Optimizing the Burning Efficiency and the Deployment Capacities of the Molten Salt Fast Reactor

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Abstract – Molten salt reactors in the present configuration, called Molten Salt Fast Reactors (MSFR), have been selected for further studies by the Generation IV International Forum. These reactors, characterized by a fast neutron spectrum, may be operated in simplified and safe conditions in the Th/233U fuel cycle with fluoride salts. Since 233U does not exist on earth and is not being directly produced today, we investigated the possibility of using, in this MSFR concept, the transuranic elements (TRU) currently produced in the world as an initial fissile load. We present here the concept, before focusing on a possible optimization in terms of minimization of the initial fissile inventory and improvement of the burning efficiency. Our studies demonstrate that an initial fissile inventory around 4 to 5 metric tons per GWe may be easily reached with a burning rate after 50 years of operation ranging from 87% to 93%. These studies also bring to light the limitations of the concept due to the irradiation damages to the structural materials. We conclude that this issue will have to be studied in depth to allow a realistic evaluation of the global possibilities of such a reactor.

I. INTRODUCTION

Starting from the Oak-Ridge Molten Salt Breeder Reactor project1, we have performed some parametric studies2,3,4,5 correlating the core arrangement, the reprocessing performances, and the salt composition, in terms of safety coefficients, reprocessing requirements, and breeding capabilities. In the frame of this major re-evaluation of the molten salt reactor (MSR), we developed a new concept that we called the Thorium Molten Salt Reactor (TMSR), which is particularly well suited to fulfil the criteria identified by the Generation IV International Forum.

This reactor may be operated in simplified and safe conditions in the Th/233U fuel cycle with fluoride salts. Its main advantages, due to the liquid fuel and coolant and to the thorium cycle, are the following: the amounts of fissile and fertile matter can be adjusted without unloading the core, avoiding any initial reactivity reserve; the Fission Products which poison the core can be quickly extracted; nuclear waste production is minimized; very high temperatures may be reached without requiring high pressure in the core.

Amongst all TMSR configurations, our recent studies have singled out the configurations with no moderator in the core: they are simple and very promising, the moderation ratio depending only on the salt composition. Such a reactor presents many intrinsic advantages, in addition to avoiding the deterioration of the moderator while ensuring excellent safety characteristics. In previous articles we presented this reactor under the name “non-moderated Thorium Molten Salt Reactor” or TMSR-NM. With a fuel salt content low in light elements a fast neutron spectrum is obtained. This concept, now called Molten Salt Fast Reactor or MSFR, has been selected for further studies by the Generation IV International Forum.

Since 233U does not exist on earth and is not being directly produced today, we investigated the possibility of using, in this MSFR concept, the transuranic elements (TRU) currently produced in the world as an initial fissile load. We thus aim at designing the most efficient, robust and simple MSR as not only an excellent Thorium-based breeder reactor, but also a device able to burn the Plutonium and the Minor Actinides (MA) produced in today’s reactors, and consequently to close the current fuel cycle while converting this Plutonium into 233U. We will concentrate in this paper on these MSFRs started with TRUs that we will refer to as the TRU-started MSFRs.

In this frame, after introducing this concept and its characteristics, we will detail an optimization of the reactor...
in terms of TRU burning efficiency and deployment capacities. This last goal can be achieved, without losing the advantages of the concept (safety, breeding capacities...), by minimizing the fissile inventory, the limits being set by irradiation damages to the structural materials and the capacities of the heat exchangers.

Our neutronic results rest on numerical simulations making use of the MCNP neutron transport code\textsuperscript{9} coupled with an in-house materials evolution code REM\textsuperscript{2,5,7,8}. The former evaluates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition within the cells. These calculations take into account the input parameters (power released, criticality level, chemistry...), by continuously adjusting the neutron flux or the materials composition of the core. Our calculations rest on a precise description of the geometry and consider several hundreds of nuclei with their interactions and radioactive decay. These codes are also used to determine the safety parameters (temperature coefficients, fraction of delayed neutrons) through complete system simulations.

