



HAL
open science

Thorium molten salt reactor : from high breeding to simplified reprocessing

L. Mathieu, D. Heuer, A. Nuttin, F. Perdu, A. Billebaud, R. Brissot, C. Le Brun, E. Liatard, J.M. Loiseaux, O. Meplan, et al.

► To cite this version:

L. Mathieu, D. Heuer, A. Nuttin, F. Perdu, A. Billebaud, et al.. Thorium molten salt reactor : from high breeding to simplified reprocessing. GLOBAL 2003 - Nuclear Science and Technology : Meeting the Global Industrial and R&D Challenges of the 21st Century, Nov 2003, New Orleans, United States. pp.1863-1872. in2p3-00020302

HAL Id: in2p3-00020302

<https://hal.in2p3.fr/in2p3-00020302>

Submitted on 22 Jan 2004

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Thorium Molten Salt Reactor : from high breeding to simplified reprocessing

L. Mathieu, D. Heuer, A. Nuttin, F. Perdu, A. Billebaud, R. Brissot, C. Le Brun, E. Liatard, J.-M. Loiseaux, O. Méplan and E. Merle-Lucotte, Laboratoire de Physique Subatomique et de Cosmologie , 53, avenue des Martyrs, F-38026 Grenoble Cedex, France
e-mail : mathieu@lpsc.in2p3.fr

S. David, IPN Orsay CNRS/IN2P3, 91406 Orsay Cedex, France

C. Garzenne, D. Lecarpentier, EDF-R&D, Département SINETICS,
1 av du Général De Gaulle, 92140 Clamart, France

Abstract

The Molten Salt Reactor (MSR) needs to be coupled to a reprocessing unit in order to extract the Fission Products which poison the core. The efficiency of this reprocessing has a crucial influence on the reactor behavior especially for the breeding. The Molten Salt Breeder Reactor (MSBR) project was based on an intensive reprocessing for high breeding purposes.

The aim of this paper is to demonstrate that a Molten Salt Reactor can be self-regenerator with a simplified reprocessing, and to present safety studies. The chemical separation is slowed down and realized in about one year instead of only several days, moreover extracted elements and extraction efficiencies are different from the MSBR reprocessing.

Introduction

Electricity nuclear production is an attractive way to cover a significant part of the future world energetic needs. Present Pressurized Water Reactor (PWR) would be able to satisfy such a need but the consequences would be critical. Indeed consumption of a great part of the uranium resources and production of high quantities of nuclear wastes are hardly acceptable.

The Thorium Molten Salt Reactor (TMSR) may contribute to solve these problems. The thorium cycle produces much less TRansUranic elements (TRU) than the U-Pu cycle and uses a more abundant resource. This cycle can also be used in fast spectra, but the fissile inventory is much lower in epithermal spectra and this property facilitates greatly a park deployment. ^{233}U initially needed to start-up a reactor must be produced, which implies to use plutonium in solid fuel reactors like PWR or FNR.

The ability to extract Fission Products (FP) is an advantage of the MSR which enables the breeding. Without any reprocessing, the fission chain stops very quickly. But on the contrary, the MSBR very efficient reprocessing may appear too ambitious for energy production. So our goal is to find some configurations of reactor and reprocessing to obtain a self-regenerator system.

The studies are based on the coupling of an evolution program and the Monte-Carlo code MCNP [1]. This code calculates cross sections and reaction rates in all cells, and the evolution code resolves the Bateman equations for the cell compositions. These calculations take into account some constraints (power, criticality, chemistry...) by adjusting regularly the neutron flux or the core composition.

After a description of the MSR concept and particularly the MSBR reprocessing, we present the new ways of reprocessing and the impact on the breeding. Next comes a way to improve breeding by changing the power density. Eventually, we present what these two major changes imply on safety and which solutions can be developed to obtain negative temperature coefficients.

1 The MSR general concept

1.1 Main characteristics

The molten salt used as fuel and coolant circulates in a channel network through a graphite matrix as shown on Figure 1 [2]. The geometric characteristics of the reactor are given in Table 1. The fuel used is a fluoride which composition is : 70% ^7LiF - 17.5% BeF_2 - 12.5% $(\text{HN})\text{F}_4$ (where HN means Heavy Nuclide). Initially HN inventory is composed of 1.5% of ^{233}U and the remaining is ^{232}Th .

