

OVERVIEW AND PERSPECTIVES OF THE MOLTEN SALT FAST REACTOR (MSFR) CONCEPT

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ABSTRACT

Recent conceptual developments on the design of fast neutron spectrum molten salt reactors (Molten Salt Fast Reactor or MSFR) using fluoride salts open promising possibilities to exploit the ^{232}Th - ^{233}U cycle. On the other hand, the MSFR concept can also contribute to significantly diminish the radiotoxic inventory from present-reactors spent fuels in particular by lowering the masses of transuranian elements (TRU). This paper provides an overview of the current status of the MSFR design, the ongoing research activities at the National Centre for Scientific Research (CNRS, France) and the future perspectives of the concept.

1. INTRODUCTION

Molten Salt Reactor (MSR) technology, including two demonstration reactors, was partly developed in the 1950's and 1960's in USA (Oak Ridge National Laboratory). These Molten Salt Reactors were considered as thermal-neutron-spectrum graphite-moderated concepts. Since 2004, the National Centre for Scientific Research (CNRS, Grenoble-France) has been focused R&D efforts on the development of a new fast-spectrum reactor based on the MSR concept. The proposed MSFR (Molten Salt Fast Reactor) combines then the generic assets of fast neutron reactors (e.g. reduced absorptions in the fission products) with those related to molten salt fluorides as fluid fuel and coolant [1,2,3] (e.g. low pressure and high boiling temperature, optical transparency). As opposed to thermal molten salt reactors, the specificity of the MSFR is the removal of the solid moderator (usually graphite) in the core. This choice is motivated by findings from parametric studies involving reactor design variables such as the breeding ratio, graphite lifespan, reprocessing requirements, ^{233}U initial inventory and safety issues (reactor feedback coefficient). The current neutronic design of the MSFR exhibits then large negative temperature coefficient, a unique safety characteristic not usually found in solid-fuel fast reactors. In addition, compared to solid-fuelled reactors, MSFR systems can operate with widely varying fuel composition and 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. The Generation IV International Forum has selected in 2008 the MSFR concept as one of the GEN IV reference reactors because of its unique capabilities for actinide burning and extending fuel resources. Since 2009 other European R&D and Russian organisations have been worked jointly with the CNRS (France) in the EVOL project (Evaluation and Viability of Liquid Fuel Fast Reactor System) to advance the conceptual design and to demonstrate that the MSFR satisfies all the goals of Generation-IV. The paper provides a general overview of the MSFR concept and then describes the research activities related to: design studies, safety, material studies, salt properties and reprocessing.

2. THE MOLTEN SALT FAST REACTOR CONCEPT

The reference MSFR design is a 3000 MWth reactor with three different circuits (or loops): the fuel circuit, the intermediate circuit and the power conversion system. The main components of the fuel circuit are: the fuel salt which serves as fuel and coolant, the core, the

inlet and outlet pipes, the gas injection system, the salt-bubble separators, the fuel heat exchangers and the pumps. The salt in the fuel loop is made of lithium fluoride and thorium fluoride with a proportion of heavy nuclei fixed at 22.5%. The total fuel salt volume in the fuel loop is about 18 m³ and the mean salt temperature of about 750°C. As shown in the sketch of Figure 1, the fuel salt flows from bottom to the top of the core cavity. After exiting the core, the fuel salt is fed into 16 groups of pumps and heat exchangers (HXs) located around the core. The fuel salt circulates in the fuel circuit in around 3-4 seconds. Potential candidates to be used as working fluids for the other MSFR circuits are still under study. They include a fluoride salt or lead (Pb) for the intermediate circuit and supercritical water for the power conversion system. As can be seen in Figure 1, the entire fuel circuit is contained inside a reactor vessel which acts as a second barrier.