II. THE MOLTEN SALT FAST REACTOR CONCEPT

II.A Reactor geometry

As shown in Fig. 1, the core is a single cylinder (the diameter being equal to the height) where the nuclear reactions occur within the flowing fuel salt. The core is composed of three volumes: the active core, the upper plenum, and the lower plenum. The fuel salt considered in the simulations is a binary salt, LiF - (Heavy Nuclei)F\textsubscript{4}, whose (HN)F\textsubscript{4} proportion is set at 22.5 mole % (eutectic point), corresponding to a melting temperature of 565°C. The choice of this fuel salt composition results from many systematic studies (influence of the chemical reprocessing on the neutronic behaviour, burn up capabilities, deterministic safety evaluation, deployment capabilities)\textsuperscript{7,8,9,10,11,12}. This salt composition leads to a fast neutron spectrum in the core.

The operating temperatures chosen for our neutronic simulations range from 700°C to 850°C, the lower limit due to the salt’s melting point, the upper limit to the structural materials chosen for our simulations (see section II.C). We have considered a thermodynamic efficiency of 50%, lower than the Carnot efficiency, to be realistic.

The external core structures and the heat exchangers are protected by thick reflectors made of nickel-based alloys which have been designed to absorb more than 80% of the escaping neutron flux. These reflectors are themselves surrounded by a 10cm thick neutronic protection of B\textsubscript{4}C which absorbs the remaining neutrons. As shown in Fig.1, the radial reflector is a fertile blanket (50 cm thick) filled with a fertile salt LiF-ThF\textsubscript{4} with 22.5% - mole \textsuperscript{232}Th. Thanks to \textsuperscript{233}U extraction every six months, this fertile blanket improves the global breeding ratio of the reactor.

Fig. 1. Schematic view of a quarter of the MSFR, the fuel salt (not represented here) being located within the orange lines

The total fuel salt volume, one of the parameters of the present study, is distributed between the core and the external circuit (see section 2.3). This distribution impacts the fraction of delayed neutrons produced in the core and thus the safety behaviour of the core. In a rough approach, these neutrons are produced anywhere in the fuel salt with a quasi-uniform probability so that the larger the volume of salt located outside the core, the fewer the delayed neutrons produced in the core. More precise studies based on safety analyses (with seven precursor families and their decay times)\textsuperscript{7} have demonstrated that the reactor behaves safely with at least half of the delayed neutrons produced in the core. Previous studies\textsuperscript{13} based on two fuel salt distributions, either 1/2 (core) – 1/2 (external circuit) or 2/3 (core) – 1/3 (external circuit), have demonstrated that the former (1/2 in the core – 1/2 in the external circuit) is less constraining for the heat exchangers. This is the fuel salt distribution that we have used in this paper.

II.B Fuel salt reprocessing

The salt management combines a salt control unit, an online gaseous extraction system and an offline lanthanide extraction component by pyrochemistry\textsuperscript{14}. The gaseous extraction system, where helium bubbles are injected in the core, removes all non-soluble fission products (noble metals and gaseous fission products). With the online control and adjustment of the salt composition, the reactivity can be kept equal to one. A fraction of salt is periodically withdrawn and reprocessed offline in order to extract the lanthanides before it is sent back into the core.
The actinides are sent back into the core as soon as possible in order to be burnt. The rate at which this offline salt reprocessing is done depends on the desired breeding performance. In the simulations presented here, we have fixed the reprocessing rate at 40 liters per day whatever the fuel salt volume.

II.C Thermohydraulic considerations

The fuel salt flows upward in the core until it reaches an extraction area which leads to salt-bubble separators after passing through salt collectors. The salt then flows downward in the primary heat exchangers and the pumps before finally re-entering the bottom of the core through injectors.

The external circuit (salt collector, salt-bubble separator, heat exchanger, pump, salt injector and pipes) is broken up in 16 identical modules distributed around the core, outside the fertile blanket and within the reactor vessel. We have divided the external circuit in two parts: the pipes (including the salt-bubble separator, the pump and the injector) and the heat exchangers. The distribution of the salt between these two parts is chosen so as to minimize the pressure drops in the circuit. The fuel salt runs through the total cycle in 3 to 6 seconds, depending on the total fuel salt volume and the salt flow velocity. The salt circulation being considered uniform, the residence time of the salt in each zone of the circuit and the core is proportional to the volume of this zone.