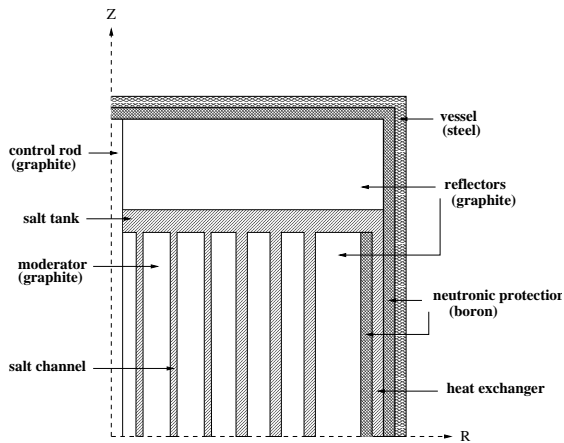


Figure 1: Schematic view of the simulated TMSR

There are two different channel radii which implies two different zones in the core. The inner “fissile” zone where the channels are smaller is more thermalized while the spectrum in the outer “fertile” zone is harder. This splitting into two different zones aims at favoring breeding by specializing the zones [3, 4].

The graphite has a density of 1.86 contrary to some previous studies made on the MSBR concept which density was about 2.3 [3, 4, 5]. This density value seems to be more realistic for graphite.

The MSBR we have simulated does not have exactly the same characteristics than the original MSBR concept, so it will afterwards be called MSBR-like.

1.2 Reprocessing

1.2.1 Modelization

The extraction rate of a chemical element is simulated by a pseudo-decay constant $\lambda = \frac{\epsilon}{T}$ where ϵ is the reprocessing efficiency and T the time in which the whole reactor salt volume is reprocessed.

So, the extracted flow of an element e is given by the following formula :

$$Q_{reprocessed} = \frac{\epsilon}{T} \cdot N_e \quad (1)$$

with N_e the amount of the element in the core.

1.2.2 Bubbling system

The first stage of the salt reprocessing is the bubbling system which extracts gaseous and metallic FP. These FP are dragged with an He gas bubbling and collected outside on filters. If these FP were not extracted they would quickly poison the core (mainly due to the high capture rate of Xe).

Table 1: Main characteristics of the TMSR

Core radius	2.3 m
Core height	4.6 m
Fissile zone radius	1.45 m
Hexagon side size	15 cm
Fuel salt density	3.3
Fissile zone channel radius	5.5 cm
Fertile zone channel radius	8.45 cm
Fissile zone channel number	138
Fertile zone channel number	114
Salt volume in the core	26.7 m ³
Salt volume out of the core	13.3 m ³
Initial fissile nucleus	²³³ U
Initial fertile nucleus	²³² Th
Initial fissile inventory	950 kg
Initial fertile inventory	58 tons
Axial reflector thickness	50 cm
Radial reflector thickness	130 cm
Reflector material	graphite
Moderator material	graphite
Graphite density	1.86
Mean core temperature	900 K
Thermal power	2500 MW
Electrical power	1000 MW

This process is crucial for the reactor operation and is present in all simulations as a basic operation. We take a period of 30 seconds for this process.

1.2.3 MSBR reprocessing

The whole salt volume is treated online by a nearby reprocessing unit in $T = 10$ days. The process is separated into several successive stages [5]:

- fluorination of U and Np (mainly) which are extracted and sent back into the core
- extraction of TRU and Pa which are stored until ²³³Pa decays into ²³³U
- extraction of FP (especially Lanthanides)
- sending back the remaining TRU and ²³³U formed into the reactor

The separation of Pa (with other TRU) enables to put it out of the neutron flux. So it decays into ²³³U without capturing neutron and forming ²³⁴Pa which decays into ²³⁴U.

FP are extracted with efficiencies which depend on their chemical properties. For example Lanthanides are extracted with $\epsilon = 20\%$ [2].

This reprocessing is considered as a fast reprocessing because of the time taken to treat the whole core, and as very efficient since the FP quantity in core is stabilized at a very low value (211 kg) and because of the Pa separation.

