THE MOLTEN SALT REACTOR (MSR) IN GENERATION IV: OVERVIEW AND PERSPECTIVES

Boussier H.[1], Delpech S.[2], Ghetta V.[3], Heuer D.[3], Holcomb D.E.[4], Ignatiev V.[5], Merle-Lucotte E.[3], J. Serp[1]

(1) Hubert Boussier – Commissariat à l‟énergie atomique (hubert.boussier@cea.fr)
(1) Jérôme Serp– Commissariat à l‟énergie atomique (jerome.serp@cea.fr)
(2) Sylvie Delpech – Centre National de la Recherche Scientifique (delpech@ipno.in2p3.fr)
(3) Véronique Ghetta – Centre National de la Recherche Scientifique (Veronique.Ghetta@lpsc.in2p3.fr)
(3) Daniel Heuer– Centre National de la Recherche Scientifique (Daniel.Heuer@lpsc.in2p3.fr)
(3) Elsa Merle-Lucotte Centre National de la Recherche Scientifique (elsa.merle@lpsc.in2p3.fr)
(4) David-Eugene Holcomb – Oak Ridge National Laboratory (HolcombDE@ornl.gov)
(5) Victor Ignatiev – Russian Research Center, Kurchatov Institute (ignatiev@vver.kiae.ru)

Abstract

The MSR is distinguished by its core in which the fuel is dissolved in molten fluoride salt. The technology was first studied more than 50 years ago. Modern interest is on fast reactor concepts as a long term alternative to solid-fuelled fast neutrons reactors. The onsite fuel reprocessing unit using pyrochemistry allows breeding plutonium or uranium-233 from thorium. R&D progresses toward resolving feasibility issues and assessing safety and performance of the design concepts. Key feasibility issues focus on a dedicated safety approach and the development of salt redox potential measurement and control tools in order to limit corrosion rate of structural materials. Further work on the batch-wise online salt processing is required. Much work is needed on molten salt technology and related equipments.

I. INTRODUCTION

Molten Salt Reactor (MSR) technology was partly developed, including two demonstration reactors, in the 1950's and 1960's in USA (Oak Ridge National Laboratory). The demonstration MSRs were thermal-neutron-spectrum graphite-moderated concepts. Since 2005, R&D has focused on the development of fast-spectrum MSR concepts (MSFR) combining the generic assets of fast neutron reactors (extended resource utilization, waste minimization) with those relating to molten salt fluorides as fluid fuel and coolant (low pressure and high boiling temperature, optical transparency) [1-5].

In contrast to most other molten salt reactors previously studied, the MSFR does not include any solid moderator (usually graphite) in the core. This design choice is motivated by the study of parameters such as feedback coefficient, breeding ratio, graphite lifespan and ^{233}U initial inventory. MSFR exhibit large negative temperature and void reactivity coefficients, a unique safety characteristic not found in solid-fuel fast reactors.

Compared with solid-fuelled reactors, MSFR systems have lower fissile inventories, no radiation damage constraint on attainable fuel burn-up, no requirement to fabricate and handle solid fuel, and a homogeneous isotopic composition of fuel in the reactor. These and other characteristics give MSFRs potentially unique capabilities for actinide burning and extending fuel resources.

MSR developments in Russia [6,7] on the MOlten Salt Actinide Recycler and Transmuter (MOSART) aims to be used as efficient burners of transuranic (TRU) waste from spent UOX and MOX light water reactor (LWR) fuel without any uranium and thorium support and also with it. Other advanced reactor concepts are being studied [8, 9], which use the liquid salt technology, as a primary coolant for Fluoride salt-cooled High-temperature Reactors (FHRs), and coated particle fuels similar to high temperature gas-cooled reactors.

More generally, there has been a significant renewal of interest in the use of liquid salt as a coolant for nuclear and non-nuclear applications [10, 11]. These salts could facilitate heat transfer for nuclear hydrogen production concepts, concentrated solar electricity generation, oil refineries, and shale oil processing facilities amongst other applications.

The paper provides an overview of the main technical activities in the countries participating to the R&D effort on the MSR in GIF and remaining issues to be addressed.

II. MSR IN GENERATION IV

The decision for setting up a Provisional System Steering Committee (PSSC) for the MSR with Euratom, France, the Russian Federation and 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 in GIF. The Policy Group took the decision to set up a Memorandum Of Understanding (MOU) for both the MSR and LFR 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 France and JRC, on behalf of Euratom, October the 6th 2010. USA and Russia will remain as observers, but Russia is considering signing the MOU in the medium term future.

The members of the PSSC MSR, France and Europe, are working on MSRF (Molten Salt Fast Reactor) in which the salt is the fuel and the coolant. The common objective of these projects is to develop a conceptual design for an MSFR with an effective system configuration – resulting from physical, chemical and material studies – for the reactor core, the reprocessing unit and waste conditioning. The conceptual design activities are intended to increase the confidence that MSFR systems can satisfy the goals of Generation-IV reactors in terms of sustainability (Th breeder), non proliferation (integrated fuel cycle, multi-recycling of actinides), resource savings (closed Th/U fuel cycle, no uranium enrichment), safety (no reactivity reserve, strongly negative feedback coefficient) and waste management (actinide burner).

Russia which participates to the PSSC as observer works on flexible MOSART (MOlten Salt Actinide Recycler & Transmuter) system fuelled with different compositions of plutonium and minor actinide (MA) trifluorides with and without Th support. 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.

III. MSR CONCEPTS

Two reactors concepts using molten salt are studied in the GIF molten salt reactor provisional system steering committee, i) molten salt reactors, in which the salt serves as both the fuel and the coolant, and ii) reactors with solid fuel cooled by molten fluoride salts.

- **MSFR concept**

Recent conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts open promising possibilities to exploit the ^{232}Th-^{233}U cycle. On the other hand, they can also contribute to significantly diminishing the radiotoxic inventory from present-reactor spent fuels, in particular, by lowering the mass of transuranic elements (TRU).

