Chapter X - Molten Salt Fast Reactors

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Abstract

Liquid-fueled reactors exhibit unusual and interesting properties compared to solid-fueled reactors, requesting a revision of some well-known conception and safety rules. Emphasis is thus put in this chapter on such differences and the need for innovative approaches. The Molten Salt Fast Reactor, based on a fast spectrum and seen as a long-term alternative to solid-fueled fast reactors, fulfills the Generation IV criteria and is studied since almost a decade mainly by calculations and determination of basic physical and chemical properties in European Union and Russian Federation. The main characteristics of this concept are presented and discussed including transient simulation, chemistry and material issues, safety analysis, research roadmap and laboratory scale experiments.

Keywords: *Molten salt Reactor, liquid fuel, Thorium fuel cycle, Generation 4, reactor physics, chemistry, materials, fuel cycle, safety*

1. Introduction

MSRs are a family of liquid-fueled fission reactor concepts using a fluid molten salt mixture as fuel. Such liquid-fueled reactors benefit from some potential advantages over solid-fueled systems, among which:

- the possibility of fuel composition (fertile/fissile) adjustment and fuel reprocessing without shutting down the reactor;
- the possibility of overcoming the difficulties of solid fuel fabrication/re-fabrication with large amounts of transuranic elements (TRU);

- the potential for better resource utilization by achieving high fuel burn-ups (with transuranic elements remaining in the liquid fuel to undergo fission or transmutation to a fissile element).

A circulating liquid fuel playing also the role of the coolant presents some more advantages, such as:

- heat production directly in the fuel which is also the coolant (no heat transfer delay);
- fuel homogeneity (no loading plan required);
- rapid, passive, fuel geometry reconfiguration via gravitational draining.

This type of reactor is still at a conceptual level, based on numerical modeling. However, very significant experimental studies were carried out at Oak Ridge National Laboratory (ORNL), in the 1950s and 60s, providing an experimental basis for their feasibility. In 1958 a water-based liquid fuel was used in a 5MWth homogeneous reactor experiment called HRE-2, demonstrating the intrinsic stability of homogeneous reactors. Later on, the Molten Salt Reactor Experiment (MSRE) (ORNL-TM-728, 1965; Haubenreich, 1970), with a liquid fluoride-based fuel at 650°C and a graphite moderated neutron spectrum, operated for four years, from 1966 to 1969, without trouble. It demonstrated the possibility of circulating a liquid fluoride mixture without corrosion problems. This was achieved by using nickel–based alloy (Hastelloy N®) and oxidation control of the fuel by use of the U3+/U4+ buffer. However this 8MWt thermal reactor only tested fissile isotopes (^{233}U, ^{235}U, Pu) and not fertile ones such as Th due to the capture cross sections which are large with thermal neutrons. Nevertheless a continuous physical processing of the fuel was successfully tested, consisting in contacting the fuel with a neutral gas to extract gaseous fission products (FP) such as Kr and Xe before they decay into Rb and Cs (poisons for thermal neutrons). Unexpectedly this processing also removed most of the metallic fission products. Although successful, these tests did not lead to the construction of the Molten Salt Breeder Reactor (MSBR) (Bettis, 1970; Whatley, 1970) studied in details by ORNL, partly because its thermal spectrum requires intensive chemical processing for fission product removal as well as Pa extraction (related to

proliferation issues due to the possible ^{233}Pa decay in pure ^{233}U in such conditions) to avoid neutron captures leading to minor actinides These drawbacks are eliminated by using a fast spectrum.

Within the MSR System Steering Committee (SSC) of the Generation-IV International Forum (GIF/MSR), two fast spectrum MSR concepts are being studied (Serp, 2014), both based on a liquid circulating fuel: the Molten Salt Fast Reactor (MSFR) concept initially developed at CNRS, France and the MOlten Salt Actinide Recycler and Transmuter (MOSART) concept under development in the Russian Federation. Simulation studies and conceptual design activities are on-going in order to verify that fast spectrum MSR systems satisfy the goals of Generation-IV reactors in terms of sustainability (closed fuel cycle, breeder system), non-proliferation (integrated fuel cycle, multi-recycling of actinides), safety (no reactivity reserve, strongly negative feedback coefficient) and waste management (actinide burning capabilities). Compared with solid-fueled reactors, fast MSR systems have lower fissile inventories, no radiation damage constraints on attainable fuel burn-up, no reactivity reserve, strongly negative reactivity coefficients, no requirement to fabricate and handle solid fuel, and a homogeneous isotopic fuel composition in the reactor.

Here, we will focus on the MSFR concept but some elements pertaining to the MOSART concept will be provided. Regarding the MSFR, presented hereafter, its design is not fixed yet but all important issues have been considered since the beginning: nuclear effectiveness, safety, proliferation resistance, in order to reach a design that does not encounter a major obstacle at any level of development. This is why, after the presentation of the physics and chemistry aspects, deployment scenarios and safety issues are discussed. Finally a path for future research is presented.

2. The MSFR Concept

2.1 Core and System Description

Conceptual design activities are currently underway so as to ascertain whether 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 saving (closed Th/U fuel cycle, no uranium enrichment), safety (no reactivity reserve, strongly negative feedback coefficient) and waste management (actinide burner). The calculation results presented here were obtained for a reactor configuration called "reference MSFR" and studied in the frame of the EVOL (Evaluation and Viability of Liquid fuel fast reactor systems) Euratom project of the Framework Program 7 (Brovchenko, 2014a; Dulla, 2014). This is not to be taken as an optimized reactor but as a basis for interdisciplinary studies.

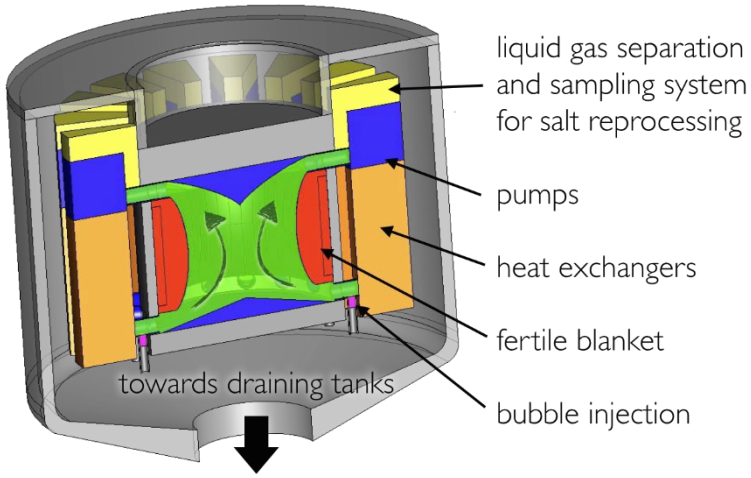

Figure 1. Schematic representation of the reference MSFR fuel circuit

The reference MSFR is a 3GWth reactor with a total fuel salt volume of 18 m^3, operated at a max fuel salt temperature of 750°C (Mathieu, 2009; Merle-Lucotte, 2012). The system includes three circuits: the fuel circuit, the intermediate circuit and the power conversion circuit. The fuel circuit, defined as the circuit containing the fuel salt during power generation, includes the core cavity, the inlet and outlet pipes, a gas injection system, salt-bubble separators, pumps and fuel heat exchangers.

As shown in the sketch of Figure 1, the fuel salt flows from the bottom to the top of the core cavity (note the absence of in core solid matter). In preliminary designs developed 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 salt acting both as fuel and as coolant. Recently, thermal-hydraulic

studies performed in the frame of the EVOL project have shown that a torus shaped core (see Figure 1) improves thermal flow (Laureau, 2013; Rouch, 2014).

The properties of the fuel salt used in these simulations are summarized in Table 2. The fuel salt considered in the simulations is a molten binary fluoride salt with 77.5 mole% of lithium fluoride; the other 22.5 mole% are a mix of heavy nuclei fluorides. This proportion, maintained throughout the reactor evolution, leads to a fast neutron spectrum in the core as shown in Figure 2. This MSFR system thus combines the generic assets of fast neutron reactors (extended resource utilization, waste minimization) and those associated to a liquid-fueled reactor.

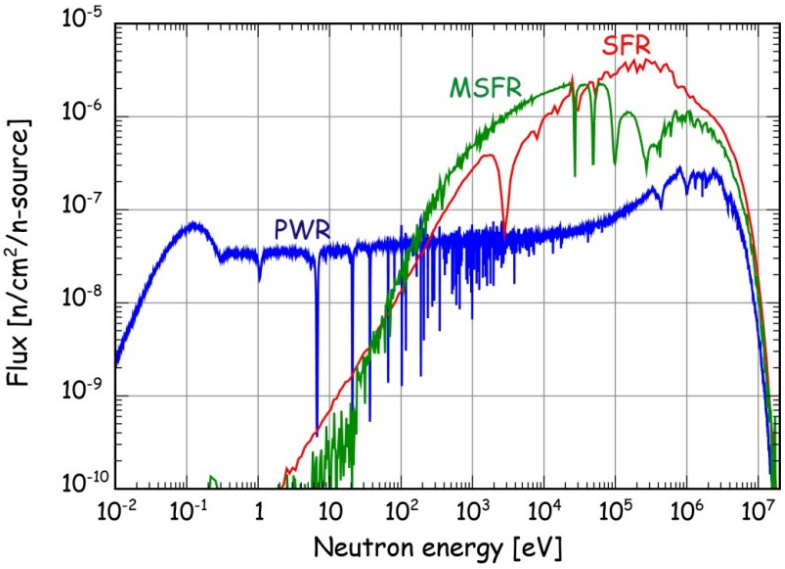

Figure 2. Calculated neutron spectrum of the reference MSFR (green curve). For comparison, a typical sodium-cooled fast neutron reactor spectrum (SFR, in red) and a typical PWR thermal spectrum (in blue) are shown

Both contributions to the feedback coefficient: density coefficient (or void, related to the salt thermal expansion) and Doppler coefficient are largely negative, leading to a total feedback coefficient of -5 pcm/K. This is a significant advantage for both the operation and the safety of the reactor as discussed below. The characteristics of the reference MSFR configuration are summarized in Table 1.