Moreover, all HN have to be thoroughly returned to the core. Indeed, the proportion of HN which are removed with FP and put to waste must be minimized. Losses are unavoidable and a small part of HN is thrown away each time the salt is reprocessed. We suppose reprocessing losses of 10^{-5} for HN in all simulations.

1.2.4 Feeding mode

In order to keep the criticality, fissile material must be added or extracted of the fuel salt. This adjustment is achieved either with the isotopic vector of uranium from the fluorination or with the ^{233}U from Pa decay. The results we present are all made under the second hypothesis.

In addition to this feeding the disappeared thorium is replaced at a rate of approximately 1 ton per year. More precisely, we impose the total HN inventory to be constant in order to keep the same physico-chemical salt properties all along the calculations. So the thorium inventory slowly decreases as far as the TRU inventory grows until equilibrium.

1.3 Discussion on the MSBR concept

1.3.1 Breeding capacity

With such a reprocessing the MSBR is a good breeder reactor. The MSBR-like reactor we simulate breeds about 34 kg of ^{233}U per year in average over one century. It implies that the doubling time, the time needed to start a new reactor with bred ^{233}U , is about 24 years. Such a doubling time enables to grow up a nuclear park in a few decades.

However start-up is a problem since ^{233}U does not exist initially and must be produced before.

1.3.2 Reprocessing constraints

The results of the MSBR project are very conclusive, due to the efficiency of the reprocessing. Unfortunately, such a reprocessing seems to be very difficult to develop because of its complexity, mainly due to the succession of specialized processes managing all elements (FP, Th, Pa, U, TRU) in 10 days.

In order to minimize the outside inventory, the time spent in the reprocessing unit has to be short in comparison with the time to reprocess the whole salt volume.

Pa storage itself is a problem because of the heat from radioactive decay. The power induced by 80 kg of ^{233}Pa decaying in ^{233}U is about 4.6 MW and such a power must be managed [4].

HN have to be separated first because it is the only way to extract Pa and because they are less stable than FP from a chemical point of view. Remaining HN in the reprocessed salt have to be minimized to reduce the reprocessing losses during FP extraction.

The large amount of Th in the salt complicates deeply the reprocessing since it must not be extracted with FP and since it is difficult to manage such a quantity in the different steps of the reprocessing [6].

All the technological constraints linked to a 10-day reprocessing make the system too complex to be really competitive as a generation-IV energy producer.

1.3.3 The problem of temperature coefficients

The MSBR is a very good breeder reactor concept, however it remains problems on temperature coefficients. The temperature coefficient $\left(\frac{dk}{dT}\right)$ characterizes the evolution of reactivity for a temperature change. For stable operation and safety this temperature coefficient has to be negative.

Various configurations were studied in the MSBR project. Temperature coefficients were found to be rather negative, but are in contradiction with the MSBR-like we simulated which coefficient is found slightly positive. The difference may be explained by calculation codes used for the MSBR which supposed an homogeneous mix of salt and graphite [4].

2 New reprocessing schemes

2.1 A new way of thinking

2.1.1 The major change

Without the constraint on the extraction of a significant amount of Pa, the reprocessing may be greatly slowed down. The whole core volume may be treated in six months or one year instead of 10 days. It deeply changes the way of thinking the reprocessing. We simulate other reprocessings based on this new point of view.

2.1.2 Principle

The simplified process is separated into several successive stages which do not have to be all achieved in a nearby chemical unit. On the reactor site are the following steps :

- fluorination of U and Np (mainly)
- waiting several months to let the Pa decay into ^{233}U
- fluorination of ^{233}U formed

The remaining salt, containing Th, TRU and FP, is sent in a specific and centralized reprocessing plant. The purpose is to recuperate the main part of thorium and salt components (Li, Be and F). Contrary to the MSBR, reprocessing time is no longer a problem and several ways can be studied for this process.

The first fluorination stage must be done as quickly as possible to return U into the core, otherwise the inventory of U would dramatically rise. In the same way but in a least importance, Th and salt components must be recovered as com-

pletely and quickly as possible to avoid increasing both fertile inventory and salt volume in the fuel cycle.

