

Thorium Molten Salt Reactor reprocessing unit: Characterization and influence on the core behaviour.

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Portrait
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Abstract :

The Thorium Molten Salt Reactor-Non Moderated (TMSR-NM), a particular configuration of Molten Salt Reactors (MSR), seems to be very promising. It works with a lithium fluoride salt on the thorium cycle without any moderator inside the core. The reactor is coupled with the reprocessing unit described in this paper. Several characteristics of this unit are not quite fixed yet, such as the reprocessing capacity. Our work shows that a core reprocessed within 10000 days is still breeder, its safety coefficients are still strongly negative but the main issue is the valence-3 element proportion. Indeed, the reprocessing capacity should be high enough to keep this proportion below the solubility limit. Still, the high limit (the whole core reprocessed faster than 10000 days) is far from the intense reprocessing considered for previous MSRs. We show that, contrary to past, the reprocessing and the core physics are no longer correlated in this configuration with no moderator inside the core, because of its fast spectrum.

This paper also shows that the radiotoxicity associated to the inventory will always be higher than the radiotoxicity associated to the wastes (hence the importance of end of game scenarios). We studied a scenario where 50 years' operation can reduce radiotoxicity by almost an order of magnitude within 10^4 to 10^5 years of storage.

Introduction:

During past studies, it has been shown that a particular configuration of molten salt reactors could perfectly fulfil the criteria selected by the Generation-4 International Forum. Studies have proved its intrinsic safety and good deployment capacity but the reprocessing is often exposed as a major issue. Together with chemists, we have developed a new reprocessing scheme, which seems to be possible with today's capability. The first part of this document will describe the reactor coupled to the reprocessing unit, a brief overview of the computational tool is also presented. Then in a second part, we will focus on the influence of this reprocessing unit on the core behaviour (breeding ratio, safety parameters, and valence-3 proportion) and finally, in a third part, we will discuss long-term wastes and their associated radiotoxicity.

1. The Thorium Molten Salt Reactor-Non Moderated (TMSR-NM):

a. Neutronic Core

Previous work has isolated a particular configuration of the core, which is really a step forward in the molten salt reactor concept [1,2].The salt plays three roles simultaneously: fuel,

coolant and moderator. A schematic view is shown in figure 1.1: the core comprises a single cylinder whose internal diameter is approximately equal to its height (in our simulations: 2.6m high, 1.25m radius) and where the nuclear reactions occur within the flowing fluoride salt (shown in yellow in Fig. 1.1). Consequently, in order to maintain three confinement barriers, the vessel should contain all the primary coolant circuit (core, heat exchangers and pumps). The neutron flux depends only on of the proportion of heavy nuclei present in the core. Previous systematic studies have led us to choose a lithium fluoride salt with 22.5% of heavy nuclei (mostly thorium and fissile matter). Our simulations are based on a 2.5GW thermal power, which corresponds to 1GW electric power if the nominal temperature is 630°C (thermodynamic efficiency of 40 %). The fertile blanket is a salt whose composition is FLi(72%) and ThF₄(28%). As shown in [1,2] such a TMSR can be started either with uranium 233 or with the transuranic elements produced in a pressurized water reactor, as fissile matter. Because the results presented in this paper, corresponds to reactor steady states, they are independent of the initial fissile matter.

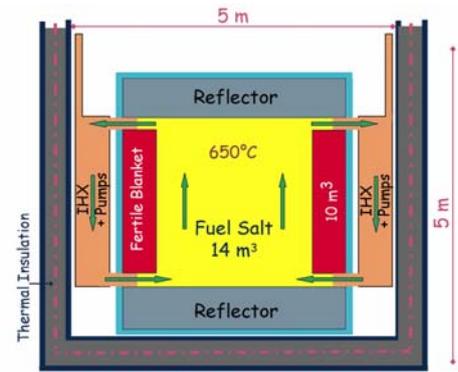


Figure 1.1: schematic view of TMSR

b. Reprocessing unit

Fuel reprocessing and adjustment of the salt composition (redox potential measurement, reactivity...) are necessary to control the operation of a MSR. The whole process is represented in figure 1.2 and can be subdivided in two parts:

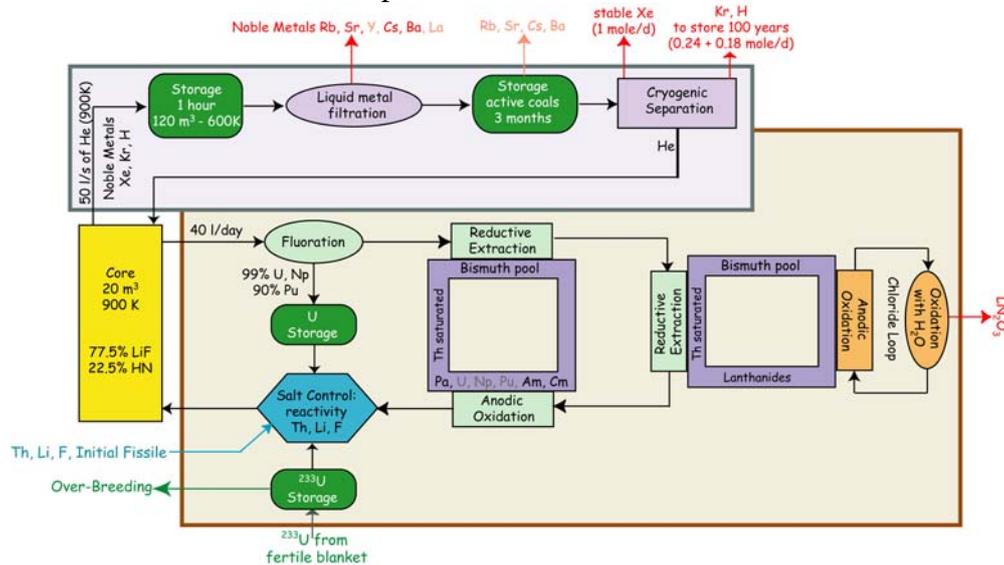


Figure 1.2: reprocessing unit scheme

Salt extraction

A small proportion of the salt fuel can be removed and processed in the power plant in order to extract the fission products and to send back all the actinides in the core. The more powerful the actinide-lanthanide separation, the less actinides will be sent to the waste, the less the long-term associated radiotoxicity will be. This treatment can be broken down in three different steps:

- ✓ First, a fluorination will extract uranium, neptunium, plutonium and a few fission products. Data from ORNL shows that we can expect 99% efficiency for uranium and neptunium and 90% efficiency for plutonium [3].

- ✓ Then reductive extraction is needed to extract the actinides, as the lanthanides cannot be removed without taking the actinides located of the fuel salt only.
- ✓ Finally, a second reductive extraction can extract all the elements other than the solvent to send them to final waste. The lanthanides are transferred on a chloride salt before being oxidised.

Helium bubbling

In parallel, there will be helium bubbling within the primary salt loop in order to remove all insoluble fission products, mostly noble metals and rare gases. The collected aerosol is filtered on liquid metal and then separated cryogenically. The fertile blankets will be reprocessed on a similar but simplified concept in order to extract, within six months, the produced uranium.

c. Calculation means

This work is based on the MCNP neutron transport code [4] coupled with an in-house materials evolution code REM [2,5]. 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. In order to simulate the coupling between the core physics and the reprocessing, we have to modify this equation. We assume that the extraction of nucleus i is proportional to its quantity. So, we can add two terms in the classical Bateman equation to obtain equation (1); one term is for the chemical extraction and the second for a possible source:

$$\frac{\partial N_i}{\partial t} = \sum_{j \neq i} \lambda_{j \rightarrow i} N_j + X_j \langle \sigma_j \phi \rangle N_j - \lambda_i N_i - \langle \sigma_i \phi \rangle N_i - \lambda_{chem} N_i + A \quad (1)$$

where λ_i represents the radioactive constant of nucleus i , X_j the fission product yield, σ_i the neutron capture cross section and A a possible input. As for any nuclear decay, λ_{chem} can be associated to a half-life time. It represents the time needed to extract half of nuclei i . 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. The REM code is a precision-driven code, i.e. it has been designed to calculate the reactor evolution while controlling the precision of the results at each step of this evolution.