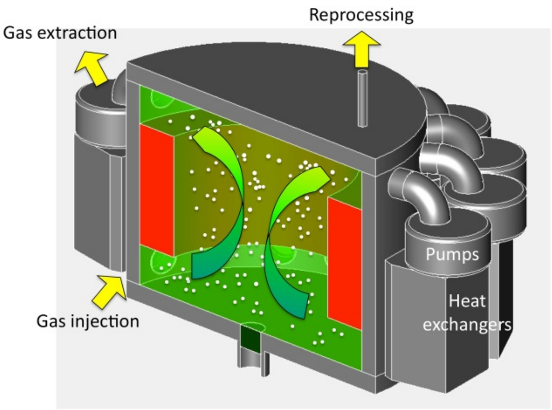


Fig. 1 Schematic conceptual MSFR design

In the MSFR, the liquid fuel processing is performed on a small side stream of the molten salt. Fission products are removed from the side stream and the remainder is then returned to the reactor. This is fundamentally different from a solid fuel reactor where separate facilities produce the solid fuel and process the used nuclear fuel. Because of this design characteristic compared to classical solid-fuel reactors, the MSFR can thus operate with widely varying fuel compositions.

Figure 1 sketches schematically general outlines for such a MSFR. The core consists of moving fuel loaded fluoride salt (note the lack of graphite moderation in core). The reference MSFR is a 3000 MWth reactor with a total fuel salt volume of 18 m^3, operated at a mean fuel temperature of 750°C. The salt is composed of lithium fluoride and thorium fluoride and the proportion of heavy nuclei is fixed at 22.5%. In preliminary drawings done in relation to calculations, the core of the MSFR is a single compact cylinder (2.25m high x 2.25m diameter) where the nuclear reactions occur within the liquid fluoride fuel salt acting also as the coolant.

The fuel salt flows freely from the bottom to the top of the central part of the core without any solid moderator. The return path of the salt (from the top to bottom) 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 outputs. The fuel salt runs through the total cycle in 3-4 seconds. 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 incident/accident that leads to a temperature increase in the core. Thus the entire fuel inventory can be passively drained by gravity into subcritical, passively cooled tanks.

- **Molten Salt Actinide Recycler & Transmuter (MOSART) concept**

MSR developments in Russia on the 2400MWt MOlten-Salt Actinide Recycler and Transmuter (MOSART) address the concept of large power units with a fast neutron spectrum in the core [6]. Promising configuration for 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 73LiF-27BeF_2 salt mixture. It is feasible to design critical homogeneous core fuelled only by TRU trifluorides from UOX or MOX LWR used fuel while equilibrium concentration for trifluorides of actinides (0.4 mole% for Li,Be/F core, with the rare earth removal cycle 300 epdf) is truly below solubility limit (~2 mole%) at minimal fuel salt temperature in primary circuit 600-620^{o}C. Recently [22], 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 2400MWt Li,Be/F MOSART core containing in initial loading 2 mole% of ThF_4 and 1.2 mole % of $TRUF_3$, with the LnF_3 removal cycle 300 epdf after 12 years of slow increasing of Th content in the solvent can operate without any $TRUF_3$ make up basing only on Th support as a self-sustainable system. The maximum concentration of TRU during this transition does not exceed 1.7 mole %. At equilibrium 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.

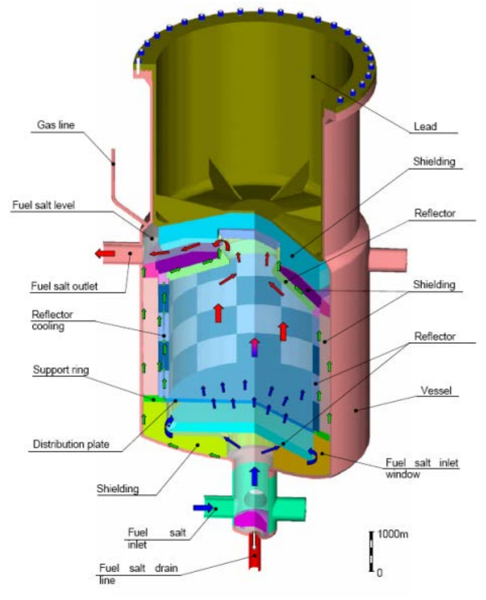


Fig. 2 MOlten Salt Actinide Recycler & Transmuter (MOSART) concept

- **Fluoride-salt-cooled high-temperature reactor concept**

FHRs are a class of reactor that, by definition, feature low-pressure liquid fluoride salt cooling, coated-particle fuel, and fully passive decay heat rejection. As high-temperature plants, FHRs can support either electricity generation or process heat production. While FHRs represent a distinct reactor class, they inherit desirable attributes from other thermal power plants whose characteristics can be studied to provide general guidance on plant configuration, anticipated performance, and costs. The extensive US molten salt reactor development program from the 1950s–1970s provides experience on the materials, procedures, and components necessary to use liquid fluoride salts. Liquid-metal reactors provide design experience on using low-pressure liquid coolants, passive decay heat removal, and hot refueling. High-temperature gas-cooled reactors (HTGRs) provide experience with coated-particle fuel and graphite components. Light-water reactors (LWRs) show the advantages of transparent, high-heat-capacity coolants with low chemical reactivity.

Several different FHRs are currently under design by different organizations. Oak Ridge National Laboratory (ORNL) is leading the preconceptual design of the Advanced High Temperature Reactor (AHTR – see Fig. 3), which is a large [1500 MW(e)] central station-type power plant focused on low-cost electricity production. The Massachusetts Institute of Technology (MIT) is leading the efforts toward developing a preconceptual design for a <20 MW(t) test reactor. The Shanghai Institute of Technology is leading a design effort to develop the first FHR critical facility/test reactor [2 MW(t)]. The University of California at Berkeley is developing a preconceptual design for a mid-sized [410 MW(e)] initial commercial prototype reactor. ORNL is also developing a preconceptual design of a Small modular Advanced High-Temperature Reactor [SmAHTR; 125 MW(t)] focused on thermal power production.

