Influence of the Reprocessing on Molten Salt Reactor Behaviour

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Abstract: The Molten Salt Reactor is one of the systems studied as a Generation IV reactor. Its main characteristic is the strong coupling between neutronics and salt reprocessing. Such nuclear reactors use a liquid fuel which is also the coolant. Elements produced during the reactor's operation, like Fission Products (FP) or TransUranians, modify the neutronic balance of the reactor by capturing neutrons. As the fuel is liquid, samples can be extracted and reprocessed to remove the poisoning elements, without stopping reactor operation.
This reprocessing includes two components: a bubbling system within the reactor which extracts the gaseous and metallic FPs quickly and a slower external unit that extracts the other FPs. A fluorination removes Uranium to reinject it immediately in the core. The rest of the salt is then treated in a decicated reprocessing unit. A salt volume equal to the core volume is cleaned in several months. We have studied the influence of different reprocessing rates on the reactor's behaviour. This mainly affects the breeding ratio, which represents the ratio of Thorium-232 converted into Uranium-233 over Uranium-233 burnt. By considering both the possibilities in chemistry and the neutronic impacts presented here, our aim is to work out an efficient, reliable and realistic reprocessing scheme.

Keywords: MSR, Thorium, TMSR, Reprocessing, Breeding

1. Introduction

Molten Salt Reactors (MSR) are one of the six systems retained as a candidate for the next generation of nuclear reactors. The on-site chemical reprocessing of such reactors is a major asset of the Molten Salt technology. MSRs are based on a liquid fuel, so that their technology is fundamentally different from the solid fuel technologies currently in use. In the thermal neutron spectrum of a MSR, poisoning due to the Fission Products (FP) being worse than in a fast neutron spectrum, the rate at which fuel reprocessing is performed can become a major issue which will be discussed in this paper.

Our work is based on the coupling of a neutron transport code called MCNP [Briesmeister, 1997] with a materials evolution code. The former calculates 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 in the cells. These calculations take into account the input parameters (power released, criticality level, chemistry,...), by adjusting the neutron flux or the materials composition of the core on a regular basis. All the data presented in this paper result from the evolution of the reactor over 100 years.

2. The Thorium Molten Salt Reactor

2.1 Reactor Definition

In 1964, the Molten Salt Reactor Experiment (MSRE) was initiated at the Oak Ridge National Laboratory (ORNL). Generating 8 MWth of power, the reactor was operated with different fuels (^{235}U then ^{233}U) over several years. The expertise gained during this experiment led, in the 1970s, to the elaboration of a power reactor project, the Molten Salt Breeder Reactor (MSBR) [EDF/DER, 1977]. The studies demonstrated that fuel regeneration is possible with the thorium fuel cycle in an epithermal spectrum, provided very efficient and, as a consequence, constraining, on-line chemical reprocessing of the salt is achieved. Over the past few years, the MSBR has been reassessed in the light of new calculating methods [Lecarpentier, 2001][Nuttin, 2005] so as to elaborate a new reactor concept that we call the Thorium Molten Salt Reactor (TMSR) [Mathieu, 2005].

The standard TMSR is displayed on figure 1. It is a 1 GWe graphite moderated reactor. Its operating temperature is 630 °C and its thermodynamic efficiency is 40 %. The graphite matrix comprises a lattice of hexagonal elements with 15 cm sides. The total diameter of the matrix is 3.20 m. Its height is also 3.20 m. The density of this nuclear grade graphite is set to 1.86. The salt runs through the middle of each of the elements, in a channel whose radius is 8.5 cm. One third of the 20 m^3 of fuel salt circulates in external circuits and, as a consequence, outside of the neutron flux. A thorium and graphite radial blanket surrounds the core so as to improve the system's regeneration capability. We assume that the ^{233}U produced in the blanket is extracted within a 6 month period.

The salt used is a binary salt, 78 % LiF – 22 % $(HN)F_4$ (where HN stands for Heavy Nuclei), whose $(HN)F_4$ proportion (eutectic point) corresponds to a melting temperature of 565 °C. This results in a 1.9 metric ton initial fissile material (^{233}U) inventory. The salt density at 630°C is set at 4.3 with a dilatation coefficient of 10^{-3}/°C [Walle, 2003].