2. Influence of the reprocessing on the core behaviour:

a. Salt composition

Reprocessing capacities will directly affect the salt composition. As the salt has to be homogeneous at all times, we give special consideration to solubility issues (of minor actinides and fission products). We give special attention to valence-3 elements (whose solubility limit is close to 5% [6]), because most of the fission products are lanthanides and their solubility is in competition with that of plutonium. As the extraction removes all lanthanides, we expect a strong influence of the reprocessing capacities on this valence-3 proportion. As valence-3 precursors are not extractible by bubbling in most cases, the bubbling time would not affect this proportion. Indeed, we plot the influence of the bubbling time in figure 2.2. and note that the bubbling induces a valence-3 proportion variation of $2 \cdot 10^{-4}$, negligible in front of the absolute proportion (from 0.3% to 4%). Thus, the valence-3 proportion depends only on the lanthanide extraction. To maintain this valence-3 proportion below 5%, the reprocessing time needs to be less than about 10000 days.

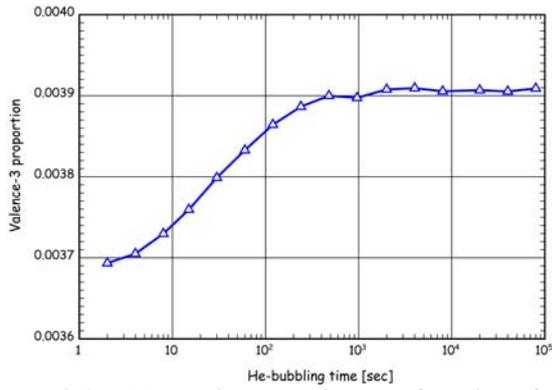


Figure 2.1 : Valence-3 proportion as a function of the bubbling (lanthanide extraction time: 518 d)

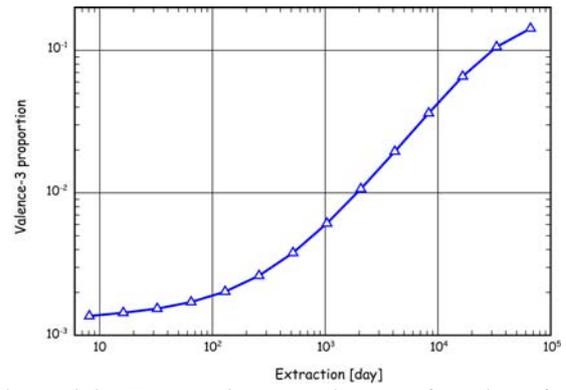


Figure 2.2 : Valence-3 proportion as a function of the lanthanide extraction (bubbling time: 30 sec)

b. Breeding ratio

The breeding ratio expresses the balance between the creation of ^{233}U through neutron capture on ^{232}Th and the destruction of ^{233}U through fission or neutron capture. It is directly linked with neutron capture rates. Lanthanides are the most capturing elements, so we can assume that the bubbling efficiency will not affect the breeding ratio, as He-bubbling doesn't affect the lanthanide population inside the core. We calculated the breeding ratio for different bubbling times, and the results are plotted in figure 2.3: as expected, there is no variation. The lanthanides population is determined by the extraction capacity so the influence of the reprocessing time will be decisive. Calculations show that in order to maintain a breeding ratio larger than 1, the entire fuel salt should be reprocessed in within less than 10000 days, as shown in figure 2.4

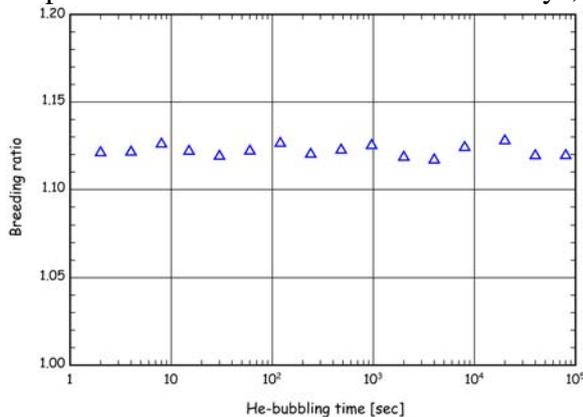


Figure 2.3: breeding ratio as a function of the He-bubbling time (lanthanide extraction time: 518 d)

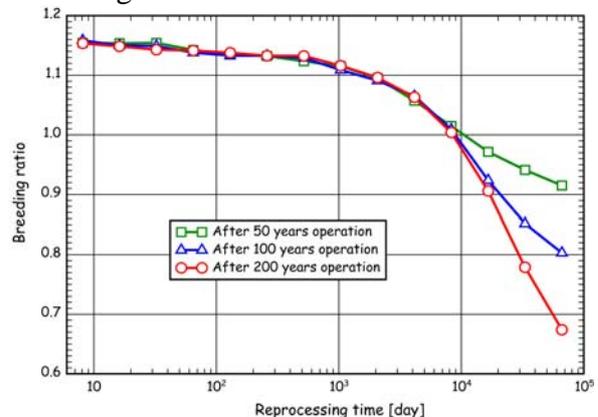


Figure 2.4: breeding ratio as a function of the lanthanide extraction time (bubbling time: 30 sec)