FHRs, in principle, have the potential to be low-cost energy producers while maintaining full passive safety. FHRs do not require any system or operator active response to avoid core damage or large off-site release of radioactive material for any design basis accident or non low-frequency beyond design basis accident, including severe earthquakes, tsunamis, large commercial plane impact, or permanent station blackout. The safety characteristics of FHRs arise from fundamental physics as well as well-designed, constructed, and maintained systems, structures, and components (SSCs). As with other high-temperature plants, FHRs can efficiently produce both electricity and process heat, including effective support for liquid hydrocarbon fuel production.

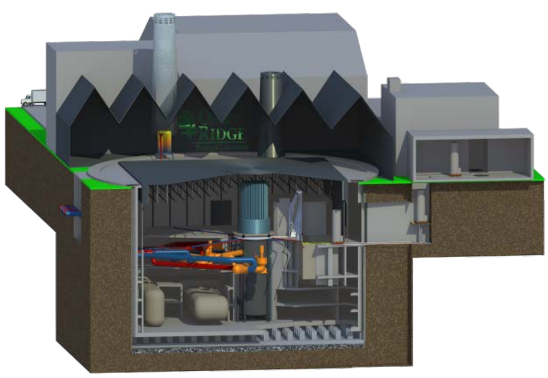


Fig. 3 AHTR reactor building layout overview

FHRs are a research focus of the U.S. Department of Energy's advanced reactor concepts program. The U.S. program includes technology development and demonstration as well as concept design studies focused on a test reactor, small modular reactors, and large-scale power plants. FHRs are also included within the research plans of both the Chinese and Indian civilian nuclear energy programs.

IV. R&D OBJECTIVES AND PROGRESS WITHIN PSSC-MSR

The common objective of these projects is to propose a conceptual design of MSFR as the best system configuration – resulting from physical, chemical and material studies – for the reactor core, the reprocessing unit and wastes conditioning. It is intended to deepen the demonstration that the MSFR system can satisfy the goals of Generation-IV 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). Those topics are the subject of the following sub-sections:

- Physical studies
- Safety
- Materials studies
- Salt properties
- Salt reprocessing
- Technological studies

IV.A Physical studies

Feedback coefficient evaluation

The potential of MSFRs without moderator in the core leading to a fast neutron spectrum while ensuring excellent safety coefficients was highlighted [1]. Various MSR configurations were studies by modulating the amount of graphite in core to obtain a thermal, an epithermal, or a fast spectrum. In particular, configurations of a fast spectrum MSR (MSFR) have been identified with outstanding safety characteristics and minimal fuel-reprocessing requirements. It has very negative feedback coefficients. This is true not only for the global temperature coefficient but also for the partial coefficients that characterize the dilatation or the heating of the salt, and the void effect.

First core and deployment capacities

Studies of the different starting modes of the MSFR have been performed [12, 13]. The MSFR concept may use as initial fissile load, ^{233}U or uranium or also the transuranic elements currently produced by light water reactors. The characteristics of these different launching modes of the MSFR and the Thorium fuel cycle have been studied, in terms of safety, proliferation, breeding, and deployment capacities of these reactor configurations.

Studies show that the MSFR configurations corresponding to various starting modes of the reactor are all characterized by excellent safety coefficients and have the same good deployment capacities. Optimizing the specific power in the MSFR configuration started directly with ^{233}U as initial fissile matter has allowed a reduction of the initial fissile inventory down to 3 metric tons per GWe. The MSFR is characterized by a low proportion of minor actinides in the salt (around one percent at equilibrium) and by its excellent safety coefficients (-5 pcm/°C).

^{233}U does not exist on earth and is not being directly produced today. The possibility of using in MSFR the transuranic elements (TRU) currently produced in the world as an initial fissile load has been investigated. MSFRs can be started with the Pu+Minor Actinides (TRU) extracted from used Uox fuel discharged from LWR reactors. The TRU-started MSFR is able to efficiently convert the plutonium and minor actinides from generation 2-3 reactors in ^{233}U while improving the deployment capabilities of the MSFR concept. A transition can be effected to the $^{232}Th/^{233}U$ cycle. The time scale for an almost complete transition is approximately one century. Its only drawback lies in its high initial plutonium concentration above its estimated solubility limit in the LiF-ThF_4 reference salt. To overcome this limitation while still using TRU elements in the initial fissile load of the MSFR to close the current fuel cycle, two optimized solutions have been proposed: mixing the TRU elements at a lower concentration (around 3 to 4 mol%) with either natural uranium with an enrichment ratio of 13% or ^{233}U produced in other reactors.

Coupling of neutronic and reprocessing simulation codes

Essentially, because the salt is the moderator, the coolant and the fuel, the study of MSFR are specific. There are strong coupling between neutronics and other part of physic field like the chemistry for instance. Simulations of the MSFR concept rely on numerical tools making use of the MCNP neutron transport code coupled with a home-made materials evolution code [14, 15] (see figure IV).

The coupling of neutronic and reprocessing simulation codes in a numerical tool has been used to calculate the extraction efficiencies of fission products, their location in the whole system (reactor and reprocessing unit) and radioprotection issues.