In the fuel circuit, after exiting the core, the fuel salt is fed into 16 groups of pumps and heat exchangers located around the core. The salt traveling time through the whole fuel circuit is 3-4 seconds (Brovchenko, 2012). The total fuel salt volume is distributed half in the core and half in the external portion of the fuel circuit.

The external core structures and the fuel heat exchangers are protected by thick reflectors made of nickel-based alloys, which are designed to absorb more than 99% of the escaping neutron flux. These reflectors are themselves surrounded by a 20cm thick layer of B_4C, which provides protection from the remaining neutrons. The radial reflector includes a fertile blanket (50 cm thick - red area in Figure 1) to increase the breeding ratio. This blanket is filled with a LiF-based fertile salt with initially 22.5mole % $^{232}ThF_4$. Due to the neutron inelastic scattering on Fluorine nuclei (see Figure 2), the MSFR spectrum is a bit less fast than that of solid-fueled fast reactors. This fact, combined to the absence of solid material in the core, results in reduced irradiation damages of the materials surrounding the core.

Table 1. Characteristics of the reference MSFR

Thermal/electric power	3000 MWth / 1300 MWe
Fuel salt temperature rise in the core (°C)	100
Fuel molten salt - Initial composition	LiF-ThF_4-(^{233}U or ^{enr}U)F_4 or LiF-ThF_4-(Pu-MA)F_3 with 77.5 mol% LiF
Fuel salt melting point (°C)	565
Mean fuel salt temperature (°C)	700
Fuel salt density (g/cm^3)	4.1
Fuel salt dilation coefficient (g.cm^{-3}/°C)	8.82 10^{-4}
Fertile blanket salt - Initial composition (mol%)	LiF-ThF_4 (77.5%-22.5%)
Breeding ratio (steady-state)	1.1
Total feedback coefficient (pcm/°C)	-5
Core dimensions (m)	Radius: 1.1275 Height: 2.255
Fuel salt volume (m^3)	18
Total fuel salt cycle in the fuel circuit	3.9 s

The fuel circuit is connected to a salt draining system which can be used for a planned shut down or in case of any incident/accident resulting in an excessive temperature being reached in the core. In such situations the fuel salt geometry can be passively reconfigured by gravity driven draining of the fuel salt

into tanks located under the reactor and where a passive cooling and adequate reactivity margin can be implemented.

The MSFR, as a liquid-fueled reactor, calls for a new definition of its operating procedures. The negative feedback coefficient provides intrinsic reactor stability. The reactor may be driven by the heat extracted, allowing a very promising flexibility for grid load-following for example. Unlike with solid-fueled reactors, the negative feedback coefficient acts very rapidly since the heat is produced directly in the coolant, the fuel salt itself being cooled in the heat exchangers.

Table 2. Physicochemical properties of the fuel salt and of the intermediate fluid, measured for the salt 78%mol LiF-22%mol ThF_4 (Ignatiev, 2012)

	Formula	Value (at 700 °C)	Validity Range [°C]
Density ρ (kg/m^3)	$4094 - 0.882\,(T_{(K)}-1008)$	4125	[617-847]
Kinematic viscosity ν (m²/s)	$5.54\ 10^{-8} \exp\{3689/T_{(K)}\}$	$2.46\ 10^{-6}$	[625-847]
Dynamic viscosity μ (Pa.s)	$\rho_{(g/cm3)}\ 5.54\ 10^{-5} \exp\{3689/T_{(K)}\}$	$10.1\ 10^{-3}$	[625-847]
Thermal conductivity λ (W/m/K)	$0.928 + 8.397\ 10^{-5} * T_{(K)}$	1.0097	[618-847]
Heat capacity C_p (J/kg/K)	$-1.111 + 0.00278\ 10^3\ T_{(K)}$	1594	[595-634][1]

2.2 Transient calculations

The definition and assessment of MSFR operation procedures requires dedicated tools to simulate the reactor's behavior and assess its flexibility during normal (e.g. load-following) or incidental (e.g. pump failure) transients. The reactor modelization requires specific treatments to take into account the phenomena associated to the liquid fuel circulation.

Classical calculation codes can't be employed directly because of the specificity of the core cavity's geometry, and because of the precursor motion. The latter and the MSFR thermal feedback effects imply

[1] The formulas have been extrapolated up to 700°C.

a strong coupling between the neutronics and the thermalhydraulics during reactor transient calculations. Dedicated tools are thus currently being developed. Coupled to a Computational Fluid Dynamics (CFD) calculation code, different neutronics models are used, as detailed below: the Transient Fission Matrix (TFM) approach, the diffusion model, or the direct coupling with a Monte Carlo (MC) approach for reference calculations with a reduced computational time. The use of a CFD code allows the calculation of the 3D velocity and temperature distributions. The latter, along with the density distribution, has a significant impact on the neutronic behavior through the induced variations in the neutron macroscopic cross-sections. Recent studies highlighted the large impact of CFD modeling hypotheses on the MSFR analysis and the need to adopt accurate turbulence models and realistic three-dimensional geometries (Rouch, 2014; Brovchenko, 2014a; Dulla, 2014). In this view, the OpenFOAM multiphysics toolkit allowed an efficient simulation of steady-state and transient cases on detailed, full core, 3D geometries (Jasak, 2007).

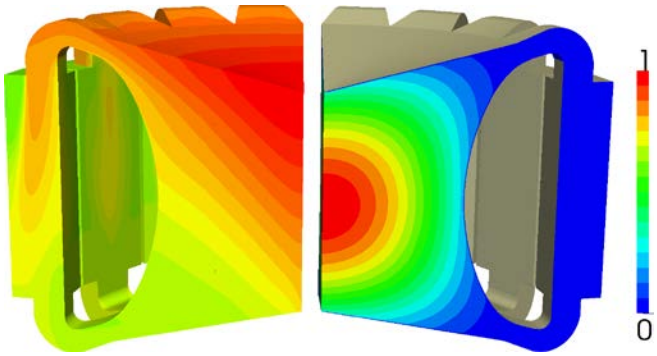

Figure 3. Delayed (left) and prompt (right) neutron sources in the MSFR

The effective delayed neutron fraction (β_{eff}) represents an important reactor kinetics parameter. In circulating-fuel systems, because of the delayed neutron precursors drift, the β_{eff} calculation requires special techniques. The coupled neutronics/CFD simulations represent a necessary step for the accurate calculation of the effective delayed neutron fraction in the MSFR (Aufiero, 2014). Figure 3 shows the distributions of the prompt (right) and delayed (left) neutron sources obtained in OpenFOAM and adopted to calculate β_{eff} in the nominal MSFR conditions.

Some simplified tools were developed for the modeling of the MSFR neutronics among which tools based on the diffusion approximation of the neutron transport equation. Other tools adopted the fine-element, the finite-difference or the finite-volume discretization of the coupled equations of the CFD/neutronics problem. All these tools proved useful as fast-running options, during the initial MSFR design optimization phase, in identifying the specifics of the reactor physics of circulating-fuel systems confronted to thermal feedbacks on the neutronics.

The Transient Fission Matrix (TFM) approach (Laureau, 2015b) has been developed specifically as a neutronic model able to take into account the precursor motion associated phenomena and to perform coupled transient calculations with an accuracy close to that of Monte Carlo calculations for the neutronics while incurring a low computational cost. This approach is based on a pre-calculation of the neutronic reactor response through time prior to the transient calculation. The results of the SERPENT Monte Carlo code (Leppänen, 2013) calculations are condensed in fission matrices, keeping the time information. These fission matrices are interpolated to take into account local Doppler and density thermal feedback effects due to temperature variations in the system. With this approach, an estimation of the neutron flux variation for any temperature and precursor distribution in the reactor can be obtained very quickly.

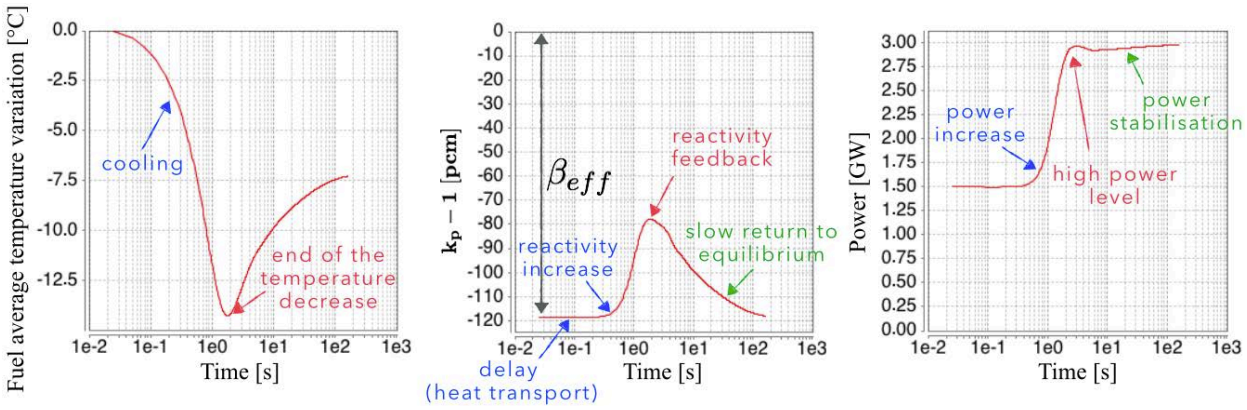

Figure 4. Instantaneous load-following transient of the MSFR from an extracted power of 1.5 GWth to 3 GWth computed with the TFM-OpenFoam coupled code (Laureau, 2015a)

The results obtained with this method applied to an instantaneous load following transient are shown in figures 4 and 5 (Laureau, 2015a). The initial condition corresponds to a critical reactor with 1.5 GWth power. At the beginning of the simulation, the temperature of the intermediate circuit is reduced to increase the power extracted up to 3 GWth. After one second, the feedback effect stops the increase of the neutron population, and the reactivity progressively returns to its initial value with a time constant corresponding to the balancing of the delayed neutron precursor population. An oscillation corresponding to the circulating time of the fuel salt can be observed. This application case highlights the good behavior of the reactor to load following transients.

