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Introduction to the Physics of Thorium Molten Salt Fast Reactor (MSFR) Concepts

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Abstract

Recent conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts have kindled renewed interest in molten salt reactors. This concept, operated in the thorium fuel cycle, may be started either with ^{233}U, enriched U, and/or transuranic elements as the initial fissile load. This paper describes some studies and developments around the MSFR concept based on the thorium fuel cycle. MSFRs are seen as a long-term alternative to solid-fueled fast neutron systems thanks to their unique potential, which includes large negative temperature and void coefficients, lower fissile inventory, no initial criticality reserve, a simplified fuel cycle, waste reduction, etc. They have been selected as one of the reference reactors of the Generation IV International Forum.

Introduction

The molten salt fast reactor (MSFR) was chosen by the Generation IV International Forum (GIF) in 2008 as a representative molten salt reactor fitting the Gen IV criteria [1] because of its fast spectrum, sustainability, and waste minimization, and the use of thorium as fertile element owing to its proliferation resistance [2–7]. In such a homogeneous reactor, the main safety characteristics are due to the absence of any moderator or construction materials in the core, which contains only the liquid fuel salt components. Thermal dilation of the liquid fuel salt gives it a thermal feedback coefficient of about −5 pcm/K, which allows power tuning by heat extraction. Because of a negative void feedback coefficient, draining the liquid fuel salt in geometrically subcritical tanks allows long term stalling with passive cooling for decay heat removal. Two advantages of having the fissile and fertile isotopes in a liquid fuel are: (1) the possibility of fuel composition adjustment without stopping the reactor and (2) the circumvention of the difficulties of solid fuel fabrication with large amounts of transuranic elements (TRU). Indeed, this reactor may be operated with a variety of fissile and fertile elements but is most efficient with ^{233}U, Pu, and Th.

This type of reactor is still at a conceptual level, based on numerical modeling. However, in the 1950s and 60s, experimental studies were conducted at the Oak Ridge National Laboratory (ORNL) in the USA. This provided a very valuable experimental base to assess the feasibility of such reactors. In 1958, a water-based liquid fuel was used in a 5 MW_{th} homogeneous reactor experiment called HRE-2, which demonstrated the auto-stability of homogeneous reactors. From 1966–1969, an 8 MW_{th} experimental graphite-moderated molten salt reactor was operated for four years without any trouble, demonstrating that using a molten fluoride salt at 650 °C was possible. However, this molten salt reactor experiment (MSRE) only tested fissile isotopes (^{233}U, ^{235}U, and Pu), not thorium, for breeding. Later, ORNL studied in detail a power reactor called the molten salt breeder reactor (MSBR), which was never built. This design was a thermal reactor with a graphite-moderated core that needed intense chemical salt treatment with about a 30-day removal time for soluble fission products, a draw-back that is eliminated with a fast spectrum.

This paper describes some studies and developments around the MSFR concept and illustrates the contemporary

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interest in fast reactor concepts based on the thorium fuel cycle, which is seen as a long-term alternative to solid-fueled fast neutron reactors.

Description of the MSFR Concept

Core and System Designs

Conceptual design activities are currently (2013) 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 savings (closed Th/U fuel cycle, no uranium enrichment), safety (no reactivity reserve, strongly negative feedback coefficient), and waste management (actinide burner). Calculation results presented here were obtained for a somewhat arbitrarily chosen reactor called "reference MSFR". This is not to be taken as an optimized reactor, but as a basis for interdisciplinary studies.

The reference MSFR is a 3 GW_{th} reactor with a total fuel salt volume of 18 m^3, operated at a maximum fuel salt temperature of 750 °C [8, 9]. More recently, thermal-hydraulic studies have been performed in the frame of the EVOL (evaluation and viability of liquid fuel fast reactor system) FP7 project, resulting in a torus shape of the core [10, 11]. As shown in Fig. 1, the fuel salt flows from the bottom to the top of the core cavity (note the absence of in-core solid matter). 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 circuit is 3–4 s [12]. The fuel salt considered in the simulations is a molten binary fluoride salt with 77.5 mol% lithium fluoride; the other 22.5 mol% consists of a mix of heavy nuclei fluorides. This proportion, maintained throughout the reactor evolution, leads to a fast neutron spectrum in the core. The total fuel salt volume is distributed half in the core and half in the external part of the fuel circuit. This MSFR system thus combines the generic assets of fast neutron reactors (extended resource utilization, waste minimization) with those associated with a liquid-fueled reactor.