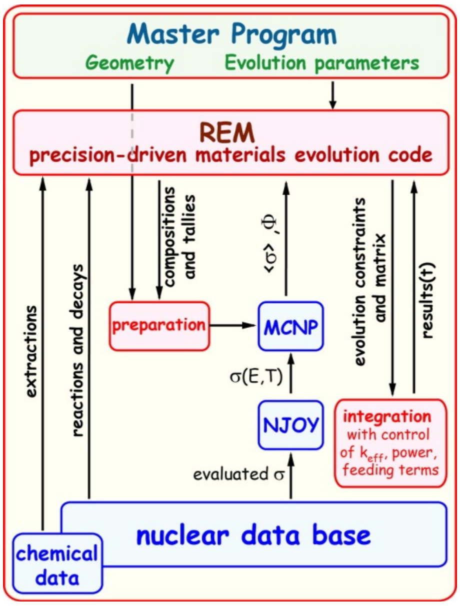


Fig. 4: Coupling scheme of the MCNP neutron transport code with the in-house materials evolution code

Preliminary results based on rough data of the pyrochemical processes involved, illustrate the potential of the neutronic-reprocessing coupling that has been developed. Studies are however still limited by the uncertainties on the design and knowledge of the chemical reprocessing processes.

Detailed neutronic and thermal-hydraulic behaviors of the core have been considered up to now with simplified coupling approach. Actions are under progress at the present time through EVOL projet at the european scale and through a local collaboration in France to developp à 3D coupled model for the MSFR core.

IV.B Safety

Molten salt reactors are liquid fuel reactors so that they are flexible in operation but very different in the safety approach from solid fuel reactors. Since this new nuclear technology is in development, safety is 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 MSFR, limited to the main systems surrounding the core [16].

Thanks to the negative reactivity feedback coefficient, the main scenarios lead to a reactor shut down.

In order to assess the behavior of the fuel salt after reactor shut down, a tool to calculate the decay heat has been developed and validated. It can be concluded that the decay heat in the core and the fuel loops of the MSFR is relatively low (3.5% of nominal power compared to 6% in a PWR) primarily thanks to the reprocessing system. The fission products that remain in the core contribute to the fuel salt heating up to 3% of nominal power. An important part of the decay heat (around 2% of nominal power) is located in the reprocessing units, mainly in the gas reprocessing unit, so that its safety assessment should be studied separately. The actinides also have an important contribution (0.5% of nominal power), that becomes dominant some hours after reactor shut down.

With a tool based on point kinetics, loss of heat sink transients can be calculated and their impact on the fuel salt temperature studied. The results of this study demonstrate the importance of the inertia of the systems. We conclude that slow transients (> 1 minute), thanks to a large system inertia, are advantageous and that, with them, the fuel salt temperature increase is slower.

These residual heat calculations will be the basis for the design of the draining system, as drainage must occur for any reactor shut down, whether in normal or in accidental conditions. The impact of the stagnant heating fuel salt on the core and fuel loop systems will be studied as well. It appears that slow transients are favorable (> 1min) to minimize the temperature increase of the fuel salt.

IV.C Material studies

The structural materials retained for MSR container are Ni-based 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. The composition of this optimized Hastelloy N (Ni- 8wt% Cr- 12wt% Mo) proved satisfactory up to 750°C, a temperature in the low range of the MFSR. The operating temperatures chosen in neutronic calculations of MSFR systems are ranged between 700 and 850°C. Due to the evolving microstructure of optimized Hastelloy N at higher temperature, it would be impossible to preserve the required material properties in the full operating temperature range required for the MSFR system.

For this high temperature domain, the replacement of Mo by W could prove beneficial for mechanical properties since tungsten diffusion is roughly ten times slower in nickel than molybdenum diffusion. A better creep resistance is expected with a Ni-W solid solution than with a Ni-Mo solid solution. This would help to reach higher in-service temperature. First results show that such material have the required properties, especially in terms of compatibility with molten salts and mechanical properties.

Experimental studies focus on the potential for using Ni-W-Cr alloys as structural materials for MSFR system. The corrosion of a specific Ni-25W-6Cr (wt.%) alloy was studied in a LiF-NaF molten salt, at 750°C and 900°C, for 350 h and 900 h. The results showed, as expected, a selective oxidation of Cr in the alloy. They also evidenced a noticeable and unexpected corrosion of W, which might be attributed to the combined presence of some pollution (by O^{2-} and Fe^{2+} ions) in the salt.

It has been demonstrated that the salt redox potential is a key parameter in the corrosion phenomena of structural materials of MSRs [10, 17]. The chemical corrosion can be controlled by a redox buffer which controls the potential of fuel salt. The redox buffer considered 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 reaction. Addition of a reducing agent leads to a decrease of the fuel salt potential. The use of an acido-basic buffer to control also the oxo-acidity of the molten salt could stabilize the chromium oxide in the alloy and contribute to the formation of a protecting layer at the alloy surface. The experimental feedback from the ORNL has demonstrated the high corrosion resistance of Ni-based alloys in fluoride molten salts. An innovative method (scanning electrochemical microscopy SECM) has been proposed to improve the understanding of the corrosion mecanisms at a microscopic scale. Efforts are also made on reactor vessel design to suppress the highest temperature points, and protect or cool some areas. This improvement process will nethertheless very correlated to the obtaining of a correct 3D simulation of the core.

A wide range of problems lies ahead in the design of high temperature materials for molten salt reactors. The Ni-W-Cr system looks promising. Its metallurgy and in-service *properties need to be investigated in further details regarding irradiation resistance and industrialization. Additional tests are being carried our in order to better understand the W behavior and eventually suppress its corrosion using a highly purified solvent. A 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.*

IV.D Salt properties

Thermodynamic properties of the salt systems are investigated (JRC/ITU) in order to collect new data which are necessary for developments of molten salt reactor designs, reprocessing scheme and simulation codes [18, 19]. A strong tool can be found in the assessment of phase diagrams. This method is based on the Gibbs free energy minimization between the different phases. With a good description of the phase diagram it is also possible to predict some properties, e.g. vapor pressure, for which no experimental information is available. Determination and modeling of molten fluoride properties salts are developed in parallel with experimental facilities:

- alpha tight glove box which for synthesis and purification of actinide fluorides,
- a Raman spectrometer set up for measurements of the Raman spectra of molten salts. This set-up will allow the determination of the local structure of the actinides in the fluoride salts.