II.D Physicochemical properties of the fuel salt

The initial fuel salt is composed of $\text{LiF-ThF}_4$-(TRU)F$_3$ with 77.5 mole % of LiF, this fraction being kept constant during reactor operation. More precisely, in our studies, this MSFR is started with the mix of Pu, Np, Am and Cm listed in Table I corresponding to the transuranic elements of an UOX fuel after one use in a standard PWR and five years of storage.

The fraction of fissile isotopes, being adjusted to have an exactly critical reactor, depends on the total salt volume. The initial TRU proportion in the fuel salt ranges from 6.8 mole % to 6.3 mole % when the fuel salt volume varies from 9 $\text{m}^3$ to 27 $\text{m}^3$.

The results presented in section III are often expressed in ‘amount of fissile isotopes’; the corresponding total TRU amount may be obtained through the proportions given in Table I.

An equivalent salt but with no fissile matter (22.5 LiF-77.5 ThF$_4$) is used as fertile salt in the radial blanket surrounding the core and serves as radial reflector.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Proportion in the mix</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np 237</td>
<td>6.3 mole%</td>
</tr>
<tr>
<td>Pu 238</td>
<td>2.7 mole%</td>
</tr>
<tr>
<td>Pu 239</td>
<td>45.9 mole%</td>
</tr>
<tr>
<td>Pu 240</td>
<td>21.5 mole%</td>
</tr>
<tr>
<td>Pu 241</td>
<td>10.7 mole%</td>
</tr>
<tr>
<td>Pu 242</td>
<td>6.7 mole%</td>
</tr>
<tr>
<td>Am 241</td>
<td>3.4 mole%</td>
</tr>
<tr>
<td>Am 243</td>
<td>1.9 mole%</td>
</tr>
<tr>
<td>Cm 244</td>
<td>0.8 mole%</td>
</tr>
<tr>
<td>Cm 245</td>
<td>0.1 mole%</td>
</tr>
</tbody>
</table>

II.E Materials

The structural materials of the reactor, even if they are located around the core and not directly in it, have to bear the neutron flux together with high temperatures. We have considered for our simulations a Ni-based alloy containing W and Cr as detailed in Table II.

<table>
<thead>
<tr>
<th>Ni</th>
<th>W</th>
<th>Cr</th>
<th>Mo</th>
<th>Fe</th>
<th>Ti</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>79.432</td>
<td>9.976</td>
<td>8.014</td>
<td>0.736</td>
<td>0.632</td>
<td>0.295</td>
<td>0.294</td>
</tr>
<tr>
<td>Mn</td>
<td>Si</td>
<td>Al</td>
<td>B</td>
<td>P</td>
<td>S</td>
<td></td>
</tr>
<tr>
<td>0.257</td>
<td>0.252</td>
<td>0.052</td>
<td>0.033</td>
<td>0.023</td>
<td>0.004</td>
<td></td>
</tr>
</tbody>
</table>

III. OPTIMIZATION OF THE CONCEPT

For the earlier versions of the TRU-started MSFR that we studied, the initial fissile load was quite large, around 8.5 metric tons of TRU fissile isotopes per GWe. This concept is reliable and robust enough to allow further optimizations, such as a minimization of the fissile inventory. The parameters for this minimization are the operating temperature of the reactor and its specific power. The operating temperature we chose is already as hot as possible for the structural materials considered here.

Thus, in this section, we will detail the impact of the specific power on the reactor’s operation. With a fixed total power of 3 GWth and a fixed proportion of fuel salt...
in/out of the core, the specific power is modified by varying the total fuel salt volume itself. We have considered values for the fuel salt volume ranging from 9 m$^3$ to 30 m$^3$, corresponding to a mean specific power in core varying from 670 W/cm$^3$ to 200 W/cm$^3$. The effects of this parameter on the neutronic behaviour of the core are detailed in section III.A.