The fuel circuit design has been optimized to reduce the fuel salt inventory outside the core cavity to about half of the total. As shown in Figure 1 the core cavity where the fuel salt produces the majority of the fissions can be roughly approximated as cylinder having a height to diameter ratio of one (2.25m x 2.25m). However, Figure 1 should be considered only as an outline since the MSFR core cavity shape is rather complicate. Three important components of the core are: i) the upper and lower axial neutron reflectors and ii) the radial fertile blankets (shown in red in Figure 1) which is part of the radial reflector. The thick axial reflectors are made of nickel-based alloys and have been designed to absorb more than 99% of the leaking neutron. The radial reflector includes a fertile blanket of about 50 cm thick to increase the breeding ratio. This blanket is filled with a fertile salt of LiF-ThF₄ with initially 22.5mole % ²³²ThF₄. The radial reflectors are enclosed by a 20 cm thick layer of B₄C, which provides additional neutron protection of the HXs. The fuel circuit includes a salt draining system which can be used for a planned shut down or in case of incident/accident leading to an excessive increase of the temperature in the core. In such situations the fuel salt geometry can be passively reconfigured by gravity draining of the fuel salt into tanks located under the reactor and where a passive cooling and adequate reactivity margin can be obtained.

Another important reactor component is the fuel reprocessing unit which has to be considered from the early stages of the design and safety studies. In the MSFR, this unit extracts a small amount of the fuel salt for fission product removal and then returned to the reactor. This is fundamentally different from a solid fuel reactor where separate facilities to produce the fuel and to process the spent fuel are needed.

3. DESIGN STUDIES

This section presents various design studies where R&D effort has been focused in order to the advance the reactor design.

Selection of the fuel salt

The selection of liquid fuel is guided by the MSFR operational criteria and also the need to fulfill the GEN IV criteria. Two liquids, a fluoride salt and a chloride salt, were retained to investigate if the above criteria could be fulfilled. A first evaluation showed that both salts have different advantages and inconvenients regarding the chemical and thermodynamics properties and thus a selection should be done on the basis of neutronics analyses. These analyses consisted on MCNP simulations of the MSFR using each type of salt. These analyses showed that there are important disadvantages in the use of a chloride salt. In particular, i) the breeding ratio of a MSFR operated with a chloride salt is degraded as compared to that of a MSR operated with a

fluoride salt, ii) the radiation damages in neutron-irradiated materials close to the reactor core are about 4 times higher when using a chloride salt (in part because neutron spectrum is harder as can be seen in Figure 2, and iii) the presence of Chlorine (Cl) in the salt leads to the production of ^{36}Cl , which is a mobile isotope, similar to the Tritium produced in the fluoride salt but with a larger radioactive period (301,000 years versus 12 years for the tritium). For the stated reasons, a fluoride salt was finally chosen as the reference fuel salt for the MSFR.

Feedback coefficient evaluation

During the initial CNRS studies of the Molten Salt Reactors, the effect of the neutron spectrum was investigated by changing the amount of graphite in core. The feedback coefficient of each MSR configuration was then calculated from three contributions: the salt Doppler effect, the salt density effect and the moderator effect (when graphite is presented). These studies shown that the fast spectrum MSRs (or MSFRs) have better safety characteristics and minimal fuel-reprocessing requirements. In particular they are characterized by very negative feedback coefficients. This is true not only for the global temperature coefficient but also for the partial coefficients.

Reactor control

The MSRF reactivity control is achieved without the use of any active control mechanism such as absorbent rods or neutron poisons but rather relying exclusively on the balance between the power generated in the fuel salt and the power extracted in the Heat Generators (HXs). As illustrated in the example of the left side of Figure 3, when the energy demand increases, the power extracted becomes larger than the power produced by fission in the core. The fuel salt temperature decreases, leading to a reactivity increment. This behavior is illustrated, for example, by the green curves in Figure 3 which correspond to sudden power increase up to 150% at the Heat Exchangers (HXs). As shown in Figure 3.b, this effect increases temporally the core power over the HXs power demand allowing restoring the initial temperature (Figure 3.c) and the zero reactivity (Figure 3.a). After a short transient of tens of seconds, the reactor returns to equilibrium at its nominal temperature but operating at a higher power level (i.e. 150%). The reactor transient responses to different reactivity insertions values are also shown on the right side of the Figure 3.

It is important to note that on the contrary of solid fuel reactors which obtain adequate reactivity margin during cold shutdown by using either absorbent rods or a soluble poison in the coolant; the MSFR do not need these mechanisms since it can takes advantage of the capacity of a liquid fuel to be transported and change the geometry. In the MSFR adequate reactivity margin during cold shutdown is then obtained by draining the fuel salt into the draining tanks where the fuel is cooled and whose geometries allow decreasing the fuel salt temperature without returning to criticality. The absence of absorbent rods simplifies the reactor operation and eliminates some accident initiators (such as for example a rod ejection).