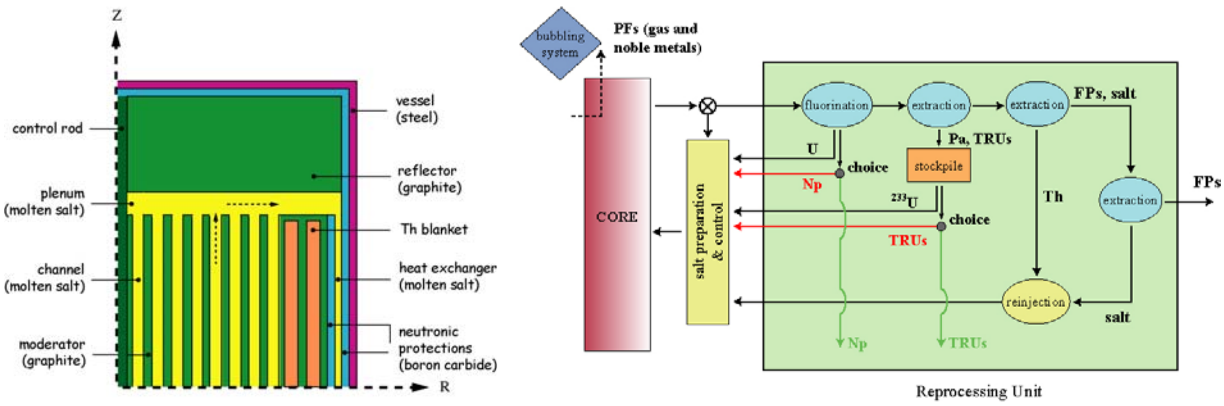

Figure 1: TMSR. Vertical Cut of a quarter (left) and Slow Reprocessing Overview (right)

2.2 Chemical Reprocessing Scheme

The MSBR suffered from major drawbacks and was discontinued. The goal being, at the time, to obtain as high a breeding ratio as possible, the on-line chemical reprocessing unit considered had to process the entire salt volume within 10 days and this was very complex [Walle, 2003]. The project was considered unfeasible. Moreover a very high breeding ratio implies that the excess ^{233}U produced be placed in storage and/or transported. Since the initial fissile matter inventory has to be produced by other means (e.g. in pressurized water reactors or fast neutron reactors) the highest possible breeding ratio does not necessarily have to be sought. As a consequence, nowadays, fast on-line reprocessing is no longer a necessity and a slow reprocessing procedure may be sufficient. In this paper, we discuss the impact of this new chemical reprocessing on the neutronic behaviour of the TMSR, our aim being to work out an efficient, reliable and realistic reprocessing scheme.

2.2.1 The Aims of the Reprocessing

As detailed in reference [Le Brun, 2005], a reactor based on the Th-^{233}U fuel cycle in a thermal neutron spectrum has a tight neutron balance so that minimizing neutron losses is essential. As a consequence, any element that consumes neutrons by capture and is not necessary for reactor operation has to be evacuated. When fissions occur in the molten salt, Fission Products (FPs) and TransUranian elements (TRU) are formed, as shown on figure 3 (left). These elements modify the neutronic balance of the reactor by capturing neutrons, thus degrading the transmutation of ^{232}Th into ^{233}U. As the fuel is liquid, samples can be extracted and reprocessed to remove the poisoning elements without stopping reactor operation.
A precise study of the neutron captures shows that they are consequent in the FPs and that some of these capture more than others.