As the MSFR uses a liquid fuel with no solid matter inside the core, the specific power can be very large compared to a reactor based on a solid fuel. The limitations are due to:

- The capacities of the heat exchangers in terms of heat extraction and the associated pressure drops. Recent studies$^{13}$ have shown that the heat exchangers are not really constraining compared to:
- The neutronic irradiation damages to the structural materials which modify their physicochemical properties through three effects detailed in section III.C.

We are also interested here in the optimization of the burning capacities of the reactor as a function of the specific power, as detailed in section III.D.

**III.A Fuel salt volume effects on the neutronics**

**III.A.1 Heavy nuclei inventory**

The initial fissile Pu inventory varies linearly from 2.6 metric tons per GWe for a fuel salt volume of 9 m$^3$ to 7.8 tons per GWe for a fuel salt volume of 27 m$^3$.

Fig. 2 illustrates the evolution of the heavy nuclei inventory in the fuel salt all along the operation of the reactor up to equilibrium, for the $^{233}$U-started (solid lines) and for the TRU-started (dashed lines) MSFR, in the case of a medium configuration containing 18m$^3$ of fuel salt. The two MSFRs are equivalent at equilibrium.

**III.A.2 Deployment capacities**

The goals of this TRU-started MSFR being to close the current fuel cycle and to allow the deployment of a fleet of MSFRs in the Th fuel cycle, the deployment capacities are based on the amount of $^{233}$U produced in excess in each reactor compared to the initial fissile ($^{233}$U) inventory necessary to start a $^{233}$U-started MSFR. The amount of $^{233}$U produced and extracted all along the lifespan of a medium MSFR configuration (18m$^3$ of fuel salt) is shown in Fig. 3 (blue line): for MSFRs directly started with $^{233}$U, the $^{233}$U extraction follows a linear growth, the production of an initial fissile load requiring 55 years in the case presented here. This production is directly related to the breeding ratio of the configuration.

![Excess production of $^{233}$U](image)

Fig. 3. Excess production of $^{233}$U expressed in term of initial fissile inventory, as a function of the reactor’s operating time

TRU-started MSFRs allow the extraction of significantly larger amounts of $^{233}$U during the first 20 years of operation (Fig. 3, red upper line), thanks to the burning of TRUs which saves a part of the $^{233}$U produced in the core. After the first 20 years, the $^{233}$U extraction rate is equivalent to that of the $^{233}$U-started MSFR. The production of an initial fissile ($^{233}$U) load thus requires only 30 years of such a reactor’s operation.

Finally, Fig. 4 presents the reactor’s first doubling time$^*$ as a function of the fuel salt volume. The shortest doubling times, less than 30 years, are obtained for a fuel salt volume ranging from 12 to 18 m$^3$ and for the TRU-started MSFR. Due to large neutron losses, the $^{233}$U-started MSFR configurations with a fuel salt volume less than 12 m$^3$ are not acceptable as a Generation 4 reactor, because of their poor deployment capacities.

* defined as the operating time necessary to produce in a reactor its initial fissile ($^{233}$U) inventory
Fig. 4. Reactor’s doubling time as a function of the fuel salt volume

III.A.3 Safety coefficients

The practical evaluation of the feedback coefficients is done as follows. The multiplication coefficient $k$ is first computed for the core with the matter compositions at equilibrium for a temperature of 775°C. It is then recalculated using the same compositions but at a different reactor temperature. In practice, the modifications concern the temperature of the salt itself, together with the density of the salt because of its dilatation (dilatation coefficient of $10^{-3}/°C$ in our case). Other temperature variations like those of the reflectors or the blanket are not considered since these materials have a very small contribution and heat up very slowly.

Fig. 5. Feedback coefficients as a function of the fuel salt volume

The total feedback coefficient at equilibrium is displayed in Fig. 5, together with its components, the contributions of the salt heating and salt density, as a function of the fuel salt volume. All these safety coefficients are comfortably negative for all MSFR configurations, ranging from -6 pcm/K to -4.5 pcm/K. The deterministic safety level is thus excellent for all these MSFR configurations.

The uncertainties indicated are a quadratic combination of the statistical and systematic uncertainties. The calculations are precise enough to lead to negligible statistical errors. Concerning the systematic uncertainties on the contribution of salt heating, the cross-sections concerned are well known, inducing only negligible uncertainties. The uncertainties on the salt density and its dilatation lead to systematic errors less than 20% on the contribution of salt dilatation.