We plot in figure 2.4 the breeding ratio at different operating times: as long as the reprocessing time is small compared to the operating time, there is no difference.

c. Safety considerations

The deterministic safety of nuclear reactors is based on two main parameters: feedback coefficients, which affect reactor stability during temperature variations, and delayed neutrons, which allow reactor control.

1. Feedback coefficients

The feedback coefficients are defined as the variation of reactivity in response to a variation of the core temperature. This coefficient has to be negative to ensure the stability of the reactor. It comprises two terms: the density and the Doppler coefficient. The density coefficient reflects the density variation and can be linked to a void coefficient, while the Doppler coefficient corresponds to the variation of neutronic cross sections with the temperature change.

Because reprocessing changes the salt composition, we can assume that those coefficients

will depend on the reprocessing time. However, the fast neutron spectrum does not vary with the reprocessing, so coefficient dependence on the reprocessing is not so obvious. The doppler and density coefficients are plotted in figure 2.5, clearly showing no noticeable variation with the reprocessing capacities provided the reprocessing is faster than 10000 days.

2. Delayed neutrons

The bubbling could lead to the extraction of neutron precursors outside the core and the fraction of delayed neutrons could thus be lower than expected (360 pcm is classically quoted [1]). Fortunately, all precursors are not removable by the bubbling. Figure 2.6 shows that we lose less than 5% of delayed neutrons with very efficient bubbling, a 30 second bubbling time, as chosen here, implying the loss of less than 1 % of delayed neutrons.