2.1.3 The two options for a simplified reprocessing

The fluorinated Np and the TRU extracted in the centralized plant may have two different destinations. So we consider two new reprocessing options as shown on Figure 2 :

- the standard offline reprocessing option : after fluorinations, FP are extracted from the remaining salt and consequently thorium and TRU return to the core. TRU inventory reaches an equilibrium value.

- the minimum offline reprocessing option : after fluorinations, thorium is extracted from the remaining salt by oxydo-precipitation. The management of the mix of TRU and FP is not detailed, but TRU do not return to the core.

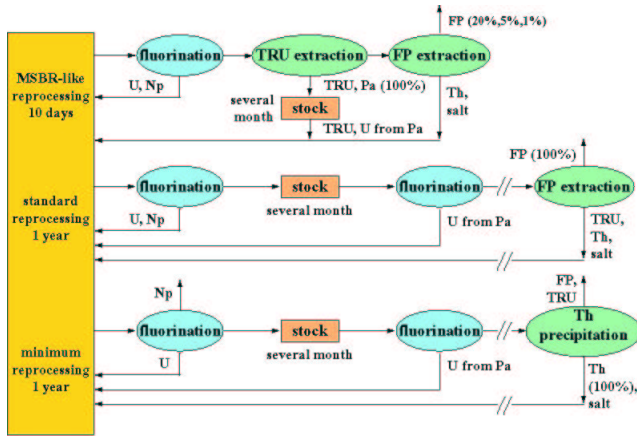


Figure 2: Schematic view of the three compared reprocessing schemes

Figure 2 presents the three reprocessing schemes with the different steps, the time taken to treat the whole salt volume and the efficiency of FP, TRU and Th extraction.

2.2 Simulation results

2.2.1 Stockpile of fissile materials

The fissile stockpile is defined as the balance between feeding and extraction of fissile material, resulting from criticality adjustment : extraction leads to a positive stockpile (breeding) while feeding implies a negative stockpile (needs).

2.2.2 Fissile stockpile for the different reprocessing schemes

In this sub-section we compare the results of our simulation for a standard reactor coupled with three different reprocessing schemes : MSBR-like reprocessing (10 days), standard

offline reprocessing (1 year) and minimum offline reprocessing (1 year and 6 months). The case of a reactor operating with only bubbling system is represented too. Figure 3 shows the evolution of fissile materials stockpile over a period of one century.

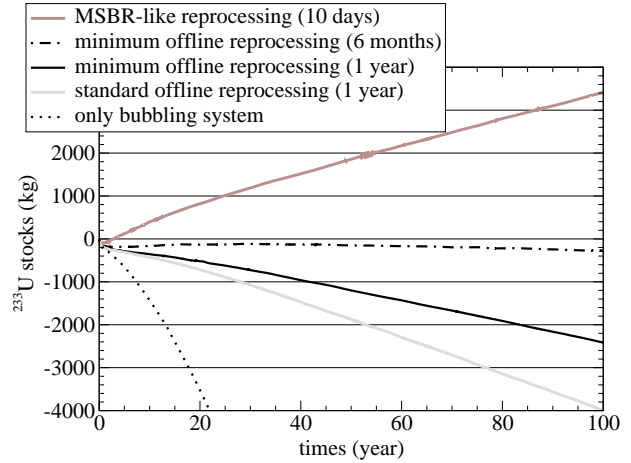


Figure 3: Stockpile of ^{233}U for several systems (1 GWe reactor with standard power density)

As it was said, a reactor with an MSBR-like reprocessing is a breeder reactor with an ^{233}U production rate of 34 kg/year. On the opposite, reactors with the slower reprocessing need continuous feedings of fissile material. However, these needs are far lower than those of a reactor without reprocessing (only with the bubbling system). In this case, the needs in ^{233}U reach 1 ton in 8 years instead of 28 years for the standard offline reprocessing and 42 years for the minimum offline reprocessing. The reactor with the 6-month minimum offline reprocessing is even about self-regenerator and so corresponds to an acceptable system.