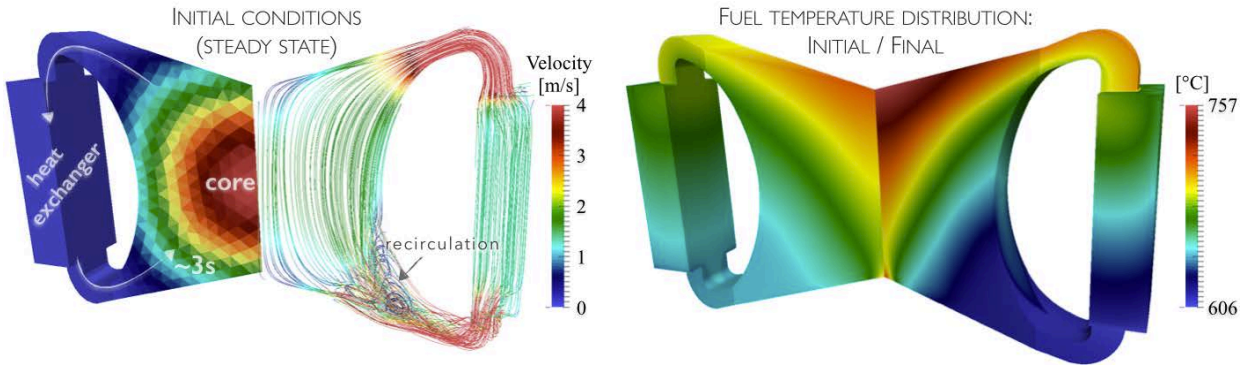

Figure 5. Distribution of power, velocity and temperature in the MSFR (Laureau, 2015a)

3. Fuel salt chemistry and material issues

3.1 Overview of the processing schemes

The fuel salt undergoes two types of treatment: on-line neutral gas bubbling in the core and delayed mini-batch on-site reprocessing (Delpech, 2009). These salt treatments aim at removing most of the fission products without stopping the reactor and thus securing a rather small fissile inventory outside the core compared to present day LWRs. The reprocessing rate itself is assumed equivalent to the present LWR rate, although it could be possible to reprocess the fuel salt every ten years but to the detriment of economical yield.

The salt treatment is schematically presented in Figure 6. It consists in two circuits. One is a continuous gas bubbling in the core to extract the gaseous fission products (FP) and the metallic particles present in the salt (metallic FP and corrosion products). The gaseous stream is sent to a provisional storage where most of the Kr and Xe decay into Rb and Cs, preventing their accumulation in the fuel salt. The remaining gas is recycled.

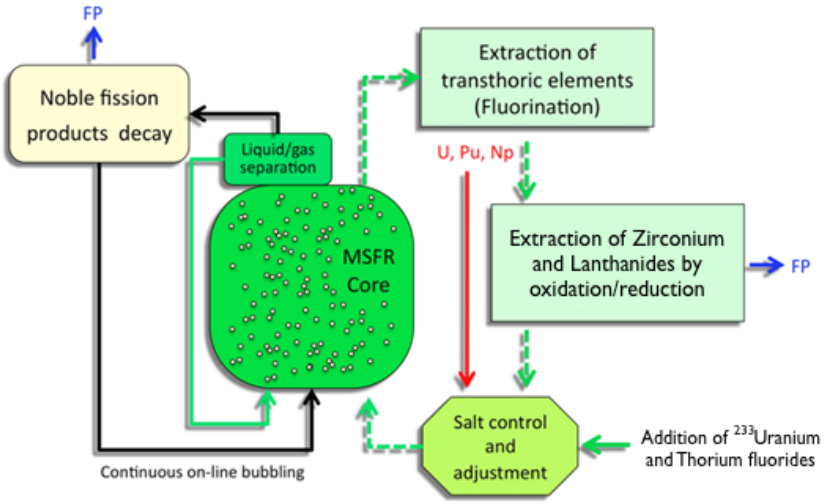

Figure 6. Schematic representation of the fuel salt treatment with two loops. On the left is the on-line treatment with gas bubbling in the core to extract noble gases and metallic particles (Fission Products). On the right is the mini-batch on-site reprocessing with two objectives: removing FP (Zr, Ln) and adjusting the fuel content in fissile and fertile isotopes.

The other is a semi-continuous salt reprocessing at a rate of about 10 liters per day, in order to limit the lanthanide and Zr concentration in the fuel salt. The salt sample is returned to the reactor after purification and after addition of ^{233}U and Th as needed to adjust the fuel composition. This is also an opportunity to tune the oxidoreduction potential of the salt by controlling the U^{4+} to U^{3+} ratio.

These two processes are aimed at keeping the liquid fuel salt in an efficient physical and chemical state for long time periods (decades). The gas bubbling has two objectives: removing metallic particles by capillarity (floating) and extracting gaseous fission product before their decay in the salt. The

pyrochemical salt batch reprocessing avoids the accumulation in the fuel salt of large quantities of lanthanides and zirconium that could be detrimental to several properties such as Pu solubility or salt volatility. Conversely to the thermal molten salt reactor, none of these processes are vital to the fast reactor operation. If they were interrupted for months or years the MSFR would not stop but would have a poorer breeding ratio and could suffer from partial clogging of the heat exchangers, leading to poorer efficiency. The effect of the batch pyro processing rate is shown in Figure 7. Note that with the reactor configuration used for the calculation, the core is under-breeder. The addition of a fertile blanket secures breeding, up to a reprocessing time of the total fuel salt volume as large as 4000 days.

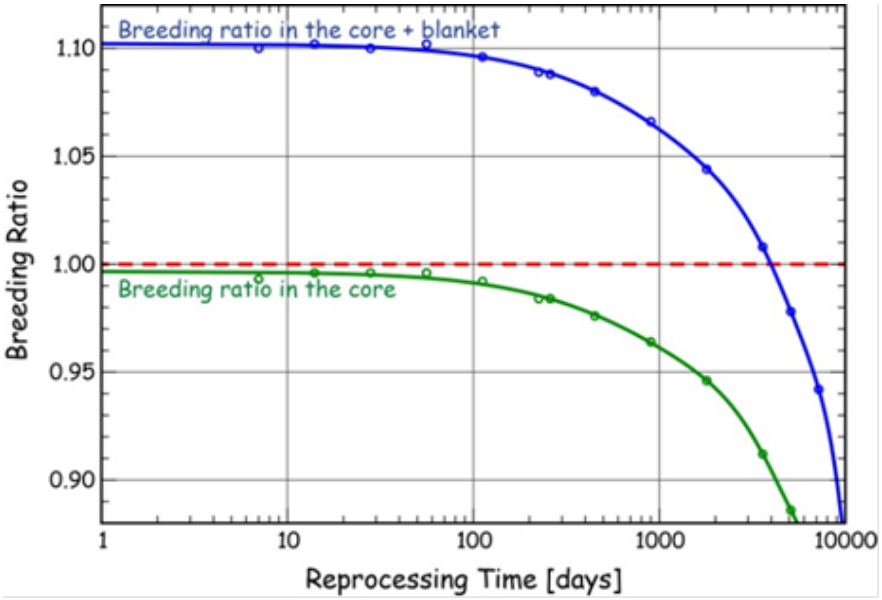

Figure 7. Influence of the batch reprocessing rate on the breeding ratio in the core and in the whole MSFR system (core + fertile blanket)

3.2 Impact of the salt composition on the corrosion of the structural materials

Material corrosion in molten salt nuclear reactors results from the evolution of the salt composition during operation: production of HF by an uncontrolled purification process or by hydrolysis reactions, production of corrosive fission products or mass transfer in thermal gradients. Ni-based alloys have been recognized the most suitable materials for their mechanical and chemical resistance up to about 700°C in the presence of fluoride salts. Graphite presents an excellent compatibility with molten fluorides but

cannot be used for structural applications submitted to a neutron flux. Silicon carbide has a good irradiation and very high temperature resistance and might be an acceptable solution for corrosion. However assembling SiC parts is not usual technology and its long term chemical behavior has not yet been tested in molten fluorides.

The historical tests carried out at ORNL have shown that a chemical potential control of the salt was necessary to prevent two types of corrosion: Cr oxidation and intergranular corrosion by Te (a fission product). This was achieved by using a chemical buffer based on the U^{4+}/U^{3+} couple. The proper U^{4+}/U^{3+} concentration ratio was obtained by contacting the salt with metallic Be from time to time to keep this ratio in a suitable range (60 to 20 for instance). The change of chemical potential of the fuel salt is intrinsic to the fission of fissile elements present in the fuel at valence IV, because the resulting fission products have a mean valence close to III. Therefore the salt becomes more oxidizing as fissions occur; an initial chemical potential control of the salt is necessary but never sufficient to prevent corrosion. It has been shown that chromium is necessary to the mechanical properties of Ni-based alloys and not only to their chemical resistance to oxidation in air. However its concentration should be limited to about 6 to 8 wt% to keep the corrosion rate at an acceptable level.