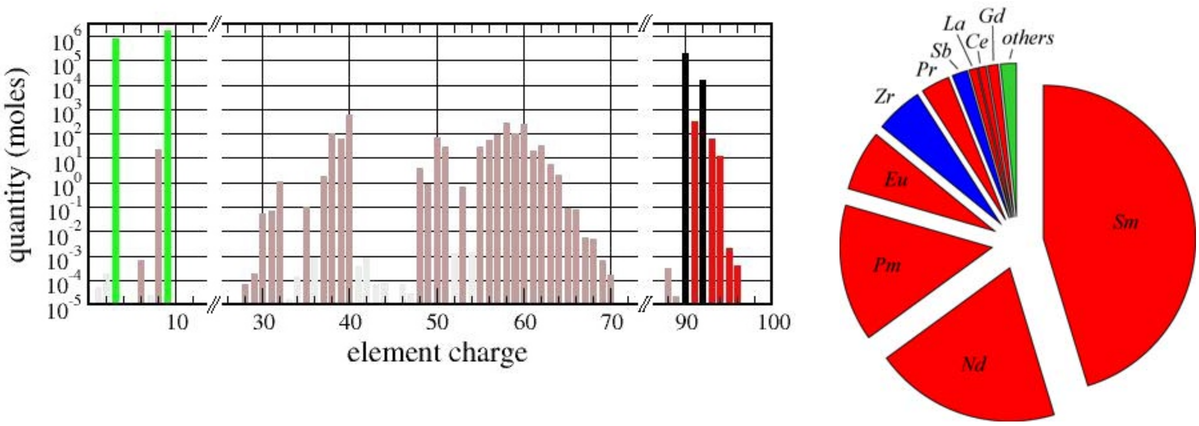

Figure 3 (Left): Composition of the TMSR at equilibrium with Fluoride+ Lithium in green, the FPs in maroon, Thorium + Uranium in black and the TransUranians in red.
(Right): Distribution of the FP capture rate in core after Helium bubbling

Among the FPs still present in the core after Helium bubbling, Samarium has the highest neutron capture rate, as shown in figure 3 (right). The other main poisons, like Neodymium or Praseodymium belong to the same chemical family, the lanthanides. All the elements of this family are drawn in the same colour to highlight their predominant contribution to the neutron captures. Among the other FPs, Zirconium stands out, it accounts for more than 50 % of the remaining captures.

2.2.2 The Slow Delayed Reprocessing Procedure

Figure 2 gives a general view of what slow reprocessing could entail. The reprocessing consists in two parts. The first one is a Helium bubbling system within the reactor which extracts the gaseous and metallic FPs within 30 seconds. The second one ensures a slower extraction of the other FPs, it is external to the reactor. The Uranium is extracted first, by fluorination, and re-injected immediately in the reactor. The remaining salt is treated in a dedicated reprocessing unit. A salt volume equal to the core volume is cleaned within several months.

Some of the stages shown in this general schematic, such as Protactinium storage or TRU extraction, can be eliminated while maintaining the primary assets of the reprocessing. The difficult part of the reprocessing is Fission Product extraction in the presence of Thorium. The idea, with slow reprocessing, is to first extract the Thorium, so as to avoid being handicapped by its presence in the FP extraction process. This method could not be applied in the MSBR because of the large Thorium flow involved, reaching several tons per day while it is only a few hundreds of kilograms per day in the case of a six month reprocessing time.

In addition, with slow reprocessing, the nuclear core can be disconnected from the processing unit, small amounts of the salt being processed individually, instead of resorting to continuous on-line reprocessing, as in the MSBR. This is a source of simplification which allows easier control of the procedure while making the core less sensitive to possible problems in the reprocessing unit.

If the time needed to reprocess the core volume is equal to the time before reinjecting the salt, there is as much salt outside the core as inside it. Thus, 6 months or more can separate the extraction of the fuel salt and its re-injection in the core, after removal of the FPs. The fissile matter inventory is not increased, however, thanks to the possibility of extracting the Uranium during a preliminary fluorination stage. In the case of slow reprocessing, we assume very good extraction efficiencies (they are set to 1 in the calculations) because plenty of time is available. The impact of this reprocessing time on the other reactor parameters of the reactor is discussed in the next section.

3. Influence of the Reprocessing on the TMSR Neutronic Behaviour

The reprocessing time is the parameter which has the greatest impact on the TMSR. In particular, it is important to study its effect on a major constraint for a Generation-IV reactor, the system's fuel regeneration capability.

3.1 Fuel Regeneration Capability

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. The breeding ratio in a critical reactor can thus be written:

$$BR = \frac{r_{c,^{232}Th} - r_{c,^{233}Pa}}{r_{f,^{233}U} + r_{c,^{233}U}} \qquad (1)$$

r_c and r_f being respectively the capture rate and the fission rate of the different isotopes.

A breeding ratio less than 1 implies that ^{233}U is consumed so that fissile matter must be fed into the core on a regular basis. In order to satisfy the regeneration constraint, we try to achieve a breeding ratio at least equal to 1, knowing that any excess neutrons can always be put to use (improved safety, transmutation capabilities ...).

3.2 Impact of the Reprocessing Time

The breeding ratios obtained at equilibrium are given in Table 1 for various reprocessing options as applied to the reactor configuration described previously. The best breeding ratio is obtained with the MSBR reprocessing and the worst with no reprocessing other than Helium bubbling in the core, and ^{233}U recovery in the blanket. The MSBR reprocessing is labelled “fast (10 days)” because of the rate at which the Pa is to be extracted. However, the extraction of the FPs is partial, making the real reprocessing rate longer (equivalent to 50 days for the FPs that capture the most). The ^{233}U stockpile corresponding to the breeding ratios are displayed on figure 4 for the same reprocessing options.