First core and deployment capacities

Studies of the different starting modes of the MSFR have been performed [3]. The MSFR concept may use as initial fissile load, ^{233}U , enriched uranium or transuranic elements currently produced by light water reactors. The characteristics of these different starting scenarios and the implementation of a Thorium fuel cycle have been studied, in terms of safety, proliferation, breeding, and deployment capacities of these reactor configurations. These studies have shown

that the MSFR corresponding to these various starting scenarios have excellent safety coefficients ($\cong -5$ pcm/K) and the same good deployment capacities.

For example, the optimization of the core specific power in the scenario where the reactor is started directly with ^{233}U as initial fissile load allows a reduction of the initial fissile inventory down to 3-4 metric tons per GWe. The possibility of using in MSFR the transuranic elements (TRU) currently produced in the world as an initial fissile load has also been investigated, since ^{233}U does not exist on earth and is not being directly produced today. In this scenario, the MSFRs is started with the Pu+Minor Actinides (TRU) extracted from used UOX fuel discharged from LWR reactors. The TRU-started MSFR is able to efficiently burn the plutonium and minor actinides from generation 2-3 reactors and breed ^{233}U while improving the deployment capabilities of the MSFR concept. A complete transition to a $^{232}\text{Th}/^{233}\text{U}$ fuel cycle can be then performed in approximately one century. The main technical problem in this scenario is high initial plutonium concentration which is usually above the solubility limit of the reference fuel salt (LiF-ThF_4). Two possible solutions to overcome this problem are using mixing a lower concentration of the TRU elements (around 3 to 4 mol%) with either uranium having an enrichment ratio of 13% or ^{233}U produced in other reactors.

Coupling of neutronic and reprocessing simulation codes

The fuel salt reprocessing creates a coupling between the reactor dynamics (i.e. variation of the neutronics parameters over the cycle length or the life span of the reactor) and the fuel salt chemistry (i.e. the performance of the fuel salt reprocessing unit). Proper numeric simulation of the MSFR fuel salt isotopic composition required then the development of a numerical tool which combines the MCNP neutron transport code coupled with a MSFR code called REM which calculates fuel composition changes due to fuel burnup and reprocessing [4,5]. An overview of the coupling between these two codes is provided in Figure 4.

This numerical tool has allowed calculating the efficiencies of the extraction of fission products, their location in the whole system (reactor and reprocessing unit) and radioprotection issues. In addition, preliminary results have shown that a relative small daily volume of reprocessed fuel salt is required in the MSFR. For the reference MSFR design, approximate data of the pyrochemical processes, suggests that reprocessed fuel salt volume is between 10 to 40 l/day. While further progress on the knowledge of the chemical reprocessing is needed to reduce these calculations uncertainties, the results underscore the importance of the use of a neutronic-reprocessing coupling tool for the MSFR design.

Neutronics and thermalhydraulic coupling models

Adequate neutronics and thermohydraulics design of the MSFR requires taking into account the phenomena caused by the liquid fuel flow on the reactor neutronics behavior. Some of these phenomena are: the circulation of the delayed neutron precursors, the fuel irradiation which depends on the salt circulation, the coupling between salt temperature distribution and the reactivity feedback, the reactor core wall temperature distribution, etc. Coupled neutronics and thermohydraulics numerical simulations are then usually necessary. Moreover, due to the complexity of the flow distribution in the core, a one dimensional flow model or a traditional subchannel approach are not enough accurate. Instead, a Computational Fluid Dynamics (CFD) approach should be used. In order to take into account for these phenomena, coupled MCNP code and CFD calculations were used to determine the power distribution, salt velocities and temperatures, and the core wall temperature. As mentioned in section 2, the MSFR core cavity

geometry is relatively complicate and have curved inlet and outlet regions that are intended to homogenize the flow velocities and thus minimize potential hot spot in the salt and on the wall surface. CFD calculations are currently being performed to optimize the core geometry.

4. SAFETY STUDIES

Liquid fuel reactors have the advantage of being very flexible in operation but require a safety methodology different from that of solid fuel reactors. Since this new nuclear technology, safety studies are an essential point to be considered all along the R&D studies. Some of the areas where effort has been focused are described in the next paragraphs.