Experimental investigation of physico-chemical properties of actinide fluorides containing salts are carried out. To elucidate the influence of the salt composition on thermo-physical properties of the MSR fuel (melting temperature, solubility of actinides and vapour pressure), it is necessary to understand the phase equilibria in the fuel system. The thermodynamic properties of all phases considered in a multi-component system such as the MSR fuel have to be assessed. Extensive thermodynamic database of various fluoride systems is thus being developed.

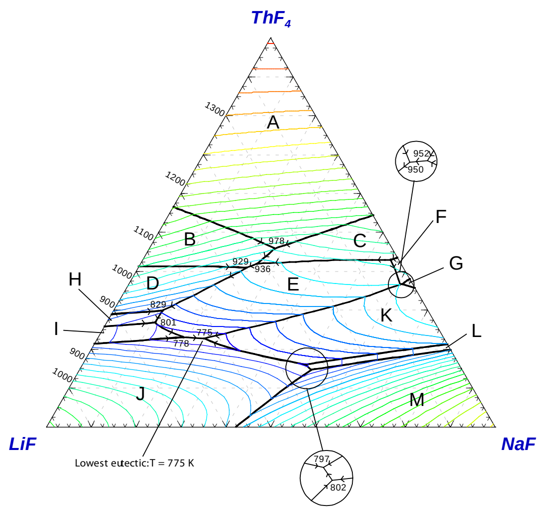


Fig. 5: The calculated LiF-NaF-ThF$_4$ pseudo-ternary phase diagram with fixed concentration of UF$_4$ set to 2.55 mol\%.

Among these properties the heat capacity, is especially important for the heat transfer evaluations within the various loops of MSR. Using a drop calorimetry, a systematic study of the heat capacity of binary LiF-AlkF (Alk = Na, K, Rb, Cs) systems has been finalized. Based on these results it appears that increased heat capacity can be expected in multi-component fluoride mixtures compared to its pure components contributing to higher safety of MSR as the higher the heat capacity the higher the buffer zone for overheating of a reactor during off-normal or accidental conditions.

Novel technique to measure mixing enthalpies of fluoride liquid solutions using a differential scanning calorimeter has been developed and first tested on the LiF-KF system showing excellent agreement to literature values. Using this promising technique mixing enthalpies of the LiF-ThF$_4$ system was first measured and the fusion enthalpy of the Li$_3$ThF$_7$ intermediate compound was determined. Furthermore, from this experimental campaign new phase diagram data points of the whole LiF-ThF_4 system was obtained.

The effort dedicated to the construction of the thermodynamic database that is being developed at ITU since 2002 will continue with the acquisition of data on TRU elements.

IV.E Salt reprocessing

The on-site salt management of the MSFR combines a salt control unit, an on-line gaseous extraction system (see V.F Molten salt technological studies) and an offline lanthanide extraction component by pyrochemistry. This salt reprocessing scheme is presented in Fig. 6.

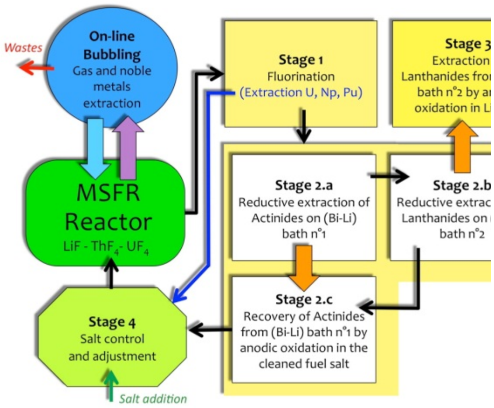
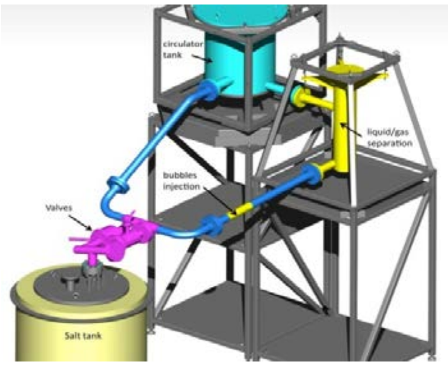


Fig. 6: MSFR reprocessing scheme

The salt properties and composition are monitored through the on-line chemistry control and adjustment unit. A fraction of salt is periodically withdrawn and reprocessed off-line in order to extract the lanthanides before it is sent back to the core. In this separate batch reprocessing unit 99% of Uranium (including ^{233}U) and Neptunium, 90% of Plutonium are extracted by fluorination and immediately reintroduced in the core. The remaining actinides are then quickly extracted together with Protactinium and also sent back to the core. Finally, the lanthanides are separated from the salt through a second reductive extraction and sent to waste disposal.

The reference scheme depicted in Fig. 6 involves 4 stages for the batch on-site fuel processing [20, 21]. The peculiarity of the concept appears in stages 2 and 3 by combining chemical and electrochemical methods for the extraction and the back extraction of actinides and lanthanides. This choice leads to fuel processing without effluent volume variation. For the core of the flow sheet, the process proposed is a reductive extraction using a liquid metal solvent. Some analytical relations have been established (considering experimental redox potentials and activity coefficients in molten salt and liquid metal) to understand the influence of the liquid solvent composition on the extraction efficiency. The liquid metal is constituted of Bi which is the metallic solvent and of Li which is the reductive



reactant.

The experimental tests of extraction process require an optimized procedure for the preparation of the metallic phase. The composition of the metallic phase is a key point for the extraction efficiency. Different procedures of metallic phases preparations have been tested. The method retained for the preparation of the metallic phase is the electrolysis of LiCl-KCl.

The progress made in core design in the last two years has opened the door for the definition of an improved fuel salt reprocessing scheme with a realistic fuel clean-up rate (40 l/day) and minimized losses to wastes. This value is almost two orders of magnitude less than the reference MSBR scheme.

