involved in the reprocessing of the liquid fuel have not yet been studied from the technological and engineering points of view and will have to be implemented as the system moves from the lab-scale to the industrial scale. Non-proliferation issues related to the fuel salt processing scheme should also be more deeply addressed.

The efficiency of the non-soluble fission products (noble metal and gases) extraction by helium bubbling, which is needed for circuit components life-time and thus safety, will have to be demonstrated. The start of the FFER loop running is foreseen for the beginning of year 2014. The future R&D studies will focus on the gas and particles extraction efficiencies (gas/salt separation, gas analysis by mass spectrometry) and the evaluation of the freezing valve used to drain rapidly the fuel salt from primary circuit to subcritical tanks in case of incidents.

FHRs will use coated particle fuel. DOE-NE is currently testing tristructural isotropic (TRISO) coated particle fuel as part of its HTGR development efforts. Identical TRISO particles are directly applicable to FHRs. The initial TRISO fuel loads for first-generation FHRs will cost substantially more than LWR fuel pellets. TRISO fuel is not currently manufactured at the commercial scale. Consequently, the cost savings resulting from manufacturing scale-up and automation cannot be reliably estimated at this time. FHRs, like LWRs, are thermal spectrum reactors intended to run on a once-through low-enrichment uranium fuel cycle. However, FHRs will require somewhat higher ²³⁵U enrichment than that currently employed at LWRs, which will incur substantial costs to modify the existing fuel infrastructure. The manufacturing fuel costs for FHR TRISO are expected to be similar to those for the increased enrichment for LWR pellets.

6.4. Design, operation & safety

Liquid fueled MSRs are flexible in operation but very different in the safety approach from solid fuel reactors (Aufiero et al., 2014; Brovchenko et al., 2013). In the new MSR concepts developed worldwide, safety will be an essential point to be considered all along the R&D studies. The first step of the safety approach is a systematic description of the reactor components, limited to the

main systems surrounding the core, in order to identify the typical accident initiators (Brovchenko et al., 2012). The development and the qualification of appropriate simulation tools to study normal and incidental situations have been initiated within the Euratom EVOL and the complementary Rosatom MARS projects (Guerrieri et al., 2012; Aufiero et al., 2013b). In order to assess the behavior of the fuel salt after reactor shut down, a tool based on point kinetics able to calculate the decay heat have been developed and validated for fast spectrum MSR (Fiorina et al., 2014). Loss of heat sink transients can be calculated allowing the evaluation of their impact on the fuel salt temperature. These residual heat calculations will be the basis for the design of the safety draining system, as drainage of the salt is foreseen for reactor shut down, whether in normal or in incidental conditions.

A complete preconceptual design incorporating all of the specialized SSCs of FHRs does not yet exist. However, initial candidate technologies have been identified for all required FHR functions. Substantial uncertainty remains in key elements of plant design and technology implementation. Developing a complete conceptual design will provide a unifying framework to enable an integrated set of technology choices to be made. Concept development also identifies technologies that require longer term development (e.g., structural alloy creep testing) that, in turn, assist in determining the overall timeline for development and deployment of the reactor class.

The licensing-related design and safety features of liquid fueled MSR combine those of a reactor and a chemical processing plant. The MSR safety approach will have to take into account a fuel in a liquid form within the coolant which depends upon keeping actinides and fission products in solution. This topic is directly linked to the previous one “Fuel and fuel cycle” where physico-chemical properties of salt are examined.

The impact of the stagnant heating fuel salt on the core and fuel loop systems will be further studied. Models improvement is primarily foreseen in the direction of increasing the reliability of the thermal-hydraulic modeling. The use of turbulence models more suitable for simulating recirculation regions should also be considered. Extension of the geometrical capability of the two models to generic 3D configurations would be of great advantage to better characterize local effects, to optimize the core design, as well as to allow the simulation of asymmetric transients like the coast-down of a single pump. A proper assessment would require a detailed hydrodynamic design of both the core and the out-of-core components (especially pumps and heat exchangers). The consequence of the in-core helium circulation on reactivity (void effect) should also be examined.

Several categories of active components are needed to be qualified for the operation of MSRs like heat exchangers, pumps, valves... These items face the same thermal and chemical compatibility challenges as the instrumentation equipment. Among these, the heat exchangers are likely to represent the most significant challenges. A heat exchanger is a cold spot in the system and supports a significant heat flux, both factors that can drive corrosion and corrosion product deposition. In addition, the heat exchangers may be required to provide a barrier for tritium diffusion to control tritium releases to the balance-of-plant and the environment. The designs must also allow inspection and be serviceable in the event of plugging or leaking. These challenges are compounded by a desire to maximize the thermal efficiency of the heat exchanger by minimizing its wall thickness and maximizing its surface area.