2.2.3 Discussion

The extraction of TRU stabilizes quickly their inventory at a low value. On the contrary, with the MSBR-like or standard reprocessing, the TRU inventories grow during 50 years before reaching their equilibrium. There is a large difference between the TRU inventories and we can compare the amount of neutrons that TRU absorb by capture with the amount they create by fission. Table 2 presents the capture and fission rates of these TRU.

The neutron balance is always negative and is equal to -0.024 n/fission in the first case and -0.008 n/fission in the second one. Although these nuclides undergo fission, they absorb more neutrons than they produce. So their extraction favors the neutronic balance and then the breeding as shown on Figure 3.

Table 2: Capture and fission rates of TRU (n/fission) with average number of neutron emitted per TRU fission (for a 1-year reprocessing)

	$\tau_{capture}$	$\tau_{fission}$	$\langle \nu_{TRU} \rangle$
standard reprocessing	0.069	0.023	2.93
minimum reprocessing	0.012	0.002	2.86

2.2.4 Heavy Nuclides inventories

Table 3 presents the core inventories for the different Heavy Nuclides after 100 years in reactor as a fonction of the simulated system. Table 4 presents the amount of material extracted from the salt.

Table 3: Core inventory of Heavy Nuclides and FP after 100 years (kg) for different reprocessing schemes (10 days for MSBR-like and 1 year for the others)

	MSBR-like	standard	minimum
Th	57 000	56 600	56 800
Pa	22.1	94.6	89.6
U	1 790	2 140	2 050
Np	34.6	44.9	14.5
Pu	49.2	66.7	5.93
Am	3.11	4.03	0.023
Cm	13.5	16.7	0.012
Bk	0.008	0.010	8e-06
Cf	0.071	0.080	7e-08
TRU	100	132	20.5
FP	211	419	421

We can see on that table the different effects of the new reprocessing options compared to the MSBR-like one :

- Pa is not extracted quickly and the inventory staying in the core is higher than for the MSBR reprocessing

- less FP are extracted so their inventory is higher and the reactor needs more uranium to reach criticality

- if TRU are extracted (in minimum reprocessing), their inventory decreases greatly, and elements of higher Z than 94 are almost inexistant

- if TRU are sent back into the core, their inventory are quite the same than with MSBR-like reprocessing. The difference is mainly due to the higher ^{233}Pa inventory which produces ^{234}U and then all the others TRU. ^{234}U is also formed by capture on ^{233}U which inventory is higher in the standard reprocessing.

2.2.5 Heavy Nuclide losses

Even when all Heavy Nuclides return into the core, reprocessing losses can not be avoided and generates waste. These quantities are proportionnal to the reprocessing losses coefficient and inversely proportionnal to the reprocessing time. We suppose in all simulations a coefficient of 10^{-5} and this leads to the results of Table 4. Results for different reprocessing times or reprocessing losses can be deduced easily from this table.

Concerning the minimum offline reprocessing only U and Th return to the core so the outside inventory of these elements are dominated by the reprocessing losses. The other elements are extracted from the salt and they accumulate outside the core.

Table 4: Inventory of Heavy Nuclides out of the core after 100 years (kg) for different reprocessing schemes (10 days for MSBR-like and 1 year for others)

	MSBR-like	standard	minimum
Th	2 080	56.8	57.0
Pa	0.033	0.001	42.9
U	63.9	2.14	2.06
Np	1.07	0.037	1 200
Pu	1.25	0.044	373
Am	0.133	0.005	10.3
Cm	0.182	0.006	0.293
Bk	4e-06	1e-07	9e-09
Cf	3e-04	1e-05	5e-07
TRU	2.63	0.091	1 590

We can check on Table 4 that reprocessing losses in the standard reprocessing are more than 30 times lower than with the MSBR-like reprocessing (the exact factor is $\frac{365}{10}$) so they are minimal with this reprocessing.

In the minimum reprocessing case, there is a large amount of HN out of the cycle due to the fact that TRU are not consumed in the core. To make a fair comparison, we have to compare the sum of inventories (in core and out of the core). Then, we can see that Np quantity is greatly increased when TRU are extracted (1.2 tons instead of several tens of kilograms). In the same way, there is 5.5 times more Pu when TRU are extracted than when they are not, and this factor is about 2 for the Am. On the contrary, there are much less Cm, Bk and Cf because TRU are significantly extracted and do not capture enough neutrons to form these elements.