Acquisition of fundamental data for the extraction processes is still needed especially for the actinide-lanthanide separation. The extraction of lanthanides has to be done because of the low solubility of these trifluoride elements and neutronic captures that decrease the reactivity balance.

IV.F Molten salt technological studies

The gaseous extraction system is a continuous salt chemistry process. Helium bubbles are injected at the lower part of the core to trap the non-soluble fission products (noble metals) dispersed in the flowing liquid as well as the gaseous fission products. A liquid/gas phase separation is then performed on the salt flowing out of the core to extract gaseous species and dragged condensed particles. Following this "physical" process of purification, a small part of the gas is withdrawn to let the fission products decay, and the remaining part of gas is sent back to the lower part of the core.

Bubbling treatment needs the insertion of an injector and a liquid gas separator in the salt circuit between the core and heat exchangers. In order to begin the conception of the bubbling components for reactor scale, an experimental project was launched, based on the construction of a molten salt loop (Forced Fluoride Flow for Experimental Research project – CNRS/LPSC). FFFER is dedicated to bubbling studies and is operated with LiF-NaF-KF salt.

Fig. 7: Forced Fluoride Flow for Experimental Research project (FFFER)

Studies dedicated to bubbling cleanup process have led to the conception of a liquid-gas separator with satisfying efficiency when measured on water mock up. The volumic gas rate domain investigated is between 0.02 to 0.5%. Specific features determined on the mock up are reported on the metallic separator internal design. The whole design of the loop has been then achieved. It is presented in Figure 7 together with a picture of the FFFR partial running test. The loop tank separation system is composed of two parts in parallel connection, a metallic valve and a cold plug. The salt tank can contain up to 100 liters but the loop circuit is designed for running with a volume ranging from 50 to 80 liters.

Fabrication of the salt mixture (LiF-NaF-KF) to be used in the French molten salt loop (FFFER project) has been achieved. Tests with liquid salt have been undertaken to prove the ability of our cold plug system to play the role of a security valve on the loop circuit. Satisfying results have been obtained; modifications on the first cold plug design have been done to improve the resistance to corrosion of the whole component. Further evolutions of this component will be made on a separated system in glove boxe, to explore other design possibilities and to acquire data for simulation.

Implementation of instrumentation (temperature, level and flow rate measurement) on the whole experimental set up is under progress. The start of the loop running is forseen for the middle of year 2013. The future R&D studies will focus on gas and particles extraction efficiency (gas/salt separation, gas analysis by mass spectrometry).

V. R&D PROGRESS FOR MOSART AND FHR CONCEPTS

- **MOSART**

In Russia, study is under progress within ISTC#3749 and MARS projects to examine the conceptual feasibility of flexible MOlten Salt Actinide Recycler & Transmuter (MOSART) system fuelled with different compositions of plutonium and Minor Actinide (MA) trifluorides with and without Th support [22]. New fast-spectrum design options and salt compositions with adequate solubility for actinide trifluorides are being examined with objective to obtain reliable and abundant source of energy through efficient use of transuranium elements from used LWR fuel as well as uranium and thorium resources. Experimental data base created within the projects is used for further development of technology as applied to consumption of actinides while extracting their energy.

Key thermal physical and chemical properties of molten binary LiF-BeF_2, LiF-NaF, and LiF-ThF_4, ternary LiF-BeF_2-ThF_4 and LiF-NaF-KF mixtures important for the design calculation were experimentally studied. Melting temperatures, plutonium and americium trifluorides solubility for the mentioned above salt solvent systems are measured. New experimental data on viscosity, density, thermal conductivity and heat capacity for selected molten binary and ternary salts are received in temperature range from liquidus temperatures till to 750°C.

Particularly, for 78LiF-22ThF_4 (mole%) fuel solvent systems used both in MOSART and MSFR designs following experimental dependences on the PuF_3 solubility (lgS, logarithm of PuF_3 molar concentration), density (ρ,g/cm^3), thermal conductivity (λ,W·m^{-1}·K^{-1}), heat capacity (c_p,J·g^{-1}· K^{-1}) and viscosity (ν, 10^{-6} m^2/s) vs. temperature (T,K) in range from liquidus up to 1100K were, respectively, obtained [23]:

$\lg S = 2.58 - 1733 / T$; $\rho = 4.742 - 8.82 \cdot 10^{-4}\, T$; $\lambda = 0.928 + 8.397 \cdot 10^{-5} T$; $c_p = -1.111 + 0.00278 \times T$ and $\nu = 1.9798 \exp\{3689 \times (1/T - 0.9698E\text{-}3)\}$.

Electrochemical behavior of the dissolved trifluorides in molten LiF-BeF_2, LiF-ThF_4, and LiF-BeF_2-ThF_4 solvent systems selected are studied. New experimental data on reductive extraction of the lanthanum, neodymium and thorium for the molten salt/liquid bismuth systems at 650°C are obtained. The measured distribution coefficients are consistent with the earlier data obtained for binary LiF-BeF_2 and LiF-ThF_4 systems as well as for ternary LiF-BeF_2-ThF_4 salt mixtures. The distribution coefficients obtained for LiF-ThF_4 and LiF-BeF_2-ThF_4 salts with relatively high concentration of ThF_4 (> 20 mole%) can not provide the effective separation between thorium and lanthanides in the fluoride salt/bismuth solutions. Excellent separation of thorium from lanthanides and alkaline-earth elements can be made by use of LiCl. The distribution coefficient for thorium is decreased sharply by addition of fluoride to the LiCl, although, the distribution coefficients for the rare earths are affected by only a minor amount [23].