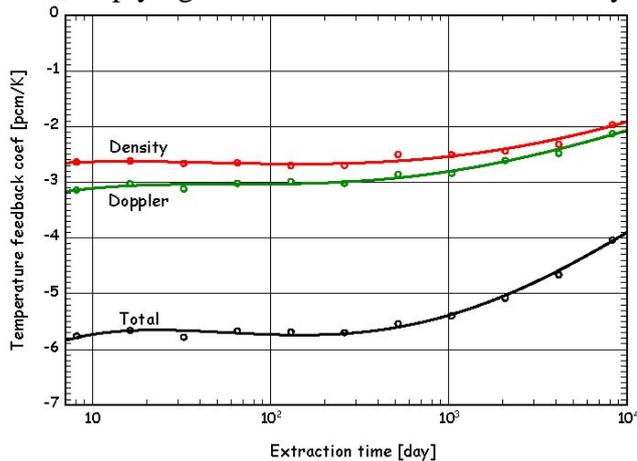


Figure 2.5: Feedback coefficient as a function of lanthanide extraction time

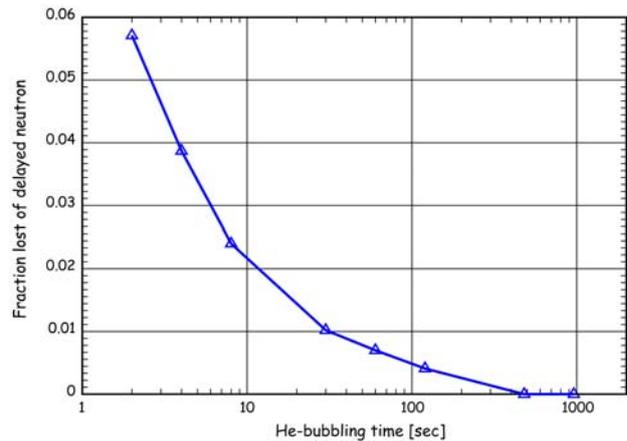


Figure 2.6: Fraction of lost delayed neutrons due to He-bubbling

3. Radiotoxicity considerations:

The purpose of recycling the actinides is to burn them in the reactor. An equilibrium will be reached for actinide quantities leading to the stabilization of the amount of long lived wastes. There are two kinds of wastes: fission products and actinides. With the process defined in paragraph 1, the actinide fraction that goes to the waste is very small: 10^{-5} of uranium and neptunium, 10^{-4} of plutonium and 10^{-3} for heavier nuclei (americium, curium, etc...). Assuming that all fission products can be extracted, the radiotoxicity, normalized to one unit of electricity produced, is plotted in figure 3.1. In this figure, we also show the radiotoxicity associated to the transuranic elements used to start a new TMSR-NM. The radiotoxicity induced by the fission products is directly linked to the number of fissions (hence the electric power) during operation. The radiotoxicity due to actinide wastes depends on the efficiency of actinides extraction. With efficiencies close to 10^{-3} (i.e. 0.1% of actinides are sent to waste) and a reprocessing time on the order of one year, about 10^3 years are necessary to reject the whole incore actinide inventory. Consequently, the radiotoxicity is dominated by the actinide inventory. This consideration holds for all GEN-IV reactors: long lived nuclei stored in the core, are not considered as wastes, but as soon as the reactor is no longer in operation, there is a huge amount of long lived wastes. Hence we should study the end of the cycle (we call this end of game operation).

The principal isotope present in the core after Th-232 is U-233, which is a fissile nucleus. The idea of the end of game is to burn this uranium and as much as possible of the other actinides present in the fuel salt. The heavy nuclei (without Th) of 7 TMSR-NMs are placed in one burner TMSR in order to reach criticality. The inventory, before and after burning is plotted in figure 3.2. We also show the radiotoxicity induced by the wastes of 200 years operation demonstrating that, even after burning, the inventory induced radiotoxicity is dominant.

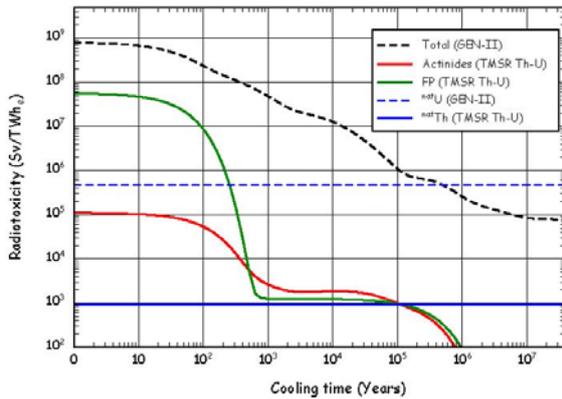


Figure 3.1: radiotoxicity of FP wastes, actinides wastes and actinides used to start a new TMSR-NM

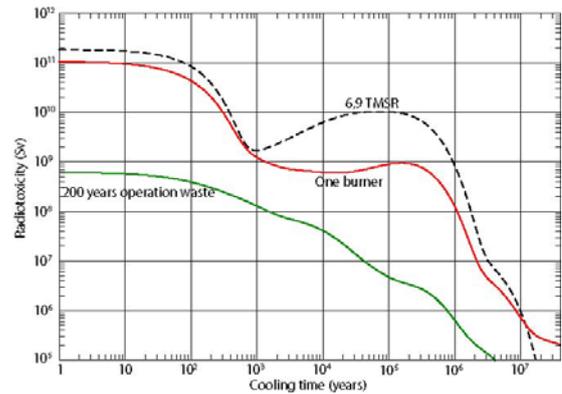


Figure 3.2: radiotoxicity of one burner TMSR-NM, 7 TMSR-NM inventories and 200 years of waste

Conclusion:

The Molten Salt Reactor concept offers great potentialities when combined with the Thorium fuel cycle and a fast neutron spectrum, thanks to its configuration flexibility.

In our simulations of the TMSR-NM, we used a 77.5 mole % LiF – 22.5 mole % (HN)F₄ salt for the fuel, and we studied various reprocessing capacities. As the neutron spectrum is not significantly modified with these capacities, changing the reprocessing time within limits has no effect on the core physics. However, it affects the chemistry directly and especially the solubility of valence-3 elements. In conclusion of this study, a reprocessing time shorter than 10000 days should be fast enough to deal with this issue. We have also shown that the bubbling has no influence on the neutronics but only on the salt physico-chemistry. We need results on bubbling extraction efficiency in order to control insoluble element deposition on structural material.

In a second part, we considered waste production and the associated radiotoxicity. Our conclusion is that the radiotoxicity associated to the waste will be negligible compared to the radiotoxicity associated to the inventory and we insist on the importance of end of game scenarios. We studied a scenario where 50 years operation can reduce radiotoxicity by almost an order of magnitude within 10⁴ to 10⁵ years of storage.

References :

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