The production of these TRU (making spontaneous fission) in the minimum reprocessing case, with less than 10^{-4} mol% of Cm, is easily managed in comparison with the current stocks.

Moreover, these quantities are still very low in comparison

with the TRU production of a PWR. The TMSR production can be integrated in the forthcoming management of large quantities of TRU coming from PWR.

In summary, the so-called “minimum reprocessing” appears as a very promising way for the future since self-regeneration is easier and since it can greatly simplify the reprocessing while its impact on extracted material management is marginal.

3 Variation of power density

The power density is a reactor parameter we can change independently of the reprocessing in order to optimize further the neutron economy. This study enables us to set up systems (reactor + reprocessing) according to our constraints : breeding, safety and simplified reprocessing.

3.1 New initial characteristics

3.1.1 Variation of the size

There are different ways to make a variation of the power density and we choose to increase the size of the core keeping the total power constant. We made a so called “size 2” reactor which has a double salt volume.

The radius and the height of the core is increased by a factor $2^{\frac{1}{3}}$, but the channel are kept the same in order to have the same neutronic and hydraulic behavior than before. Then the core contains 198 channels in the fissile zone and 222 in the fertile one instead of respectively 138 and 114 for the standard “size 1” reactor. Due to lattice effect the effective volume variation of size “2” is in fact 2.24.

3.1.2 Variation of the inventories

As there is twice more salt in a size 2 reactor and as the salt keep the same initial composition, the fissile and fertile inventories are twice higher too. It means that such a reactor needs about 2 tons of ^{233}U and 130 tons of ^{232}Th to be started.

3.1.3 Variation of the reprocessing time

The reprocessing capacity is the salt mass which can be reprocessed by time unit. To keep the same capacity for the size 2 reactor we have to double the time in which the salt volume is reprocessed.

3.2 Stockpile of fissile materials

We can see on Figure 4 that, for both reprocessing schemes, the size 2 reactor has better breeding characteristics. The average ^{233}U formation is about 70 kg/year for the MSBR reprocessing and 40 kg/year for the minimum offline reprocessing.

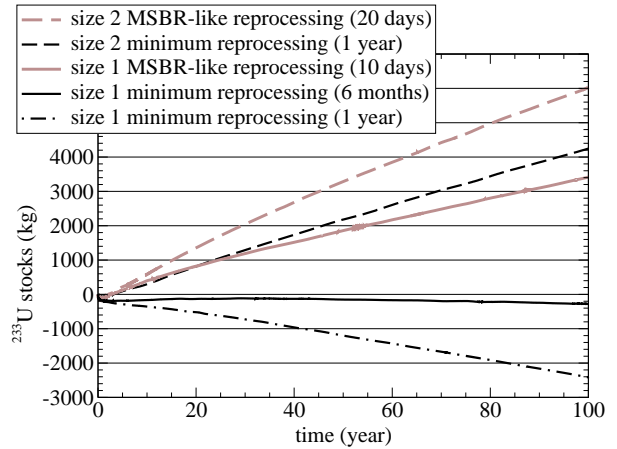


Figure 4: Stockpile of ^{233}U for several systems

As a comparison, these values are respectively 34 kg/year and about 0 kg/year for the size 1 reactor.

3.3 Influence of the power density

3.3.1 Neutron flux

These differences are mainly due to the change of the neutron flux induced by the power density variation. As the salt volume is doubled but the total power kept constant, the neutron flux is divided by a factor two.

All reaction rates are given by the formula :

$$\tau = N \cdot \sigma \cdot \phi \quad (2)$$

In a first approximation we can say that the cross section variation is low enough to be ignored. As the neutron flux is divided by a factor two, the only important parameter to discuss is the amount N of an element in the salt.

3.3.2 Evolution of the capture and fission rates

Among the different chemical elements, some have their inventory doubled while others stay at their initial value. Table 5 shows the neutron balance for two different size of reactor in order to point out the reaction rate variations.