Development of a MSFR safety analysis methodology

The design characteristics of the MSFR have a significant impact on the safety analysis [6]. For example: i) the principle of defense in depth and multiple barriers has to be adapted, ii) the diverse and independence of the reactivity control mechanisms have to be demonstrated, iii) a specific approach on the safety analysis during a severe accident is required since the MSFR fuel is in liquid state at normal conditions, the salt interactions with groundwater have not been studied yet and the source term is not well known, iv) Evaluation of the risk posed by the residual decay heat and the radioactive inventory existing in the reprocessing unit is also needed. Some of the objectives of the MSFR safety analysis methodology are then: i) Developing a MSFR safety analysis methodology based on current safety principles (e.g. defense-in-depth) but adapted to the MSFR particularities, ii) allow reactor designer to estimate impact of design changes and implement adequate safety barriers, iii) help to identify accident initiators and high risk scenarios that require detailed transient analysis, iv) Integrate both deterministic and probabilistic approaches. In order to fulfill these objectives, a MSFR safety methodology is being developed according to the following steps:

- (1) Systemic modeling of all reactor components by using a model-based risk analysis tool,
- (2) Identification the safety functions: this step involves defining the plant operating conditions and the associated design/safety requirements (such as in the ANS/ANSI 18.2). Safety functions can then be identified from the components functional criteria.
- (3) Identification of reactor abnormal events: possible components fail modes and dangerous phenomena (loss of a barrier) are postulated and accident initiators identified.
- (4) Risk evaluation: evaluation of the probability and the severity is performed by using MSFR transient analysis code.

Fraction of Delayed Neutrons

The fraction of delayed neutrons β is an important reactor cinetic parameter that affects the safety performance, and depends on the fissile composition of the reactor and, to a minor extent, on the neutron spectrum. The value of β has been numerically estimated to be about 330 pcm. In the evaluation seven precursor families for the two fissile nuclei, ^{233}U and ^{235}U were considered. It is important to realise when solving the reactor kinetics equations that the concentration of the delayed neutrons precursors depends on the core flow rate. Therefore a variation of the fuel salt flow (e.g. a Loss of Flow Accident) impacts the reactor reactivity although the effect can be easily accommodate thanks to the temperature feedback coefficient.

Decay heat

In order to assess the behavior of the fuel salt after reactor shut down, a model to calculate the decay heat has been developed. Analyses performed with this model concluded that the decay heat in the fuel circuit 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 fuel salt contribute to the salt heating up to 3% of nominal power. An important part of the decay heat (around 1.5% 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. The decay heat model has been used to study various reactor transient behaviors such as the Loss of Heat Sink accidents. The decay heat calculations during reactor transients are the basis for the design of the MSFR draining system. Preliminary calculations show that slow transients are favorable (pump coast-down > 1min) to minimize the temperature increase in the salt. Finally, the impact of the stagnant heating fuel salt on the core and fuel loop systems has to be studied in the future.

Transient studies

Thanks to the MSFR negative reactivity feedback coefficient and the absence of a reactivity reserve (which in a typical PWR is about 10,000 pcm), credible accident scenarios led to a safe reactor shutdown. However, since the reactor design is not yet completed neither the safety analysis methodology, the identification of accident initiators and the subsequent transient analyses can only be considered as being at an early stage. Most of the safety transient studies have been focused up to now on two categories of accidents: i) Reactivity and Power Distribution Anomalies and ii) Decrease in the fuel salt mass flow rate. In the first category, accidents such as a large unaverted injection of the fissile material, the rupture of the fertile blanket tank, a change of core geometry or the effect of the protactinium decay after shutdown have been studied. In the second category (these accidents involves changes on the heat transfer rate and on the precursors concentration), partial and total loss of flow accidents and partial or total blockade of the fuel circuit have been considered. In all cases, the reactor response (e.g. maximum fuel salt temperature) was found to be acceptable under most credible scenarios (such as the reactivity insertion rate, pump coastdown value, the salt draining rates, etc.). An example of such transient analysis is illustrated in Figures 3(d)-(e)-(f).