Prior to the use of the U^{4+}/U^{3+} chemical buffer a salt purification is required for the initial salt preparation or when recycling the actinides after lanthanide extraction. H_2O and HF are the most oxidizing compounds present as impurities in solid fluorides and in the molten salt. High oxidation state, H_2O and dissolved oxides can be eliminated by using gaseous H_2/HF mixtures but some HF may remain dissolved in the salt. Care should be taken to limit this dissolved amount. For a salt not containing Be ions the ultimate reduction can be achieved by addition of U^{3+} when recycling U into the fuel salt, or by reduction with metallic Th (Th should be added anyway, to compensate for neutron captures).

4. MSFR fuel cycle scenarios

To produce power, a fission nuclear reactor requires fissile material. Generation 2 or 3 reactors (PWR, CANDU, EPR…) being under-breeder systems, i.e. using more fissile material than they produce, need to be regularly re-fueled with fissile material all along their operation time. On the contrary, breeder generation-4 reactors (SFR, MSFR, GFR…) require only one (or two in the case of solid fuel reactors) initial fissile material load. They then produce at least the fissile material they need to be operated during their entire lifespan. Molten salt reactors require only one fissile load since no fuel re-fabrication is necessary and the fuel salt composition is controlled on-line without stopping reactor operation whereas 2 loads are necessary for solid-fueled reactors with one fissile load inside the reactor and the other in the reprocessing/fuel manufacturing process.

According to our simulations results, the Thorium based MSFR can be started with a variety of initial fissile loads as discussed below (Heuer, 2014; Merle-Lucotte, 2009):

- With U235 enriched uranium - the only natural fissile material available on earth is ^{235}U (0.72% of natural uranium). Enriched uranium can be used directly as initial fissile material to start MSFRs, with an enrichment ratio less than 20% due to proliferation resistance issues.
- With ^{233}U directly as initial fissile material, assuming that this ^{233}U can be produced in fertile blankets of other reactors (SFR…) or by irradiating ^{232}Th in an Accelerator Driven System (ADS) for example. Once an initial park of MSFRs based on the Th-^{233}U cycle is launched, ^{233}U will also be produced in breeder MSFR reactors, allowing the deployment of such ^{233}U-started MSFRs in a second phase even if no ^{233}U is produced elsewhere.
- With the plutonium produced in current PWRs or in future EPRs or, even better, the mix of TRU produced by these Generation 2-3 reactors as initial fissile load.
- With a combination of the previous starting modes. For example, ^{233}U may be produced by using special devices containing Thorium and Pu-Mox in current PWRs or in future EPRs.

- Figure 8 presents two examples of fuel composition evolutions for a "3GWth reference MSFR" reactor started with ^{233}U or TRU. An optimized fuel salt initially composed of LiF-ThF4-$^{enriched}UF4$-$(TRU)F_3$ with uranium enriched at 13% in ^{235}U and a TRU proportion of 3% (see Figure 9), has been selected in the frame of the EVOL project taking into consideration the neutronics, chemistry and material issues.

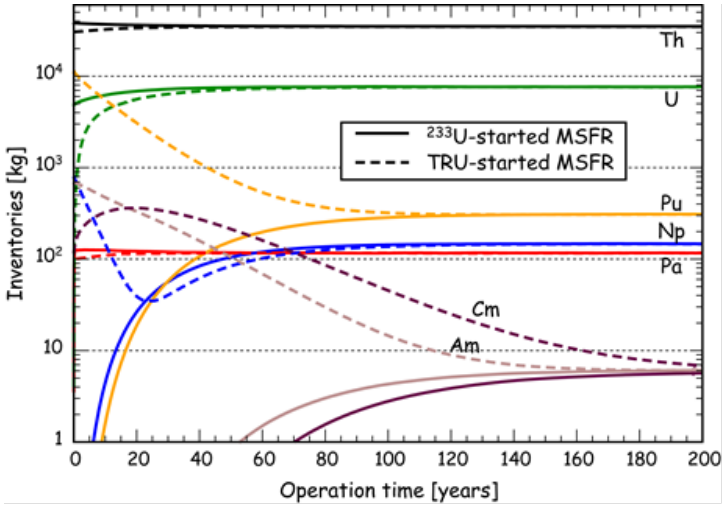

Figure 8. Time evolution up to equilibrium of the heavy nuclei inventory for the ^{233}U-started MSFR (solid lines) and for the TRU-started MSFR (dashed lines) - Operation time is given in Equivalent Full Power Years (EFPY)

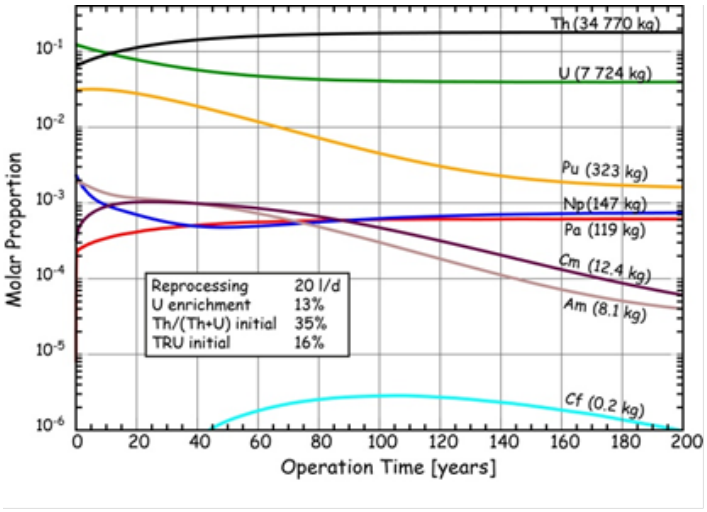

Figure 9 . Time evolution up to equilibrium of the heavy nuclei inventory for the optimized MSFR configuration started with enriched Uranium and TRU elements. Operation time is given in Equivalent Full Power Years (EFPY)

Given the absence of naturally available ^{233}U, a standing question is whether a park of MSFRs can be deployed whether at the French national, the European or the worldwide scales. In this section, we illustrate the flexibility of the concept in terms of deployment and end-of-game capacities of the MSFR at the French national scale.

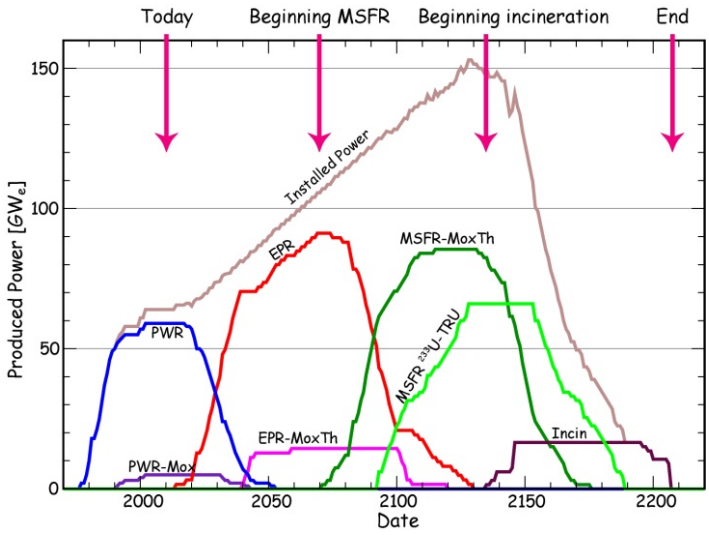

Figure 10. French nuclear power deployment exercise based on PWRs, EPRs and MSFRs

The deployment scenarios of a park of nuclear reactors also led to an estimation of the amount of heavy nuclei produced by such a deployment. We aim at evaluating the complexity of the management of these heavy nuclei stockpiles, as well as their radio-toxicity. The French scenario, displayed on Figure 10, assumes that the natural uranium resources available are large enough to require Generation 4 reactors in 2070 only. The deployment scenario starts with the historical French nuclear deployment based on light water reactors (PWRs followed by EPRs). By 2040, some Generation 3 reactors are fueled with Pu-Uox in a Thorium matrix both to reduce minor actinide production and to prepare the launching of the Thorium fuel cycle in MSFRs. The park of these Generation 3 reactors is then progressively replaced with MSFRs started with this Th-Pu Mox fuel from the last Generation 3 reactors. The deployment is finally completed with MSFRs started with a mix of ^{233}U produced in the existing MSFRs and the remaining stockpiles of Pu-Uox and Pu-Mox irradiated in the light water reactors.