Results of five corrosion tests with exposure time 250 hrs each done with Li,Be,Th,U/F fuel salt containing also Te additions at 720–740°C in the range of U(IV)/U(III) ratios from 1 to 500 demonstrated that high temperature operations are feasible using carefully purified molten salts and loop internals. In these tests device for voltammetric redox potential evaluation was successfully used [24]. The nickel-based alloys selected for testing had the following compositions (in % mass): HN80M-VI (Mo 12, Cr–7.6, Nb 1.5), HN80MTY (Mo–13, Cr–6.8, Al–1.1, Ti–0.9), HN80MTB (Mo 9.4, Cr 7.0, Ti 1.7, W 5.5) and EM 721 (Cr 5.7, Ti 0.17, W 25.2) [23].

After materials exposure in the fuel salt with the [U(IV)]/[U(III)] ratio from 1 to 100 there was revealed no traces of tellurium intergranular cracking on specimens surface for all alloys under study except HN80MTB. Tellurum intergranular cracking was found on tested alloys only after exposure in fuel salt with [U(IV)]/[U(III)] = 500. For each of the tested alloys the intensity of tellurium intergranular cracking was essentially lower in unstressed state than in stress condition. Study on deuterium permeation through nickel-based HN80MTY and EM721 alloys is also carried out. Temperature dependences of deuterium solubility, coefficients of permeability and diffusion in alloys were built. Next Te corrosion test will focus on Li,Be,U/F fuel salt at temperature 750°C.

- **FHR**

The U.S. Department of Energy’s (DOE) Office of Advanced Reactor Concepts (ARC) sponsors the U.S. FHR development efforts. Oak Ridge National Laboratory has technical leadership for the program with Idaho National Laboratory performing key fuel qualification and heat exchanger design tasks. During 2011, DOE awarded a significant new university based integrated research program with multiple, interrelated research tasks and a focus on developing a conceptual design for an FHR test reactor. The project is being performed by a team lead by the Massachusetts Institute of Technology along with the University of California at Berkeley and the University of Wisconsin.

In 2012 the ARC FHR development program focused on maturing the design for the Advanced High Temperature Reactor (AHTR). The AHTR is a design concept for a central generating station type [3400 MW(t)] FHR. The overall goal of the AHTR development program is to demonstrate the technical feasibility of FHRs as low-cost, large-size power producers while maintaining full passive safety. A pre-conceptual design study on a small, modular FHR (SmAHTR) was also completed in late 2010. The AHTR design studies focused on developing a reasonable core and fuel design and placing the proposed core within a power plant.

Development of a fluoride salt component test facility is under progress. The principal activity was construction of a fluoride salt test loop (see Fig 8). Demonstration of wireless (inductive) heating of fuel element surrogates in a salt environment, integrating silicon carbide components into a fluoride salt loop, and development and demonstration of a fluidic diode for liquid fluoride salt application were the main research topics.

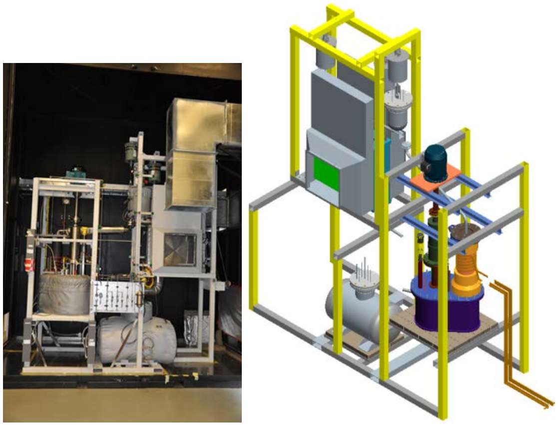


Fig. 8 ORNL Liquid Salt Test Loop (as design drawing (left) and as constructed (right))

A cooperative research program between U.S. Department of Energy and the Czech Republic Ministry of Industry and Trade was also initiated during 2011. The project's technical objective is improving the understanding of the reactivity worth of lithium isotopically selected $2LiF\text{-}BeF_2$ salt. The program involves provision of U.S. produced isotopically separated salt to the Nuclear Research Institute at Řež for testing the salt's reactivity worth at their LR-0 critical facility.

VI. CONCLUSION

Since 2005, R&D on MSR has been focused on fast spectrum concepts (MSFR) which have been recognized as long term alternatives to solid-fuelled fast neutron reactors with attractive features (very negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle…). MSFR designs are available for breeding and for minor actinide burning. They are robust reference configurations (with significant improvement compared to MSBR), allowing to concentrate on specific R&D issues.

Although the European and USA interests are focused on different baseline concepts (MSFR MOSART and FHR), large commonalities in basic R&D areas (liquid salt technology, materials) exist and the Generation IV framework is useful to optimize the R&D effort.

A network on MSR R&D has been active in Europe from 2001 with financial support by EURATOM. Partners of the MSR PSSC are involved in the Euratom-funded EVOL (Evaluation and Viability of Liquid Fuel Fast Reactor Systems) project and ISTC#1606 and #3749 projects. ISTC has provided another efficient way of collaboration between Russian research organizations, European partners and non-European partners (USA, Japan, Korea, Canada). A complementary ROSATOM programme named MARS (Minor Actinides Recycling in Molten Salt) project between Russian researches organizations is carried out in parallel to Euratom EVOL project.

The common objective is to propose a conceptual design of MSFR by 2013 as the best system configuration – resulting from physical, chemical and material studies – for the reactor core, the reprocessing unit and wastes conditioning. It is intended to deepen the demonstration that the MSFR system can satisfy the goals of Generation IV 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).

Acknowledgements

The paper has been written with major contributions by M. Allibert, P. Rubiolo from CNRS.