Additionally to the reduction of neutron leakage, increasing the reactor size greatly reduces ^{233}Pa capture rate. Indeed, its inventory is the same in both case contrary to the other elements which inventories are doubled. As ^{233}Pa capture rate is lower, less ^{234}U is formed (just as heavier U isotopes) and their capture rate is reduced too. Neutrons which are no longer captured on these elements participate to the breeding by captures on ^{232}Th .

Moreover, as the neutron flux is twice lower and the flux shape is the same, the graphite life time is doubled. But as the amount of graphite needed to build a size 2 reactor is twice

Table 5: Neutron balance *(n/fission) after 100 years in reactor for size 1 and size 2 with minimum offline reprocessing (respectively 6 months and 1 year)

	size 1	size 2	difference
^{233}U	0.107	0.107	=
Th	1.015	1.060	+0.045
Pa	0.024	0.013	-0.011
FP	0.034	0.032	-0.002
TRU	0.006	0.005	-0.001
other U	0.179	0.161	-0.018
graphite	0.059	0.054	-0.005
salt	0.047	0.050	+0.003
leakage	0.020	0.009	-0.011
(n,xn)	0.011	0.011	=
$\langle v \rangle$	2.480	2.480	=

bigger, the amount of graphite to treat is kept constant. However, the time between two graphite replacement is doubled.

3.3.3 Reactions rate and breeding ratio

The breeding ratio is an adimensionnal value which represents the number of ^{233}U nuclei formed over the number disappeared :

$$\tau_{breeding} = \frac{\tau_{capture}(^{232}\text{Th}) - \tau_{capture}(^{233}\text{Pa})}{\tau_{capture}(^{233}\text{U}) + \tau_{fission}(^{233}\text{U})} \quad (3)$$

Table 6 presents the calculation of the breeding ratio for the size 1 and size 2 reactor with minimum offline reprocessing (respectively 6 months and 1 year).

Table 6: Reaction rates (n/fission) and breeding ratio for a size 1 and a size 2 reactor with minimum reprocessing

	size 1	size 2
capture Th	1.015	1.060
capture Pa	0.024	0.013
capture ^{233}U	0.107	0.107
fission ^{233}U	0.887	0.896
$\tau_{breeding}$	0.997	1.044

4 Temperature coefficients and breeding ratio

4.1 Generalities

Breeding is a priority aspect of a TMSR, but safety is without contest an important point to address. The reactor has to be stable without human or automatic intervention. Such a constraint is quantified by the total temperature coefficient $\left(\frac{dk}{dT}\right)$ which represents the variation of reactivity following a variation of temperature. As previously said, this coefficient must be negative in order to prevent that an increase of temperature implies an increase of reactivity.

The total temperature coefficient can be splitted into several terms which represent different phenomena. As the cross terms are negligible the formula becomes :

$$\left(\frac{dk}{dT}\right) = \left(\frac{dk}{dT}\right)_{\text{Doppler salt}} + \left(\frac{dk}{dT}\right)_{\text{salt density}} + \left(\frac{dk}{dT}\right)_{\text{graphite}} \quad (4)$$

- the Doppler effect characterizes the widening of capture or fission resonances. Thorium cross section has its first resonances around 20 eV and it implies a decrease of reactivity ; the Doppler coefficient is negative.

- the density effect is due to the fact that a small part of salt is ejected from the core when its density decrease. On the contrary graphite does not expand, the spectrum is more thermalized and the reactivity increases since the reactor is under-moderated ; the density coefficient is positive.

- the graphite effect concerns a shift of the thermal part of the neutron spectrum due to the temperature increase. The fission cross section of ^{233}U has a shoulder starting at 0.2 eV, while the Th capture cross section slope does not change. The spectrum hardening implies then an increase of reactivity and the graphite coefficient is positive.

It is important to point out that the total salt coefficient is negative since Doppler coefficient is dominant. Moreover the graphite takes a "long" time to heat (several minutes to several hours), and its positive temperature coefficient is not effective immediatly.

A reactor with a positive total coefficient and a negative salt coefficient can then be managed. But such a reactor is not intrinsically stable, and further studies have to be made to improve these coefficients.