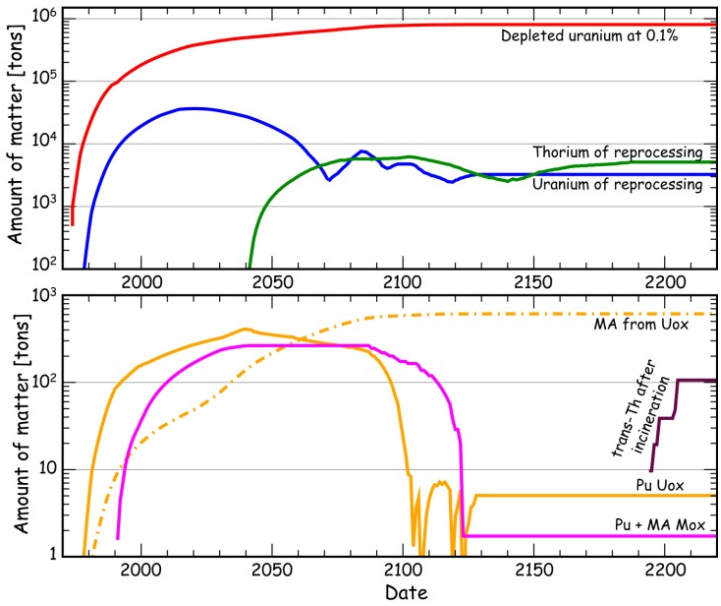

Figure 11. Evolution of the actinide stockpiles during the scenario considered

Assuming that, at any time in the future, here in the first half of the XXIInd century, France resolves to dispense from the production of fission based nuclear energy, the scenario ends with the introduction of burners with a view to optimizing the end-of-game and further reducing the final TRU inventories after MSFR shutdown. Note that the end-of-game situation would not be different if it occurred after hundreds of years of operation; it depends only on the installed power.

The evolution of the radioactive element stockpiles other than the fission products during the scenario is shown in Figure 11. The final stockpiles that will have to be managed as the scenario ends are the following:

- Depleted uranium at 0.1%: 803 700 t
- Uranium from reprocessing (minimized by the scenario management): 3 250 t
- Irradiated Thorium: 5 100 t
- Irradiated Uox fuel (minimized by the scenario management) represented in Figure 10 by its Pu content (labeled 'Pu-Uox'): 5 t of Pu standing for 450 t of irradiated Uox

- Irradiated Mox fuel (minimized by the scenario management) represented in Figure 10 by its Pu content (labeled 'Pu+MA Mox'): 0.76 t standing for 12.4 t of irradiated Mox
- Minor actinides separated from the Pu when the latter is used as Mox fuel in light water reactors, and vitrified (labeled 'MA from Uox'): 612 t
- Final burner inventories: 106 t

The evolution of the radiotoxicity corresponding to the final radioactive stockpiles of this scenario including the fission products is displayed in Figure 12, where it appears that the short-term radiotoxicity (a few dozen years) is dominated by the FP while the long-term radiotoxicity (10^3 to 10^6 years) is mainly due to the vitrified minor actinides produced in light water reactors and not re-used in Mox fuel.

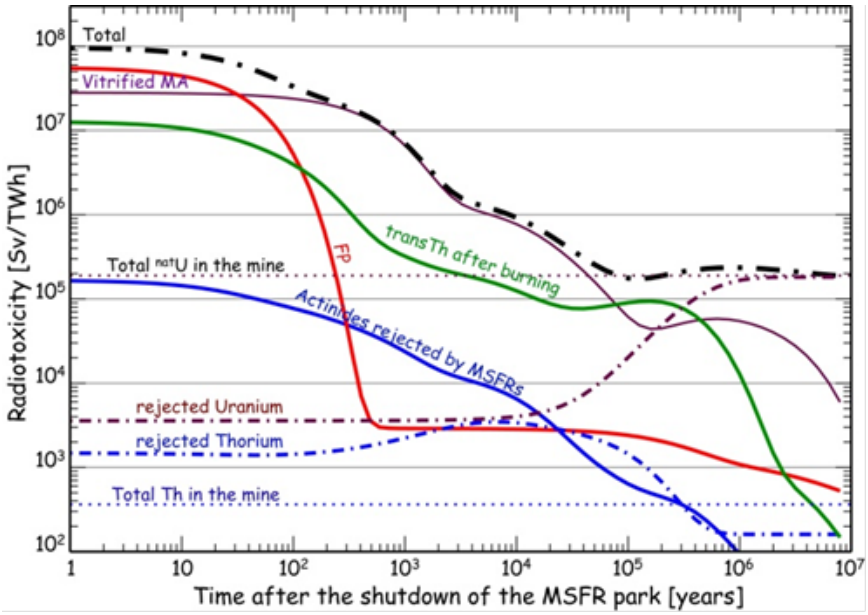

Figure 12. Time evolution of the various contributions to the radiotoxicity of the final radioactive stockpiles

5. Safety Issues

In the frame of the EVOL Euratom project in collaboration with Russian research organizations cooperating in the ROSATOM project MARS (Minor Actinides Recycling in Molten Salt) (Ignatiev, 2012),

design and safety studies of the Molten Salt Fast Reactor (MSFR) system have been led (Brovchenko, 2014b).

A molten salt reactor has some specific safety features because the fuel salt geometry can be modified quickly and passively by draining to subcritical tanks. It is possible to design the system with a maximum of passive devices to cool the fuel in all circumstances and for long times without human intervention. Moreover, the MSFR reactor stability is enhanced by its largely negative feedback coefficients. Some of these features are discussed below but not all safety provisions are detailed.

5.1 Safety Approach and Risk Analysis

The unique characteristics of a liquid-fueled reactor strongly impact its design and safety analyses. For example:

- The safety principle of defense-in-depth and multiple barriers must be re-adapted since conventional barriers (such as clad, primary circuit and containment in LWRs) are no longer applicable.
- The diversity and mutual independence of the MSFR reactivity control mechanisms have to be demonstrated (no control or shutdown rods or burnable poisons…).
- New safety criteria to evaluate reactor response during normal, incidental and accidental conditions are needed since the MSFR fuel is in liquid state - which is not an acceptable situation for the LWR fuel.
- In the evaluation of severe accident scenarios with leakage to the environment, any interactions between the fuel salt and groundwater should be investigated in detail and the source term be determined.
- The risk posed by the residual decay heat and the radioactive inventory in the reprocessing unit must also be evaluated.

A novel methodology for the design and safety evaluations of the MSFR is needed. Nevertheless, it would be desirable that the MSFR methodology rely on current accepted safety principles such as the principle of defense-in-depth, the use of multiple barriers and the three basic safety functions: reactivity control, fuel cooling and radioactive product confinement. In addition, due to the limited amount of operation experience and some of its novel features, any new methodology shall be robust and comprehensive, and integrate both deterministic and probabilistic approaches. In order to fulfill these objectives, a MSFR design and safety analysis methodology is currently being developed (Brovchenko, 2013a) according to the following steps:

1. Systemic modeling of all reactor components using a model-based risk analysis tool.
2. Identification of the safety functions, to be defined from the components' functional criteria.
3. Identification of reactor abnormal events (failure modes and dangerous phenomena)
4. Risk evaluation: evaluation of the probability and the severity of events.

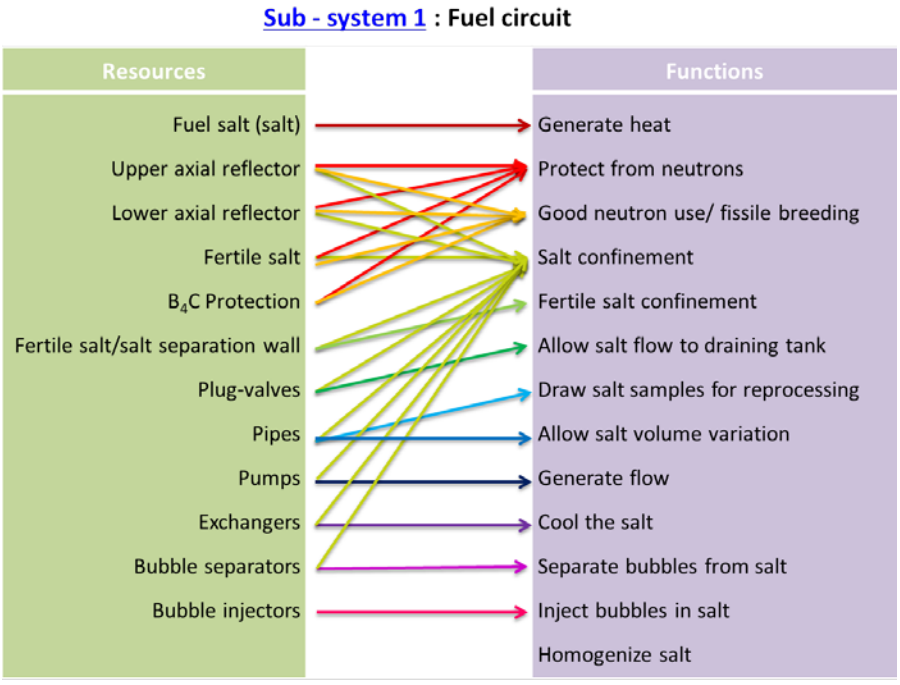

Figure 13. Resources and functions of the fuel circuit sub-systems, the correspondence of resources to functions are shown by arrows that are color coded to improve the legibility of the graph

The design and safety criteria should ensure that all the reactor components adequately perform the safety functions in order to meet the requirements defined for each plant operating condition. The MSFR development being at its early stages, the idea is to adopt an inherent safety-by-design approach.

Figure 13 gives a preliminary view of a systematic description of the MSFR fuel circuit in terms of components and safety functions.

5.2 Liquid-fueled Reactor Specificities

The design characteristics of the MSFR have been evaluated regarding safety issues. An example has been chosen here to illustrate this approach: one of the assets of the liquid-fueled MSFR systems is the homogeneity of the fuel. In a general way, this type of reactor can be placed in a category with all the reactors that run with a circulating fluid fuel (whether gaseous or liquid). These are referred to as homogeneous reactors. Since the 1960s, it has been shown that, in the case of homogeneous reactors without reactivity reserve, control rods are not necessary to control reactor operation (Briggs et al., 1955). The MSFR, which is self-controlled thanks to its negative temperature feedback coefficients and the absence of in-core reactivity reserve fits in this category and, consequently, control or safety rods are not included in the design being considered. Contrary to a PWR, it does not require neutron flux shape control since the fuel is permanently homogenized and the coolant, here the fuel salt itself, can undergo large temperature increases (100°C to 200°C) with no risk of a boiling crisis susceptible to threaten the integrity of the cladding.