In preliminary designs developed in relation to calculations, the core of the MSFR is a single compact cylinder (2.25 m high × 2.25 m diameter) and the nuclear reactions occur within the liquid fluoride salt, which acts both as fuel and as coolant. 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 20-cm 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 Fig. 1) to increase the breeding ratio. This blanket is filled with a LiF-based fertile salt with initially 22.5 mol% $^{232}ThF_4$.

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 leading to an excessive temperature being reached in the core. In such situations, the fuel salt geometry can be passively reconfigured by gravity-driven draining of

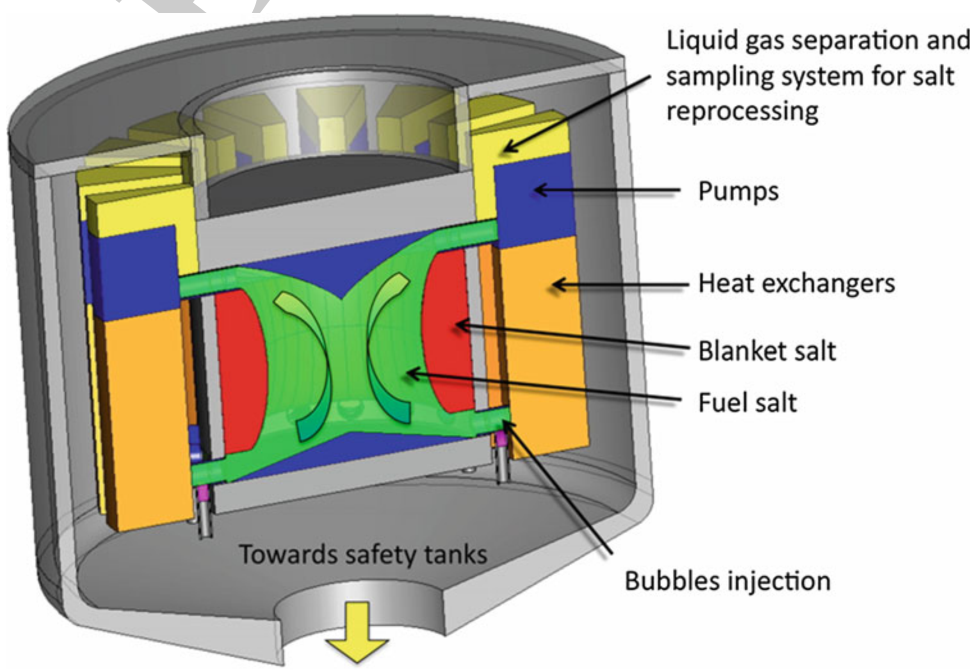

Fig. 1 Schematic drawing of the MSFR design. Fluoride-based fuel salt is *green*, fertile blanket salt is *red*

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

Figure 2 is a general view of what a reactor could look like, with its elements represented as generic boxes for the various functions because they have not yet been studied in detail.

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 subcritical 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.

Salt Cleaning and Reprocessing

The fuel salt undergoes two types of treatment: on-line neutral gas bubbling in the core and remote mini-batch reprocessing on-site [13]. These salt treatments aim to remove most of the fission products without stopping the