III.B Materials damages

The neutronic irradiation damages to the structural materials modify their physicochemical properties through three effects: the displacements per atom, the production of Helium gas via nuclear reactions in the materials, and finally the transmutation of the Tungsten (component of the Ni-based alloy selected for this study) to Osmium through nuclear reactions. We have calculated these damages for the axial reflectors, which are the most irradiated elements of the core. For improved legibility, precise results will be given only for the medium case corresponding to a fuel salt volume of 18 m$^3$.

III.B.1 Displacements per atom

The radiation damages in neutron-irradiated materials, dependent on many factors like the irradiation dose and the neutron spectrum, are expressed in dpa (displacements per atom), corresponding to the number of times an atom is displaced for a given fluence. We have calculated these damages for the axial and radial reflectors, which are the most irradiated elements of the core. The damages in both reflectors are of the same order of magnitude.

Fig. 6. Radiation damages to the axial reflector as a function of distance from the core axis (zero corresponding to the centre of the core), for different depths of the reflector
Our calculations, displayed in Fig. 6 in the case of the MSFR with 18 m$^3$ of fuel salt, show that the damages are largest in the first two centimeters of the central area (radius 20 cm and thickness 2 cm) of the axial reflector and are quite small, varying from only 1.17 dpa/year with a fuel salt volume of 12 m$^3$ to 0.47 dpa/year with 27 m$^3$. Thus, this is not a limiting factor for our present studies. We did not consider here the dpa due to the fissions occurring near the materials which cause damage on the first few ten micrometers only.

III.B.2 Helium Production

The Helium concentration in the structural materials is directly determined by its production rates through nuclear reactions. This He production depends on the boron and nickel contents of the alloy used as structural material, He appearing through the two reactions: $^{10}$B$(n, \alpha)^7$Li and $^{58}$Ni$(n, \alpha)^{55}$Fe. As shown in Fig.7 and contrary to the case of thermal reactors, the reaction involving $^{58}$Ni is dominant here. Moreover the boron content of the alloy may be easily reduced, the values indicated here being a maximum. The largest acceptable amount of Helium in the alloy is not presently known: the diffusion of Helium at the temperatures involved in the system has not been studied.

Fig. 7. Contributions of the nuclear reactions involved in Helium production in the most irradiated part of the axial reflector (radius 20 cm and thickness 2 cm), as a function of the reactor operating time, for a fuel salt volume of 18 m$^3$

We assumed that the acceptable limit is equal to 100 ppm of He and that there was no He leak by diffusion. The results are displayed in Fig. 8 for a fuel salt volume of 18m$^3$ in terms of reactor operation times necessary to produce this amount of 100 ppm of He in different depths of the axial reflector. This operation time for deeper zones of the reflector being equal to more than 170 years (for 14 cm to 30 cm), it is not indicated in the figure. As a conclusion, regular replacements of the most irradiated area of the upper axial reflector will have to be planned, but this concerns only its first 15 centimeters.

Fig. 8. Operation time necessary to produce 100 ppm of He in different depths of the axial reflector, as a function of the irradiated area considered (zero corresponding to the centre of the core), with a fuel salt volume of 18 m$^3$

III.B.3 Osmium Production

The neutronic simulations of the MSFR reveal the transmutation of the Tungsten contained in the alloy into Rhenium and Osmium through nuclear reactions as detailed in Fig. 9.

Fig. 9. Transmutation cycle of Tungsten, Rhenium and Osmium, due to neutronic captures; the blue boxes represent the unstable nuclei that decay through the purple arrows

The consequences of this effect on the materials resistance are to be investigated; if a loss of less than 1 at% of tungsten is acceptable, then the most irradiated part of the upper reflector will have to be changed every 5 to 10 years as shown in Fig. 10. This part will anyway be regularly replaced because of its Helium content (see section III.B.3).
III.B.4 Summary

We present in Table III the results obtained for the different irradiation damages as a function of the fuel salt volume, in terms of reactor operation times necessary to produce these damages. With the hypotheses taken here in terms of damage limitations, the most constraining effect seems to be the Helium production in the structural materials but this has to be thoroughly studied by specialists.