5. MATERIAL STUDIES

The structural materials retained for MSR were Ni-based alloys with a low concentration of Cr. The composition of the alloy was optimized by ORNL researchers for corrosion resistance, 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. Due to the evolving microstructure of optimized Hastelloy N at higher temperature, it is impossible to preserve the required mechanical properties in the full operating temperature range required for the MSFR system (up to 750-800°C). For the MSFR, the replacement of Mo by W may be beneficial since tungsten diffusion is roughly ten times slower in nickel than molybdenum diffusion. A better creep resistance is then expected with a Ni-W solid solution than with a Ni-Mo solid solution. This would help to reach a 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. In particular, experimental studies on the corrosion of a specific Ni-25W-6Cr (wt.%) alloy has been performed

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 O²⁻ and Fe²⁺ ions) in the salt. Although these preliminary results Ni-W-Cr system seem promising, a wide range of problems still lies ahead in the design of high temperature materials for molten salt reactors. Ni-W-Cr alloy metallurgy and in-service properties need to be further investigated regarding irradiation resistance and industrialization. Additional tests need to be carried-out in order to better understand the W behaviour 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 [7].

6. SALT PROPERTIES

Thermodynamic properties of the salt systems necessary for developments of molten salt reactor designs, reprocessing scheme and simulation codes are being investigated by European partners in the EVOL. These investigations involve the use of numerical tools for the assessment of phase diagrams and the use of experimental facilities to measure physico-chemical properties of actinide fluorides salts. Although significant progress have been made, more efforts are still needed to extend the domain of validity of the measured salt properties (e.g. extending these properties to temperatures prevailing during severe accidents) and to measure other salt properties that are still not well known (e.g. the salt thermal emissivity).

7. SALT REPROCESSING

The on-site salt management of the MSFR combines a salt control unit, an on-line gaseous extraction system and an offline lanthanide extraction component by pyrochemistry. This salt reprocessing scheme is presented in Figure 5 [8,9]. 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 ²³³U) and Neptunium, 90% of Plutonium are extracted by fluorination and immediately reintroduced in the core. The remaining actinides are then extracted through a first reductive extraction 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 progress made in core design in the last years has allowed improving fuel salt reprocessing scheme with a more realistic fuel clean-up rate (between 10 to 40 l/day) and minimized losses to wastes. This value is almost two orders of magnitude less than the reference MSBR scheme and it is one of the advantages of the MSFR concept. Acquisition of fundamental data for the extraction processes is still necessary 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.

8. MOLTEN SALT TECHNOLOGICAL STUDIES

The gaseous extraction system is a continuous salt chemistry process. Helium bubbles are injected at the lower part of the MSFR 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. In order to design the bubbling components at the reactor scale, an experimental project was started, based on the construction of the molten salt loop (Forced Fluoride Flow for Experimental Research project supported by the CNRS/LPSC and Grenoble INP) shown in Figure 6 which is dedicated to bubbling studies and is operated with LiF-NaF-KF salt.

CONCLUSIONS

A general overview of the current status of the Molten Salt Fast Reactor (MSFR) design, the ongoing research activities at the National Centre for Scientific Research (CNRS, France) and the future perspectives of the concept have been presented. Although the design and safety studies are still at an early stage, they shown that the MSFR has significant attractive features including a large negative feedback coefficients, smaller fissile inventory, simplified fuel cycle, etc. These features make this concept very suitable for breeding and for minor actinide burning. No design stopper have been identify in the design and safety studies. However, there are a number of technical areas, discussed in the paper, that require further progress to prove the industrial feasibility of this concept.

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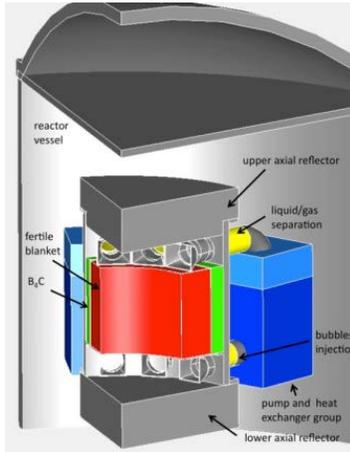


Figure 1. Fast-spectrum MSR (MSFR).