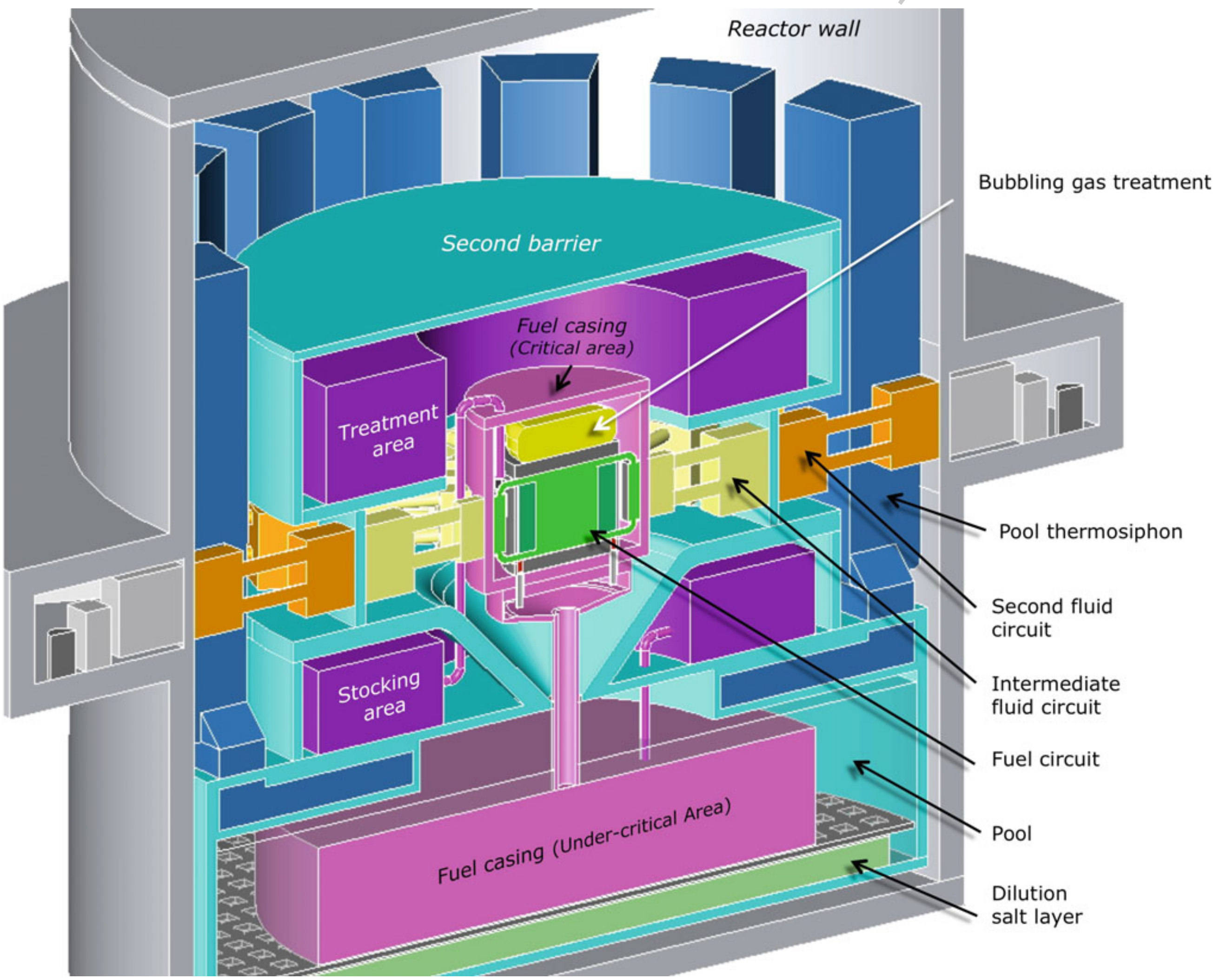

Fig. 2 Illustration of the main functions associated with the MSFR operation. In the middle is the *green* fuel salt circuit surrounded by a *pink* envelope representing the first confinement barrier. The *cyan* envelope represents the second barrier, including storing and chemical salt processing units in *violet*. The third barrier is in *gray*. Two heat transfer circuits between the three barriers are represented as loops in *yellow* and *orange*