To conclude, we notice that the damages are inversely proportional to the fuel salt volume of the reactor, thus logically favoring the larger MSFR configurations.

TABLE III
Reactor operation time necessary to produce 100 dpa (second column), 100 ppm of He (third column) and the loss of 1 at% of Tungsten (fourth column) in the most irradiated part of the axial reflector (central area of radius 20 cm and thickness 2 cm), for different fuel salt volumes of the MSFR

<table>
<thead>
<tr>
<th>Fuel salt volume</th>
<th>t(100 dpa)</th>
<th>t(100 ppm of He)</th>
<th>t(-1at% of W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12 m³</td>
<td>85 years</td>
<td>2.2 years</td>
<td>4.7 years</td>
</tr>
<tr>
<td>18 m³</td>
<td>133 years</td>
<td>3.2 years</td>
<td>7.3 years</td>
</tr>
<tr>
<td>27 m³</td>
<td>211 years</td>
<td>5.5 years</td>
<td>10.9 years</td>
</tr>
</tbody>
</table>

III.C Burning rate efficiency

Fig. 11 shows the burning rate\(^\dagger\) obtained for all the transuranic elements after 25, 50, 100 and 200 years of operation, for different fuel salt volumes. Operation times greater than 60 years (a reactor lifespan) are obtained by transferring the salt contained in the phasing out MSFR into a new MSFR. This is made easier because the fuel is liquid.

\(\dagger\) The burning rate is defined as the TRU inventory in the reactor at a given time divided by its initial TRU inventory

Fig. 11. Burning rate of all the transuranic elements after different operation times, as a function of the fuel salt volume

The smallest fuel salt volume configuration, corresponding to the highest specific power, allows faster TRU reduction since more than 90% of the TRUs are burned after only 25 years of operation, and nearly 96% after 50 years of operation. The configurations with larger fuel salt volume also reach this maximal burning rate of 96% but on the longer run, after more than around 100 years of operation.

Finally burning efficiency optimization favors the smaller MSFR configurations.

IV. CONCLUSIONS

The Molten Salt Fast Reactor (MSFR) concept is particularly well suited to fulfill all the criteria defined by the Generation IV International Forum (GIF) and has been selected for further studies by the GIF. We have presented in this paper the MSFR versions started with transuranic elements from the current PWRs. This concept is reliable and robust enough to allow further optimizations, such as the minimization of the fissile inventory and the maximization of the burning efficiency. More specifically, we have evaluated the impact of the specific power on these reactors’ characteristics by varying the total fuel salt volume.

Regarding the neutronic considerations, the reactor doubling times, driving the deployment capacities, favor the medium fuel salt volumes ranging from 12 to 20 m³. The safety parameters are excellent for all fuel salt volumes.

The irradiation damages are inversely proportional to the fuel salt volume of the reactor, thus logically favoring...
the larger MSFR configurations. These damages are dominated by Helium production through the (n,\(\alpha\)) reaction on \(^{58}\)Ni. Two solutions may be considered: either regularly replacing the superficial layers (around 15 cm) of the axial reflectors and the fertile blanket container; or selecting another material without boron or \(^{58}\)Ni. The latter solution may prove more expensive but will be reserved to the more exposed areas. The effects of Helium on the structural materials (maximal acceptable amount, diffusion) have to be thoroughly studied by specialists to confirm our preliminary conclusions. Likewise, the effects of the transmutation to Osmium of the Tungsten contained in the structural materials have to be studied for a better understanding of the long-term materials resistance.

Optimization of the burning capacities of the system favors the configurations with the smaller fuel salt volumes.

As a conclusion, the MSFR configurations containing medium fuel salt volumes, between 15 and 20 \(m^3\), are a good compromise to satisfy all viewpoints (deployment, safety, materials damages and burning capacities). For these configurations, our studies demonstrate that an initial Pu fissile inventory around 4 to 5 metric tons per GWe may be reached even with the limitations assumed for the irradiation damages to the structural materials. Realistic evaluations of the overall possibilities of the MSFR strongly depend on further research in the field of materials.

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