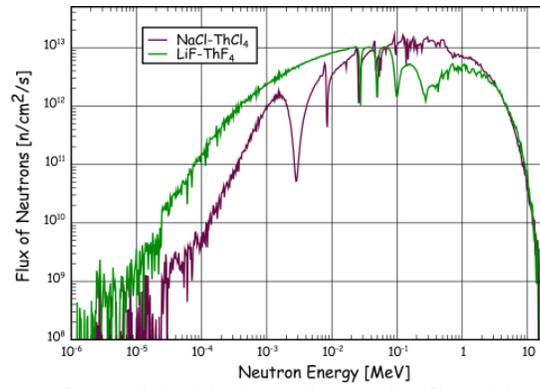
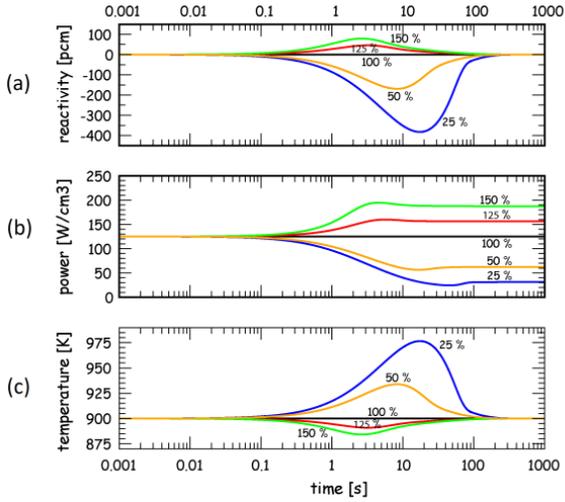
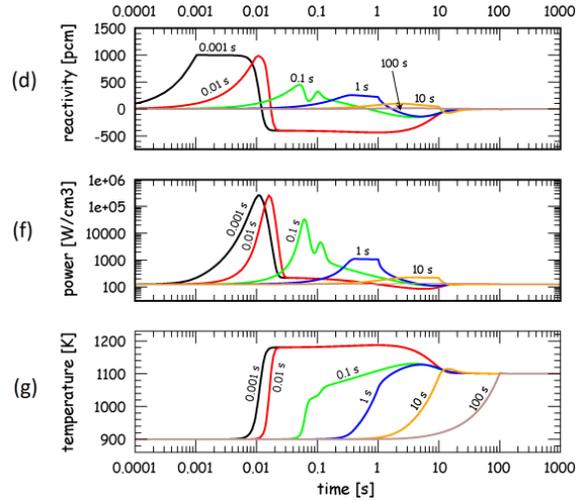


Figure 2. Neutron Spectra for a chloride (purple) and a fluoride (green) salt in a MSFR.



MSFR transient resulting from a variation of the extracted power in the HXs (% of the nominal value).



MSFR transient response as a reactivity insertion of 1000 pcm (insertion rate between in 0.001s and 100s).

Figure 3. MSFR reactivity, power density and the salt mean temperature responses during selected transients (a feedback coefficient of -5 pcm/K was assumed in the simulations).

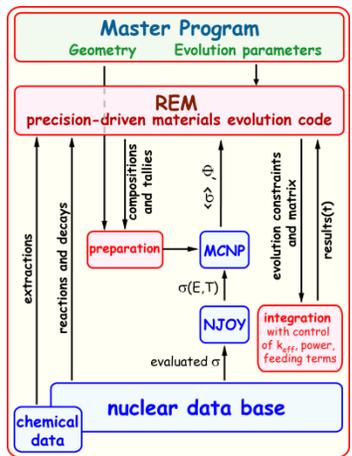


Figure 4. MCNP neutron transport code and the REM fuel evolution code coupling.

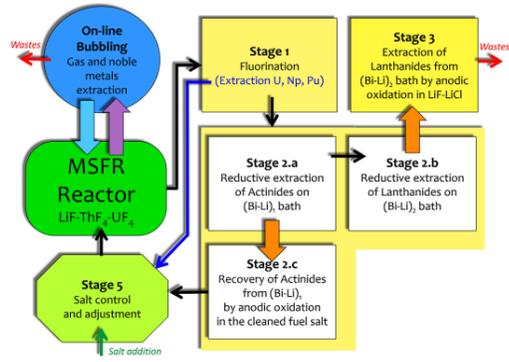


Figure 5. MSFR reprocessing scheme.

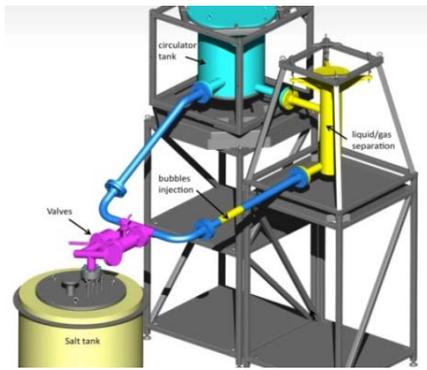


Figure 6. Forced Fluoride Flow for Experimental Research project (FFER).