The three barriers traditionally used in the defense-in-depth approach were defined in the specific frame of the PWR reactor development or, more generally, in the frame of solid-fueled reactors. Like other safety notions, the transposition of the confinement barriers first mandates more general consideration of the origin and application of this concept. Eventually, these barriers will have to be redefined according to their usefulness for each reactor design rather than seeking an equivalence with PWRs. An extensive study adapted to the sequence of potential accidental events will have to determine or

confirm the number of c necessary onfinement barriers in the case of the MSFR as well as their configuration. However, as a first step and as a pedagogical illustration describing the overall facility, the 3 fuel salt confinement barriers in the MSFR can be identified by analogy with PWRs as shown in Figure 14:

- Pink: the fuel circuit (heat exchangers, pumps, ...) and the draining system (tanks and pipes) totally within the fuel casing;
- Light blue: the reactor vessel, the intermediate circuit and the draining system's water circuit;
- Grey: the reactor containment structure (the building) and the emergency cooling chimney, not shown on the drawing.

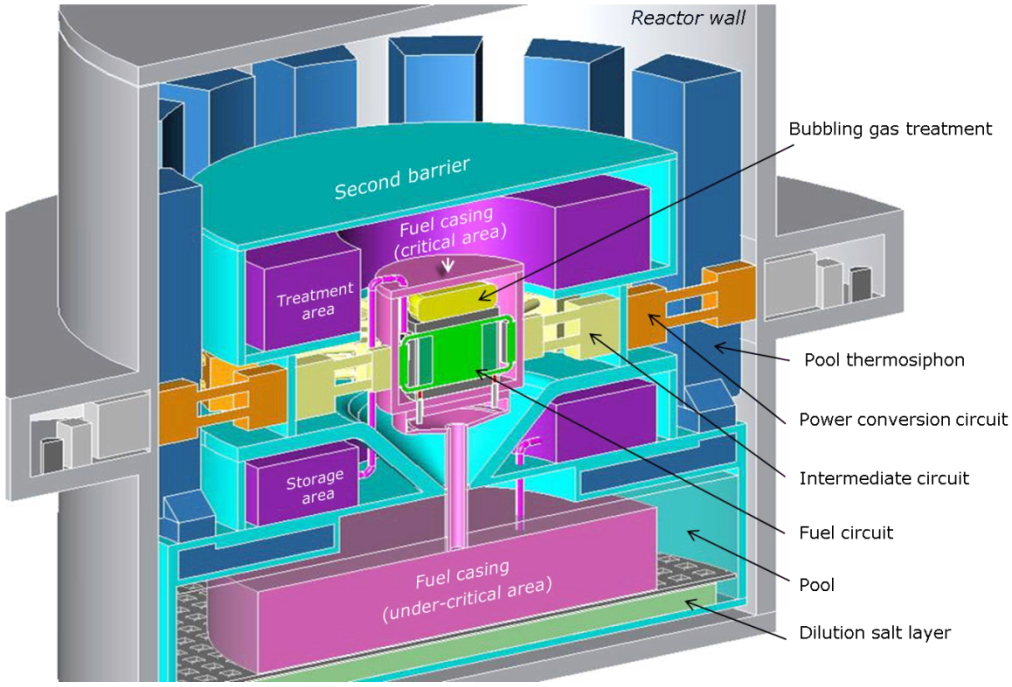

Figure 14. Illustration of the main functions associated with the MSFR operation. In the middle is the green fuel salt circuit surrounded by a pink envelope representing the first confinement barrier. The cyan envelope represents the second barrier including storing and chemical salt processing units in violet. The third barrier is in gray. Two heat transfer circuits between the three barriers are represented as loops in yellow and orange.

The first barrier (pink) includes three zones. The upper zone contains the fuel circuit (green) and the neutral gas reprocessing (yellow). A collector for salt draining is represented (funnel and vertical tube)

leading the drained salts to containers with sub critical geometry (not detailed) situated in a large water pool. This large water pool acts as a thermal buffer in case of high temperature emergency draining. At the bottom of this pool is located a layer containing a dilution salt that can passively mix with the fuel salt in case of a large first barrier failure. This can provide neutron poisons to the fuel and create a large salt-wall interface for passive cooling in the event of a severe accident. Heat pipes (dark blue) are used to transfer the decay heat to the atmosphere. This means that decay heat can be removed into the atmosphere in case of a heat sink failure.

Other systems which also contain radioactive materials have to be studied, in particular the fertile blanket salt system including the storage and processing of the associated gases, as well as all the related inter-system transfers.

As a brief conclusion to this paragraph, let us recall that the global safety objectives are fully transposable to the MSFR reactor. The difficulty lies, among other things, in the identification of severe accidents for this type of reactor. Thus, a core melt in the case of solid-fueled reactors is central to present safety studies and has no immediate equivalent in a liquid-fueled reactor. A safety analysis for the MSFR must then proceed from the fundamentals of nuclear safety.

5.3 Decay Heat Removal

The decay heat generation is represented versus time in Figure 15. The MSFR design implies that fission products are present in two different places when the reactor is stopped. Some are in the liquid fuel salt and some in the gas processing unit. About 1/3rd of the heat is produced in the gas processing unit and 2/3rd in the liquid fuel. The power of both heat sources decreases rapidly (by a factor 10 in about one day) from the value at shut down, which depends on the history of power generation. The total amount of power at shut down is about 5% of the nominal power. This value is lower compared to solid fuel reactors because fission products are continuously removed in this concept.

In case of cooling problems the fuel salt and the fluid containing fission products (salt or metal) of the gas processing unit can be drained into a subcritical tank located in a water pool. A large amount of water is used as a decay heat thermal buffer so as to reduce the heat to cold sink transfer rate need by a factor ten, for instance. This heat transfer is achieved by passive thermos siphons or heat pipes to the atmosphere through the reactor building walls (the third barrier). If unattended for a very long time, the fuel salt will solidify.

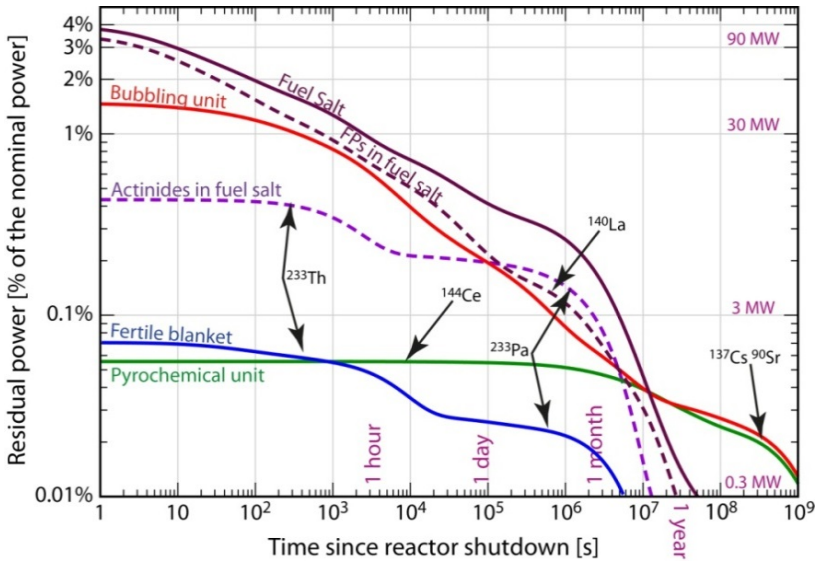

Figure 15. Residual heat in the different radioactive fluids of the MSFR, after the total fission shutdown of the reactor previously in steady-state [Brovchenko, 2012; Brovchenko, 2013b]

5.4 Preliminary accidental transient identification

- A direct transposition to liquid-fueled reactors of the traditionally identified accidents of solid-fueled reactors is not possible. In a liquid-fueled reactor, the fuel is also the coolant so that a LOCA (Loss Of Coolant Accident) implies the simultaneous loss of the fuel and of the coolant. We can study these initiators by equating the primary circuit coolant to the liquid fuel while keeping in mind that the phenomena related to the accidents will not necessarily be comparable to those of a solid-fueled reactor. Another interpretation could identify the MSFR's intermediate circuit with a solid-fueled reactor's primary circuit. In order to retain more clarity, we prefer to re-define the accident types as outlined below for the fuel circuit:

- LOF - Loss Of Flow: In the fuel circuit Loss Of Flow accident, we gather all the accidents that are not associated to a slowing down or stalling of the intermediate fluid circulation and are not due to a loss of fuel.
- LOH - Loss Of Heat sink: In a Loss Of Heat sink accident, the fuel salt circulation continues unchanged but its cooling is no longer ensured.
- TLOP - Total Loss Of Power: In the event of on-site Total Loss Of Power all the pumps are stalled in the fuel, intermediate and conversion circuits; all active systems connected to the power supply are assumed non-operational; in this type of accident, the on site security power supply is considered deficient as well.
- TOP - Transient Over-Power or OVC - OVer-Cooling: An OVer-Cooling accident increases the reactivity and, as a consequence, the power generated because the reactor's thermal feedback coefficient is negative.
- LOLF - Loss Of Liquid Fuel: In the Loss Of Liquid Fuel accident, we consider a significant leak of the fuel salt outside the fuel circuit. RAA - Reactivity Anomalies Accident: Since the reactivity reserve is very small in the MSFR, reactivity related accidents have to do with reactivity anomalies rather than accidents of the TOP type (control bar ejection). In fact, reactivity variations incurred in this reactor are much smaller than they are in a PWR.