7. Conclusion

Molten salt fluorides as coolants offer interesting features such as chemical inertia, very good transport properties, strong

irradiation resistance, high thermal stability and boiling points. They share some advantages with liquid metal coolants like reactor operation at low pressure. This constitutes a significant safety and cost advantage.

The MSR has many advantages like the capability of burning actinides or breeding fissile fuel, no need for fuel manufacturing and a high level of sustainability because MSR can be run as a breeder reactor in the uranium fuel cycle or in the thorium fuel cycle or as a transmuter of long-lived actinides from spent nuclear fuel. Worldwide, all these operation modes of the MSR are investigated.

Molten salt cooled reactors can be operated with either a solid fuel or a liquid fuel and different MSR concepts are developed worldwide depending on the different goals and needs (sustainability, actinide burner...). The favorable aspect of reactors loaded with TRISO particle fuel and cooled with a molten salt (FHR) over a liquid fuel reactor is the robustness of the cladding of this fuel which adds an extra barrier to fission product release and increase the safety margins in case of incidents.

Introduction of molten salt technology to nuclear power can be done in two stages: (1) fluoride salt as coolant + solid HTR fuel; (2) replacement of the pebble bed solid fuel by graphite reflector/moderator in the core with subsequent fuel addition to the fluoride salt.

The Molten Salt Reactors (MSR) with liquid fuel can be designed with a neutron spectrum ranging from thermal to fast. R&D on molten salt reactors in Europe has been focused on fast spectrum concepts with or without thorium support (MSFR and MOSART) which have been recognized within GIF as long term alternative to solid-fueled fast neutron reactors with attractive features (strongly negative feedback coefficients, smaller fissile inventory, simplified fuel cycle). A negative temperature reactivity coefficient is universally recognized as a desirable safety feature for power reactors. The consideration done demonstrated the potential of the MSFR and MOSART as systems with flexible configurations and fuel cycle scenarios which can operate within technical limits with different loadings and make up based on TRUs (from spent LWR fuel with MA/TRU ratio up to 0.45) as special actinide transmuter, as self-sustainable system (CR = 1) or even as a breeder (CR > 1).

The risks of corrosion by the impurities (oxygen, water mainly) dissolved into a molten salt coolant or by the fission products present in fuel salt are a significant issue in MSRs that has been the subject of R&D work since the 1950s. The R&D on advanced materials for MSFR and FHR designs are under way now in different countries. Although more experimental and qualification efforts for special materials involved in the MSR design are still needed to demonstrate the viability of MSRs, past experience gained at the ORNL in the 1960s and 1970s remains a source of knowledge that demonstrated many aspects of the reactor technology.

The safety analysis methods in their current form cannot be applied to liquid fueled MSRs, in particular due to the fact that the fuel is molten during normal operation and due to the absence of cladding. Its outstanding safety level is ensured by the strongly negative feedback coefficients, even in a fast neutron spectrum, and the capability to drain the liquid fuel in dump tanks, which excludes overheating due to stagnant decay heat removal. A novel methodology for the design and safety evaluations of liquid fueled MSRs is needed, relying on current accepted safety principles such as the principle of the defense-in-depth, the use of multiple barriers and the three basic safety functions: reactivity control, decay heat removal and radioactive products confinement. New methodology applied to the reactor and reprocessing units shall be robust and comprehensive, integrate both deterministic and probabilistic approaches.

The various molten salt reactor projects like FHR, MOSART, MSFR, and TMSR have common themes in basic R&D areas, of

which the most prominent are the liquid salt technology and materials behavior, the fuel and fuel cycle chemistry and modeling, and the numerical simulation and safety design aspects of the reactor. Research in these areas will not only support the development of the MSR and FHR, but can find applications in other Generation IV systems as well, like the SFR, LFR, and VHTR, e.g., for the development of alternative fluids for intermediate heat transport, where liquid salts offer potential advantages regarding in particular the high volumetric heat capacity, reduced equipment size, absence of chemical exothermal reactions, intermediate loop and power cycle coolants.

The strong international cooperation between the countries involved, among which China, European Union, France, Japan, Russia, and the USA has shown to be very effective in tackling the most prominent remaining R&D issues. However, a continued effort is needed by all parties to converge to a basic reactor design excelling in safety and sustainability around 2020.

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