Nomenclature

AHTR Advanced High Temperature Reactor
GIF Generation IV International Forum
LWR Light Water Reactor
MA Minor Actinides
MOSART MOlten Salt Actinide Recycler & Transmuter
MSR Molten Salt Reactor
MSFR Molten Salt Fast Reactor
ORNL Oak Ridge National Laboratory
PSSC Provisional System Steering Committee
UOX Uranium Oxide
TRU Transuranic elements

References

1. L. Mathieu, D. Heuer, E. Merle-Lucotte et al., "Possible Configurations for the Thorium Molten Salt Reactor and Advantages of the Fast Non-Moderated Version", *Nucl. Sc. and Eng.*, **161**, 78-89 (2009)

2. C.W. Forsberg et al, "Liquid Salt Applications and Molten Salt Reactors", *Revue Générale du Nucléaire* **N°** 4/2007, 63 (2007)

3. E. Merle-Lucotte, D. Heuer et al., "Introduction of the Physics of Molten Salt Reactor", *Materials Issues for Generation IV Systems, NATO Science for Peace and Security Series - B, Editions Springer*, 501-521 (2008)

4. E. Merle-Lucotte, D. Heuer et al, "Minimizing the Fissile Inventory of the Molten Salt Fast Reactor", *Proceedings of the Advances in Nuclear Fuel Management IV (ANFM 2009),* Hilton Head Island, USA (2009)

5. E. Merle-Lucotte, D. Heuer et al., "Optimizing the Burning Efficiency and the Deployment Capacities of the Molten Salt Fast Reactor", *Proceedings of the International Conference Global 2009 - The Nuclear Fuel Cycle: Sustainable Options & Industrial Perspectives,* Paper 9149, Paris, France (2009)

6. Ignatiev V. et al. (2005), Characteristics of molten salt actinide recycler and transmuter system. In: *Proceedings of International Conference on Emerging Nuclear Energy Systems.* Brussels, Belgium, 21-26 August: paper ICQ064
7. ISTC# 1606 final report, International scientific technical centre, Moscow, July, 2007

8. Forsberg, C.W., P.F. Peterson and R.A. Kochendarfer, "Design Options for the Advanced High-Temperature Reactor", *Proc. 2008 International Congress on Advances in Nuclear Power Plants (ICAPP '08),* Anaheim, CA USA, June 8-12, 2008

9. Bardet, Ph., *et al.*, "Design, Analysis and Development of the Modular PB-AHTR", *Proc. 2008 International Congress on Advances in Nuclear Power Plants (ICAPP '08)*, Anaheim, CA USA, June 8-12, 2008

10. V. Ignatiev, A. Surenkov, Material Performance in Molten Salts, *Comprehensive Nuclear Materials*, Volume 5, 2012, Pages 221-250

11. O. Beneš, R.J.M. Konings, Molten Salt Reactor Fuel and Coolant, *Comprehensive Nuclear Materials*, Volume 3, 2012, Pages 359-389

12. E. Merle-Lucotte, D. Heuer, M. Allibert, M. Brovchenko, N. Capellan, and V. Ghetta, "Launching the Thorium Fuel Cycle with the Molten Salt Fast Reactor"**,** Contribution 11190, *International Congress on Advances in Nuclear Power Plants (ICAPP)*, Nice, France (2011)

13. E. Merle-Lucotte, D. Heuer, M. Allibert, X. Doligez, V. Ghetta, "Optimizing the Burning Efficiency and the Deployment Capacities of the Molten Salt Fast Reactor", Contribution 9149, *Global 2009 - The Nuclear Fuel Cycle: Sustainable Options & Industrial Perspectives*, Paris, France (2009)

14. X. Doligez et al., "Numerical tools for Molten Salt Reactors simulations", *Proceedings of the International Conference Global 2009 - The Nuclear Fuel Cycle: Sustainable Options & Industrial Perspectives*, Paris, France (2009)

15. E. Merle-Lucotte, D. Heuer, M. Allibert, X. Doligez, V. Ghetta, "Simulation Tools and New Developments of the Molten Salt Fast Reactor", Contribution A0115, *European Nuclear Conference ENC2010*, Barcelone, Espagne (2010)

16. M. Brovchenko, D. Heuer, E. Merle-Lucotte, M. Allibert, N.Capellan, V. Ghetta, A. Laureau "Preliminary safety calculations to improve the design of Molten Salt Fast Reactor", *PHYSOR 2012 Advances in Reactor Physics Linking Research, Industry, and Education*, Knoxville, Tennessee, USA, April 15-20, 2012, on CD-ROM

17. S. Delpech, E. Merle-Lucotte, T. Augé, D. Heuer, "MSFR: Material issued and the effect of chemistry control", *GenerationIV International Forum Symposium*, Paris, France (2009)

18. M. Beilmann, O. Beneš, R.J.M. Konings, Th. Fanghänel, "Thermodynamic assessment of the (LiF + UF3) and (NaF + UF3) systems", *The Journal of Chemical Thermodynamics*, Volume 57, February 2013, Pages 22-31

19. O. Beneš, M. Beilmann, R.J.M. Konings, "Thermodynamic assessment of the LiF–NaF–ThF4–UF4", Journal of Nuclear Materials, Volume 405, Issue 2, 15 October 2010, Pages 186-198

20. S. Delpech, E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le-Brun, L. Mathieu, G. Picard, "Reactor physics and reprocessing scheme for innovative molten salt reactor system", J. of Fluorine Chemistry, 130 Issue 1, p. 11-17 (2009)

21. S. Jaskierowicz, S. Delpech, P. Fichet, C. Colin, C. Slim and G. Picard, "Pyrochemical reproces-sing of thorium-based fuel", *Proceeding of ICAPP2011*, Nice, France (2011)

22. V. Ignatiev et al, “Molten salt reactor: new possibilities, problems and solutions”, *Atomnaya energia* 2012, **112**: 3, p.135

23. V. Ignatiev, et al, “Progress in Development of MOSART Concept with Th Support”, ICAPP’12, Chicago, USA, June 24-28, 2012, Paper No. 12394

24. V. Afonichkin, A. Bovet and V. Shishkin, “Salts purification and voltammetric study of the electroreduction of U(IV) to U(III) in molten LiF–ThF_4”, *J. Nucl. Mater.* 2011; **419:** 1-3, p. 347