This preliminary list of accidents results from the application of the general safety assessment methodology mentioned above and currently under development for liquid-fuel reactors. The next steps for this safety evaluation will take place under the framework of the Horizon2020 European Commission project SAMOFAR (Safety Assessment of Molten Salt Fast Reactors) starting in the second half of 2015 up to 2019.

6. Concept Viability: Issues and Demonstration Steps

6.1 Identified limits

Although the MSFR is still at the preconception design stage, several limiting factors can be identified in the development of the concept.

The first, obvious, issue is materials resistance to high temperatures under irradiation, if the reactor is to be operated with a reasonably high power density. A first temperature limit is given by the fuel salt melting point (565°C) to which a safety margin should be added to avoid local solidification (50°C for instance). To this, add 100 to 150°C for in core temperature heating corresponding to a salt circulation period of 3-4 seconds, so as to satisfy heat transfer dynamics in the heat exchangers without incurring an excessive pressure drop within these. This leads to a temperature of about 750°C at the core outlet to the gas-salt separation device and the pump (hot leg). Those devices may be maintained at 700°C by cooling i.e. the same temperature as the heat exchanger plates during the heat transfer, the intermediate coolant salt being at about 650°C. It seems that there are today alloys that can withstand such temperatures for a long time but this could be a limit, unless the material is replaced regularly as is done with solid fuel cladding.

The second issue arises in the attempt to limit the per GW fissile inventory. This implies restricting as much as possible the proportion of fuel salt out of the core, in the tubing, pumps and heat exchangers. One of the main constraints on the design of the MSFR fuel circuit is the ability to evacuate the heat generated while restraining the fuel salt volume mobilized for that task. It seems technically challenging to reduce this “useless” amount of salt to less than 50% of the total load and 30% appears as a limit.

The third issue is a question more than it is a real limit: the safety evaluation. Indeed, as discussed above, today's safety evaluation techniques apply to solid fuel water reactors but are partly irrelevant for liquid

fuel reactors. A new way of tackling the problem should find a consensus before any national safety authority can approve of a liquid fuel reactor design and this will take time and resources.

From the parametric studies that were carried out on the MSFR the concept does not exhibit any major stumbling blocks and the various limits can all be circumvented by reducing the power density.

6.2 Progression in safety demonstration and design optimization

It is possible to design a low power demonstration reactor in which to test all the features expected for a full size "Reference MSFR" with a single fuel loop, as shown in Figure 16. Its fissile inventory lies in the range of 400 to 500kg of ^{233}U for a zero power version and up to 670kg for a 200 MWth version.

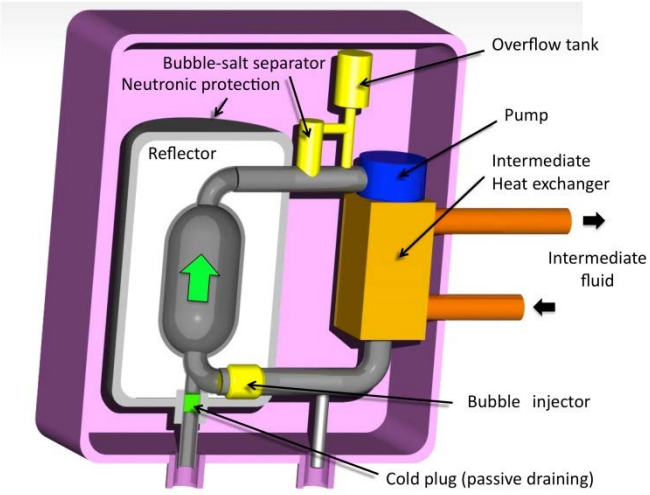

Figure 16. Sketch of a liquid fuel single loop reactor for demonstration purposes or modular conception. The fuel volume (1.8 m^3) is reduced by a factor 10 from the 3GWth reactor and the power (200MWth) by a factor 15 to use the same intermediate heat exchanger.

The size of the reactor liquid fuel loop is not a limit as shown by the calculation of a single loop 200MW reactor instead of a 16 loop 3GWth reactor. The low power demonstration version (Merle-Lucotte, 2013) is sketched in Figure 16 could be replaced by a regenerator version if the reflectors were replaced by a blanket. The size of this fuel loop assembly is about 2.5m in diameter and 3m high (core: 1.1m diameter

and 1.1m high). The power is limited by the intermediate exchanger size which is assumed to be the same as that of the 3GWth reactor.

Before reaching this advanced level it will be necessary to bring evidences of safety for all experiments involving nuclear materials, under the supervision of nuclear safety agencies. To get the clearance of these authorities the reliability and safety of the technical solutions involved should be demonstrated before on pieces of equipment operating with non-nuclear materials (Simulant salts or chemicals). Therefore the following simplified scheme is foreseen:

- basic data determination and assessment (It is the present stage up to about 2020);
- technical devices testing on non-nuclear simulants up to the full scale;
- chemical separation tests on nuclear materials at small laboratory scale and by remote handling;
- development of numerical simulation tools validated on experimental equipment using circulating simulant salts at high temperature.

Obviously all the stages mentioned above will overlap in time, not only for practical reasons but because all the aspects of the design should be kept in mind and documented during the whole development procedure. According to present international standards, safety and proliferation resistance should be analyzed from the beginning of the conception in order to be inherent in the design and not “added-after”.

6.3 Presently Ongoing Laboratory Scale Experiments

Several experimental set-ups are being operated at LPSC-Grenoble-France to acquire some technical experience on the handling and processing of molten salts..

One piece of equipment is called FFFER for Forced Fluoride Flow for Experimental Research. It is a 70 liter FLiNaK loop with a liquid salt circulating rate of about 2 liters/second at 600°C. This reduced scale loop

aims at studying gas injection and separation for the continuous extraction of gaseous and metallic fission products in the MSFR fuel salt. At present only the gas injection and the hydrocyclone efficiency for bubble-salt separation are being studied, but important technical devices are tested in the process.

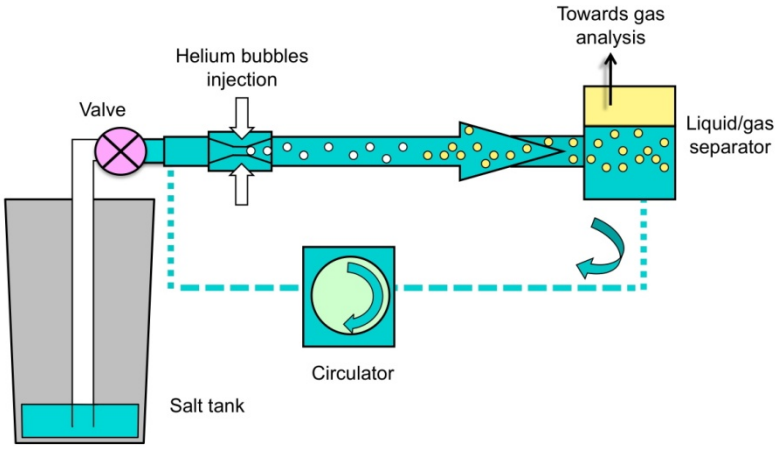

Figure 17. Scheme of the FFFER loop

The FFFER loop comprises a tank where the FLiNaK load is prepared before the experiment and stored after. The circulating loop is situated above this tank and is filled with liquid salt only for the duration of the experiment. It is isolated from the tank by two valves in parallel: a mechanical ball valve and a “freeze plug”. In case of electrical shut-down the “freeze plug” melts within a few minutes and the salt goes back in the insulated storage tank where its solidification may take place without any disturbing effects. The main elements of this equipment are shown in Figures 17 and 18: the melting tank is in grey, the valves are in pink, the light blue tank contains the circulation pump and the yellow one the hydrocyclone for bubble/salt separation. The building material is 304 and 316L steel for all the parts. The 55mm inner diameter pipes (mean velocity 1m/s) are fitted with a Venturi gas injector and an ultrasonic salt velocimeter. The salt level in the three tanks (melting, separation, pump) is measured and regulated by probes and the corresponding gas pressures are controlled according to experimental need.