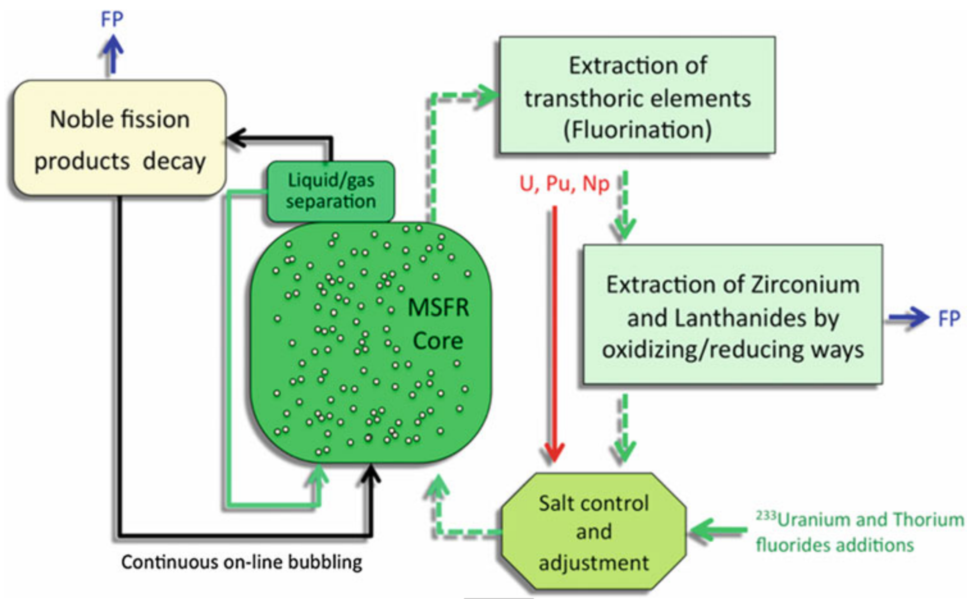

Fig. 3 Schematic representation of the fuel salt treatment with two loops

reactor and, thus, secure a rather small fissile inventory outside the core compared with present-day light water reactors (LWRs). The reprocessing rate itself is assumed to be 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 Fig. 3. It consists of 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 area, where most of the Kr and Xe decay into Rb and Cs, preventing their accumulation in the fuel salt. The remaining gas is recycled.

On the left is the on-line treatment with gas bubbling in the core to extract noble gases and metallic particles (FP). 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 L per day to limit the lanthanide and Zr concentrations in the fuel salt. The sampled salt 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 oxide reduction 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 products before they decay in the salt. The pyrochemical salt batch reprocessing avoids the accumulation of large quantities of lanthanides and zirconium in the fuel salt, a process 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 it 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 Fig. 4. Notice that with the reactor configuration used for the calculation, the core is an under-breeder. Breeding is reached for the reprocessing of a full load up to 4000 days owing to the addition of the fertile blanket.

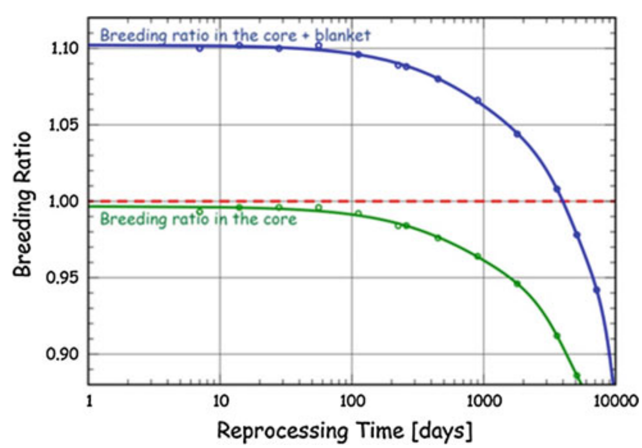

Fig. 4 Influence of the batch reprocessing rate on the breeding ratio in the core and in the whole MSFR system (core+fertile blanket)

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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, that is, using more fissile material than they produce, need to be regularly re-fueled with fissile material throughout their operation time. Conversely, breeder generation IV reactors (SFR, MSFR, GFR) require only one (or two in the case of solid-fuel reactors) initial loading of fissile material. They then produce at least the amount of fissile material they need to be operated during their entire lifespan. Molten salt reactors require only one fissile load as no fuel re-fabrication is necessary and the fuel salt composition is controlled on-line without stopping reactor operation, whereas two loads are necessary for solid-fuel 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 [15, 16]:

- ^{235}U, the only natural fissile material on earth (0.72% of natural uranium). It can be used directly to start MSFRs with enriched uranium as the initial fissile material, with an enrichment ratio of less than 20% due to proliferation resistance issues;
- MSFRs can be directly started with ^{233}U as the initial fissile material, assuming that this ^{233}U can be produced in fertile blankets of other reactors (SFR, etc.) 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 MSFRs that are breeder reactors, allowing the deployment of such ^{233}U-started MSFRs in a second phase even if no ^{233}U is produced elsewhere;
- Using the plutonium produced in current pressurized water reactors (PWRs) or in future EPRs as the initial fissile material. An even better scenario would be the use of mixtures of TRU produced by these Generation II or III reactors.
- A mixture of these starting modes. For example, ^{233}U may be produced by using special devices containing thorium and Pu–MOX in cur-rent PWRs or in future EPRs.