Varying the reprocessing time from 3 months to 2 years induces about a 0.06 loss in the breeding ratio, while the ^{233}U accumulation rate decreases from +20 kg / year (over-breeder reactor) to -40 kg / year (under-breeder reactor). This means that a doubling of the reprocessing time induces a breeding ratio loss of about 0.02 and a reduction of around 20 kg / year in the ^{233}U stockpile.

Table 1: Breeding ratio for several reprocessing options

Reprocessing Time	Breeding Ratio
Fast (10 days)	1.062
Slow (3 months)	1.024
Slow (6 months)	1.000
Slow (1 year)	0.986
Slow (2 years)	0.961
Bubbling only	0.562

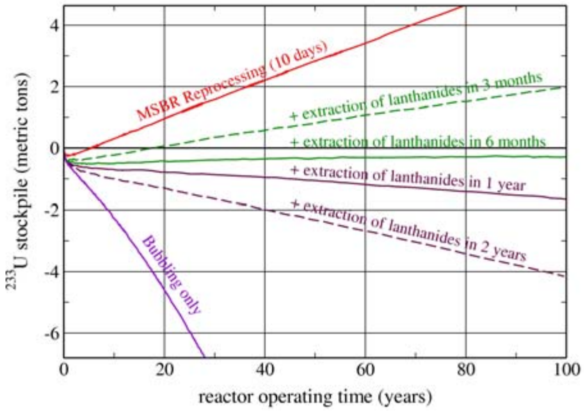

Figure 4: ^{233}U stockpile in the TMSR for different reprocessing rates, compared to the MSBR reprocessing

The change in the breeding ratio and, as a consequence, in the ^{233}U stockpile, is mainly due to the change in the capture rate of the FPs. The good breeding ratio of the fast reprocessing (MSBR) is directly due to the fact that 80 % of the protactinium is stored outside of the neutron flux instead of 30 % for a 3-months reprocessing. This effect has a much larger impact than the FPs and the TRUs in this case. Thus, unless it is extracted rapidly, the Pa's incidence on breeding is minor.

5. Conclusions

The Molten Salt Reactor is a very attractive concept especially for the Thorium fuel cycle which allows nuclear energy production with a very low formation of radiotoxic minor actinides, so that it has been selected by the Generation-IV International Forum. Its main characteristic is the strong coupling between neutronics and salt reprocessing.

In this paper, we presented a reference configuration called the Thorium Molten Salt Reactor (TMSR) and studied the influence of different types of reprocessing on the behaviour of this reactor. Indeed, many different reprocessing schemes are possible, the processing time being one of the adjustable variables. This affects mainly the breeding ratio, which represents the ratio of Uranium-233 consumed over Thorium-232 converted into Uranium-233. The reprocessing time has a negligible impact on reactor safety, on the inventory of fissile materials or the life-time of the structure.

We conclude that is possible to assert that the simplification of the reprocessing presented in this study improves the feasibility of the MSR system. But the reprocessing procedure has to be refined, the possibilities offered by chemistry and the neutronic impacts studied above have to be considered together.

References

M.W.Rosenthal et al., 1970, "Molten Salt Reactors - History, Status, and Potential", Nuclear Applications and Technology, Vol. 8, pp. 107-117

J.F.Briesmeister, 1997, "MCNP4B-A General Monte Carlo N Particle Transport Code", Los Alamos Laboratory report LA-12625-M

A.Nuttin et al., 2005, "Potential of Thorium Molten Salt Reactors: Detailed Calculations and Concept Evolution With a View to Large Scale Energy Production", Progress in Nuclear Energy, Vol. 46, No.1, pp. 77-79

C. Le Brun, 2005, "Molten Salts and Nuclear energy Production", MS7 Conference, Toulouse, France

L. Mathieu, 2005, "Potentialités des réacteurs à sels fondus en cycle Thorium dans les scénarios du futur : le concept de Thorium Molten Salt Reactor", PhD thesis, Institut National Polytechnique de Grenoble, France (in French)

EDF/DER, 1977, "Analyse critique du projet MSBR", internal report HT-12/24/77 (in French)

E.Walle, J.Finne, G.Picard, S.Sanchez, O.Conocar, J.Lacquement, 2003, "Molten Salt Reactors: Chemistry of Fuel Salt and Fuel Salt Cleanup", Global, New Orleans, USA

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