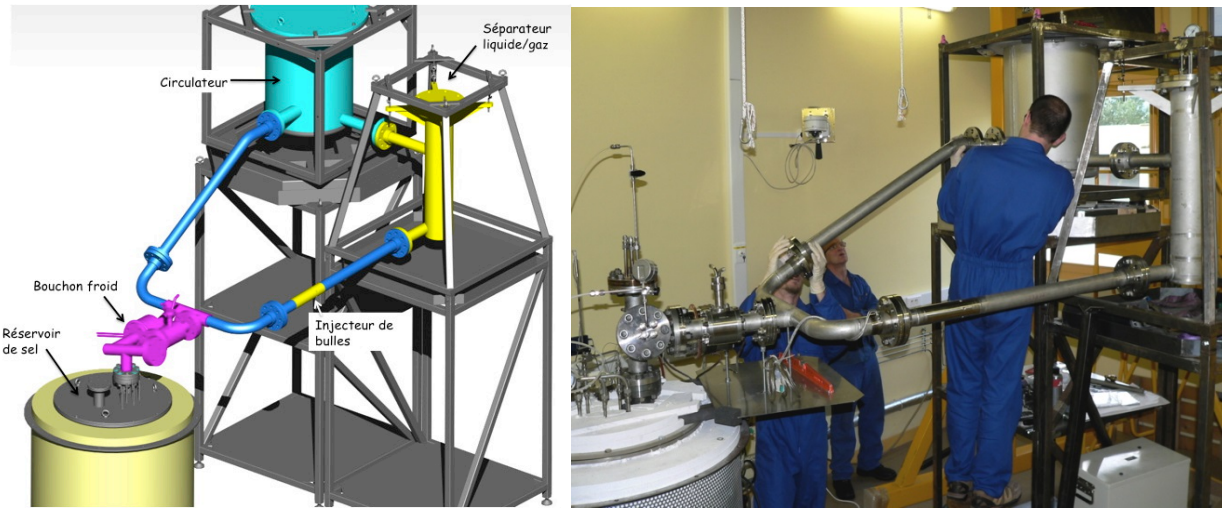

Figure 18. Design of the FFFER loop and view during assembly

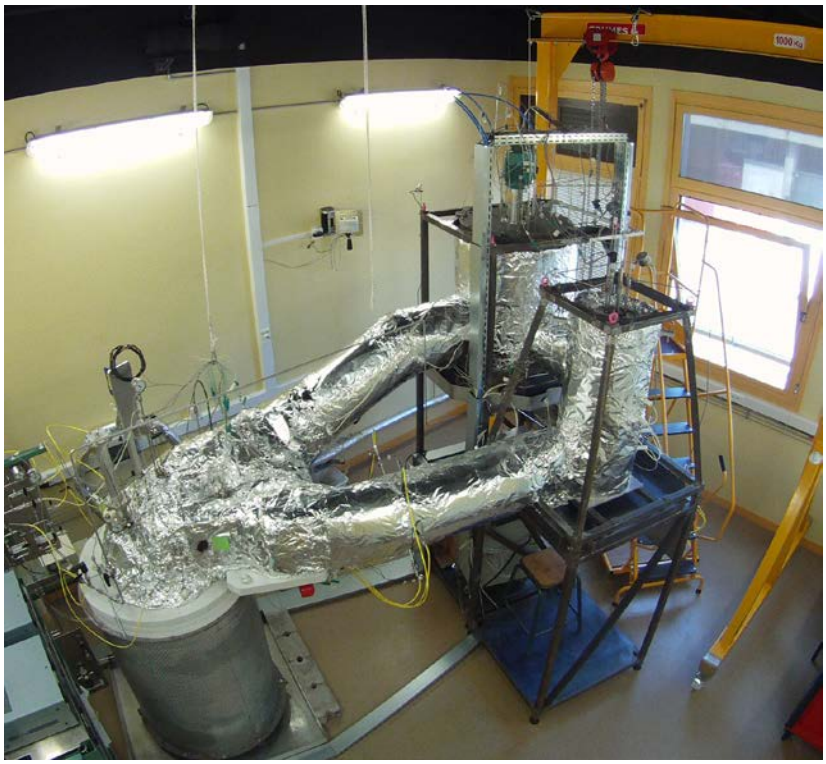

Figure 19. Completed loop with its thermal insulation

The injection and separation devices were designed after a transparent water mock-up (scale 0.72) was operated, allowing to gain familiarity with the tuning of all the parameters from the circulation pump to the separator as well as with the ultrasonic measurement of the velocity. An illustration of the vortex

created by the tangential fluid inlet at the base of the separator is shown in Figure 20. The bubble water separation efficiency reached about 85% at 0.1% volume fraction of gas, up to more than 95% for a 0.4% volume fraction of gas.

Figure 20. Water mock-up of the separator showing the concentration of bubbles in the vortex center and their coalescence. The gas is evacuated at the top and the liquid through the pipe on the left

The ultrasonic velocimetric technique is based on ultrasonic reflections on bubbles to depict the velocity profile across the pipe. This gives information about the bubble distribution and their mean velocity. However, this is a new technique which requires further studies and some tuning before it can be used outside the laboratory.

This experiment allowed casual observations of corrosion that are being studied separately on static small scale experiments.

A second loop is planned in the Euratom SAMOFAR project to identify and measure the salt's thermal behavior during thermal exchanges.

6.4 Other R&D activities on Molten Salt Systems

MSR development worldwide is still at a conceptual design stage, with most investigations around these concepts based today on numerical modeling, with the notable exception of the People's Republic of China, where a large project to develop a thorium MSR prototype has started very recently.

Recent MSR developments in the Russian Federation are focused on the 1000MWe molten-salt actinide recycler and transmuter (MOSART). The primary specifications for a MOSART core were to provide the fissile concentration and fuel salt geometry such that about 2.4GWt nuclear heat would be released at conditions affording efficient transmutation & recycling of TRUs from MOX PWR spent fuel (Ignatiev, 2012). The MOSART reference core with no graphite moderator is a cylinder 3.4 m in diameter and 3.6 m high. The fuel salt inlet and outlet pipe diameters are fixed at 1m. Radial, bottom, and top reflectors are attached to the reactor vessel. This leaves a ring filled with fuel salt surrounding the core to cool reflector and reactor vessel. The molten salt flow rate is 10000 kg/s. In nominal conditions, the fuel salt enters the core at 600°C and transports 2.4GWt to the secondary salt in the primary heat exchanger. The fluoride fuel salt mixture is circulated through the reactor core by four pumps operating in parallel. Other pumps circulate the salt through the heat exchangers and return it to a common plenum at the bottom of the reactor vessel. In the reference MOSART design, the out of core salt volume is 18 $m^{3.}$ The MOSART concept is being studied in different configurations which consider different core dimensions and different compositions of the fuel salt and/or salt blanket that allow for different modes of utilization. A detailed description of MOSART can be found in (Afonichkin et al, 2014).

7. Conclusion and perspectives

The MSFR concept has been recognized as a long term alternative to solid-fueled fast neutron reactors because of attractive features that remain to be confirmed.

It is characterized by:

- fluoride-based liquid fuels of various compositions (solvent, fertile and fissile) allowing operation as breeder or burner with many different possible fertile and fissile compositions;
- fast neutron spectrum;
- homogeneous fuel composition thanks to fast fuel circulation (in-core turbulence and multiple heat exchanger channels). This homogeneity allows continuous fuel monitoring;
- continuous extraction of volatile or metallic fission products via neutral gas bubbling.
- quasi continuous light chemical fuel processing (rate comparable to LWR solid fuel but on a daily basis) without stopping the reactor.

These characteristics result in a reactor with a high safety potential due to:

- negative temperature feedback reactivity coefficients (Doppler and density) leading to high thermal stability in operation and in all perturbing circumstances
- homogeneous liquid state allowing passive draining of the core fuel into passively cooled geometrically non-critical tanks
- absence of significant reactivity reserve because of the quasi continuous adjustment of the fuel composition
- No pressurization required due to the absence of any volatile fluid susceptible to be contaminated by fuel leaks.

The international MSFR collaboration is presently focused on technology-independent safety issues, considering that only a high safety level may convince safety agencies to authorize the development of such a new reactor concept. Since 2001, calculations and experimental research were conducted in Europe in national programs (CNRS-France, KI-Russia) and in a European network supported by Euratom and Rosatom (MOST, ALISIA, ACSEPT/ PYROSMANI, EVOL/MOSART,). This collaboration is presently continuing with the SAMOFAR/SMART-MSFR joint projects (2015/2019) where industrial partners (EdF, AREVA) and the French technical safety organization (IRSN) will be actively involved. This common program is devoted to the acquisition of experimental data and simulation tools for safety studies. The specific objectives of the Euratom program are:

- To develop and apply a new safety methodology for liquid fuel reactors, which could partly be woven into into the safety methodology of other Generation-IV reactors as well.

- To measure all relevant safety-related data of the fuel salt and of the whole system needed for the assessment of the MSFR.

- To design and build a software simulator to verify the safe operation of the MSFR including start-up, shut-down and load-following operation, and to identify normal operation accident initiators.

- To extract a complete set of accident initiators and scenarios, and to evaluate these using best-estimate simulation tools including uncertainty analysis.

- To prove experimentally and numerically the safe and reliable operation of the freeze valves and the draining of the fuel salt, and to measure the natural circulation dynamics of the (internally heated) fuel salt in a loop, representing the primary circuit and drain tanks.

- To demonstrate experimentally the reductive extraction processes for lanthanides and actinides, and to assess the safety of the high temperature chemical processes to clean and control the fuel salt.

Since the beginning, the common philosophy of the MSFR community was to give priority to knowledge over technology assuming that a long time will be devoted to assess the safety of technological solutions, i.e. assuming that safety is the primary concern for public acceptance of new nuclear reactors. The resulting roadmap for future developments is presently concerned with all the chemical and physical knowledge that help to assess the MSFR characteristics and design, including basic data measurements and multiphysics simulation tools. A second step will be the development of technological means, using simulant salts instead of real fuel, in order to demonstrate, at the proper scale, the validity of the proposed technology and to validate fluid flow and heat transfer models. The third step is the zero power demonstration small reactors, with the objectives of checking the neutronic properties (eliminating data uncertainties) and testing the start-up and shut-down processes. Then, it will be possible to test a small power reactor with two new tests: the heat transfer with internal heat source and the fission product extraction (continuous and quasi-continuous). This means that the pyroprocessing of the fuel by remote handling should be studied and tested in parallel to the first three steps, as well as the safety and proliferation issues. Indeed, the option of studying all the aspects of the concept was taken from the beginning to render the safety constraints inherent to the design and not have them added after. This implies using new approaches in agreement with the GIF community for safety and proliferation resistance. All these steps are mandatory to develop the technical and scientific background and knowledge for further practical demonstrations of the flexibility and viability of Molten Salt Reactors on a reactor scale. Such R&D activities are being conducted in the world, particularly by a European network supported by EURATOM and ROSATOM to confirm the validity of the theoretical advantages of this concept and to assess the potential advantages of fast spectrum molten salt reactors.

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