Figures 5 and 6 present comparisons of fuel composition evolutions of a "3 GW$_{th}$ reference MSFR" reactor started with ^{233}U, TRU, Th–MOX, or enriched U and TRU.

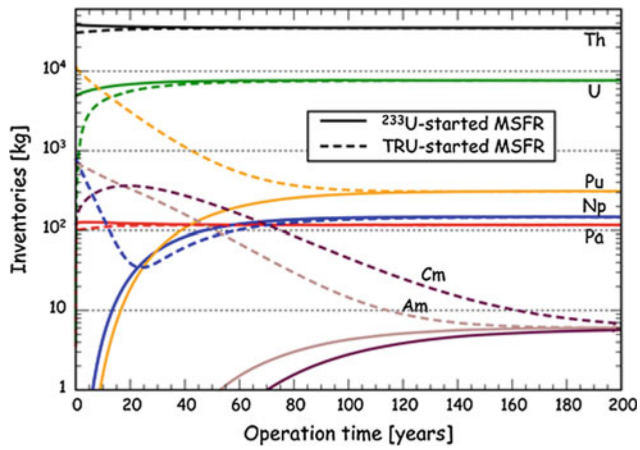

Fig. 5 Time evolution up to equilibrium of the heavy nuclei inventory for the ^{233}U-started MSFR (*solid lines*) and for the TRU-started MSFR (*dashed lines*). Operation time is given in equivalent full power years (EFPY) [14]

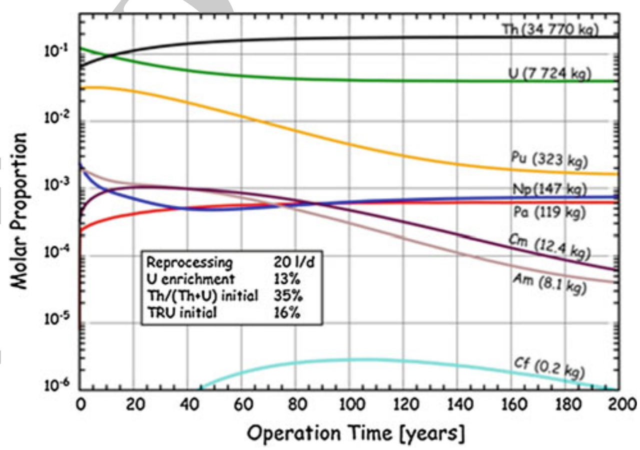

Fig. 6 Time evolution up to equilibrium of the heavy nuclei inventory for the optimized MSFR configuration started with enriched uranium and TRU elements. Operation time is given in EFPY

Safety Issues

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 attendance. Moreover, for the MSFR, reactor stability is strengthened by its large negative feedback coefficients. Some of these features are discussed below, but not all safety provisions are detailed.

Safety Approach and Risk Analysis for a Liquid-Fueled Reactor

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

1. The principle of defense in depth and multiple barriers has to be adapted as the conventional barriers (such as cladding, primary circuit and containment in a PWR) are no longer applicable;
2. Diversity and independence of the MSFR reactivity control mechanisms have to be demonstrated (no control or shutdown rods or burn-able poisons);
3. New safety criteria to evaluate reactor response during normal, incidental, and accidental conditions are needed as the MSFR fuel is in the liquid state, which is not an acceptable situation for the LWR fuel;
4. 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;
5. Evaluation of the risk posed by the residual decay heat and the radioactive inventory in the reprocessing unit is also necessary.

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 currently 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, owing to the limited amount of operation experience and some of its novel features, any new methodology should be robust and comprehensive, and integrate both deterministic and probabilistic approaches. To fulfill these objectives, a MSFR design and safety analysis methodology is currently being developed [17] according to the following steps:

1. Systemic modeling of all reactor components by 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 (including failure modes and dangerous phenomena);
4. Risk evaluation based on evaluation of probability and severity.

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 plants' operating conditions. With MSFR development being at its early stages, the idea is to adopt an inherent safety-by-design approach.

Decay Heat Removal

The decay heat generation versus time is represented in Fig. 7. Based on the concept described above, 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 a third of the heat is produced in the gas processing unit and two thirds in the liquid fuel. The power of both heat sources decreases rapidly (by a factor of ten in about one day) from the value at shut down, which depends on the history of the power generation. The total amount of power at shut down is about 5 % of the nominal power. This value is lower compared with 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 placed 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 by a factor of ten, for instance. This heat transfer is achieved by passive thermosiphons 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.

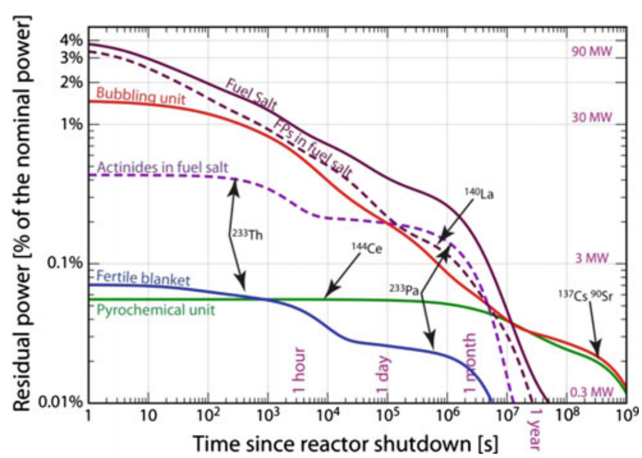

Fig. 7 Residual heat in the different radioactive fluids of the MSFR, after the total fission shut-down of the reactor previously under steady-state conditions [12, 17]

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Issues and Demonstration Steps of the Concept Viability

Despite the status of preconception design of MSFRs, several limiting factors can be identified in the development of the concept.

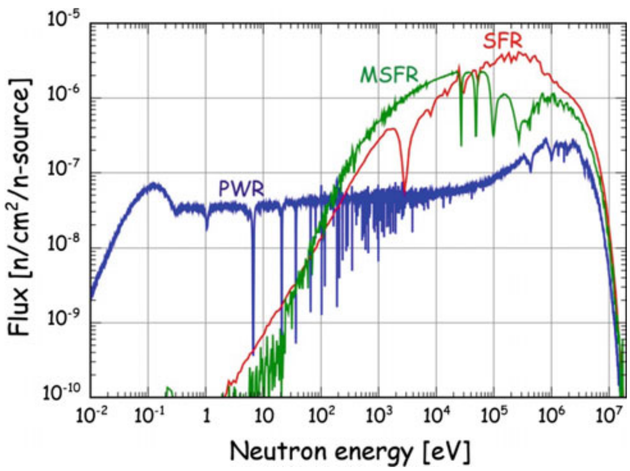
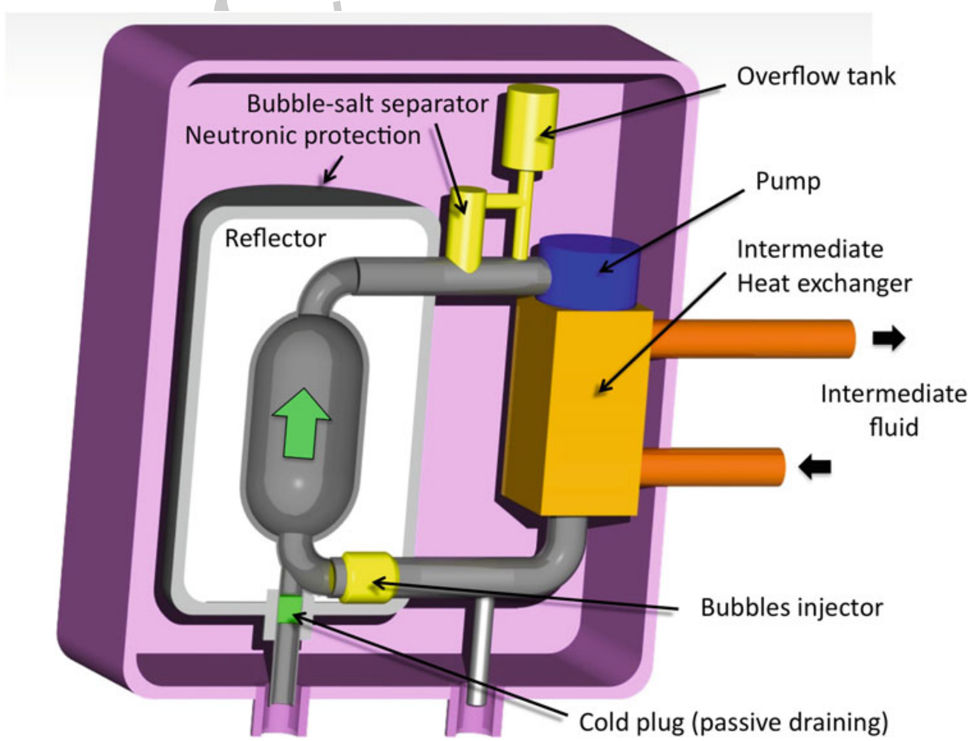


Fig. 8 Fast neutron spectra of the reference MSFR (*green curve*) and of a sodium-cooled fast neutron reactor (SFR, *red curve*) compared with the thermalized spectrum of a pressurized water reactor (PWR, *blue curve*) [14]

The first, obvious, issue is materials resistance to high temperatures, 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 100–150 °C for in-core temperature heating corresponding to a salt circulation period of 3–4 s, 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, that is, the same temperature as the heat exchanger plates during the heat transfer, the intermediate coolant salt being at about 650 °C. Although it seems that there are current alloys that can withstand such temperatures for a long time, this could still be a limit unless the material is replaced regularly, as is done with solid-fuel cladding.

The second issue is resistance to the neutron flux at high temperature, unless low power density operation is chosen. Calculations of the maximum displacement per atom (dpa) of the core walls yield 7.5 dpa/year for a power density of 330 W/cc. This is less than expected for solid-fuel fast reactors because of the neutron spectrum difference that is due to neutron inelastic scattering on fluorine nuclei, as shown in Fig. 8, and the absence of solid material in the core.

Fig. 9 Sketch of a single liquid fuel loop reactor for demonstration purposes or modular conception. The fuel volume (1.8 m^3) is reduced by a factor of ten from the 3 GW_{th} reactor and the power (200 MW_{th}) by a factor 15 in order to use the same intermediate heat exchanger



The third issue appears when trying to limit the per GW fissile inventory. This means restricting as much as possible the proportion of fuel salt out of the core, that is, in the tubing, pumps, and heat exchangers. It is technically challenging to reduce this "useless" amount of salt to less than 50 % of the total load and 30 % appears as a limit.

The fourth issue is a question more than a real limit: the safety evaluation. Indeed, present-day safety evaluation techniques are suitable for solid-fuel water reactors but partly irrelevant for liquid-fuel reactors. A new way of tackling the problem should find a consensus before any national safety authority can approve a liquid-fuel reactor design and this will take time and resources.

The size of the reactor liquid-fuel loop is not a limit, as shown by the calculation of a single-loop 200 MW_{th} reactor instead of a 16-loop 3 GW_{th} reactor. A low power demonstration version [18] is sketched in Fig. 9, but a regenerator version could be implemented by replacing the reflectors with a blanket. The size of this fuel loop assembly is about 2.5 m in diameter and 3 m high (core: 1.1 m diameter and 1.1 m high). The power is limited by the intermediate exchanger size, which is assumed to be the same as for the 3 GW_{th} reactor.

From the parametric studies that were carried out on the MSFR, no stumbling blocks appeared and the various limits can all be circumvented by reducing the power density.

Conclusion

Since 2005, R&D on molten salt reactors has been focused on fast spectrum concepts (such as the MSFR), which have been recognized as a long-term alternative to solid-fuel fast neutron reactors as MSFRs have attractive features such as very negative feedback coefficients, smaller fissile inventory, and a simplified fuel cycle. Experimental research on basic data is being conducted by a European network supported by EURATOM and ROSATOM to confirm the validity of the theoretical advantages of this concept. No insurmountable obstacles have been identified thus far, but almost all the technology remains to be tested, and demonstration experiments will have to be conducted to continue to assess the potential advantages of fast spectrum molten salt reactors, regardless of whether they are based on the thorium fuel cycle or are used as TRU burners.

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