Can we have at the same time breeding, simplified reprocessing and intrinsic safety? All temperature coefficients we obtain for the several presented systems are slightly positive as shown in Table 7. However, these results are difficult to interpretate according to the uncertainties (at least ± 0.05 pcm/ $^{\circ}\text{C}$). Effects involved are quite low and differences between coefficients are not significative enough. Moreover, un-

certainties on data base cross sections prevent us from doing more precise calculations.

Table 7: Total temperature coefficient (pcm/°C) for presented systems

	$\left(\frac{dk}{dT}\right)$	$\tau_{breeding}$
size 1 MSBR-like (10 days)	0.30	1.032
size 2 MSBR-like (20 days)	0.69	1.056
size 1 standard (1 year)	0.26	0.955
size 1 minimum (1 year)	0.44	0.975
size 1 minimum (6 months)	0.32	0.997
size 2 minimum (1 year)	0.74	1.042

In order to obtain negative coefficient, other ways must be studied.

4.2 Erbium

A solution is to put some poison in the core [7]. This poison may be placed in the graphite matrix, in several bars surrounding each channel. We have chosen to limit the place of these bars to the fissile zone only, since the power density is maximum there.

The most promising poison is the ^{167}Er thanks to a great resonance of 10 000 barns at about 0.5 eV. This situation improves mostly the graphite temperature coefficient, since spectrum hardening implies more captures in the erbium resonance.

To simulate this system, we use the geometry and the equilibrium salt of a size 1 reactor with MSBR-like reprocessing. Six bars which radius is 1 cm surround each channel in the fissile zone. These bars are made of graphite with an ^{167}Er concentration of 10 ppm. So there is about 300 g of ^{167}Er in the reactor which corresponds to an antireactivity of 620 pcm. To adjust the criticality, we have to add 3% of uranium to the core (54 kg of U which contains 30 kg of ^{233}U).

Table 8 shows the total temperature coefficient and the breeding ratio with erbium bars in comparison with the standard case.

Table 8: temperature coefficient (pcm/°C) and breeding ratio for erbium study in a size 1 with MSBR-like reprocessing

	$\left(\frac{dk}{dT}\right)$	$\tau_{breeding}$
without Erbium	0.30	1.032
with Erbium	-0.60	1.022

This method is in agreement with other results [7], showing that we can obtain a negative total temperature coefficient

by keeping breeding characteristics, however the system simulated included the efficient MSBR-like reprocessing. This solution is limited while breeding mode is not given up.

Meanwhile, the erbium capture rate reaches 20 g/day, and a significant part of the poison has disappeared only in a few days. The erbium inventory management has thus to be further studied.

Another solution may be to put erbium directly in the salt. The effect on the different temperature coefficients have to be evaluated, but the erbium renewal is much easier than with bars in graphite.

4.3 Influence of channel radius on temperature coefficients and breeding ratio

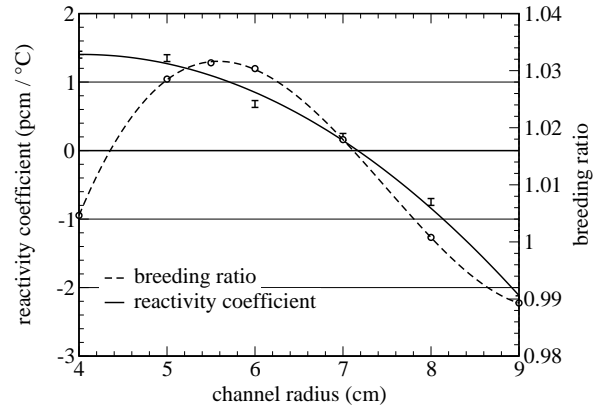


Figure 5: Temperature coefficient and breeding ratio as a function of the channel radius

The total temperature coefficient and the breeding ratio depend on the channel characteristics [3]. Figure 5 shows the influence of the channel radius on these parameters. This study was made by using the equilibrium of a reactor with only one zone and MSBR-like reprocessing, but the results are valid for the other systems.

We can see that the total temperature coefficient becomes negative for large channels as the breeding ratio decreases. The neutron flux is maximal in the central zone and the associated total temperature coefficient has more weight than the one of the peripheral zone. By placing large channels in the central zone, it improves the reactor stability but degrades the breeding. For this reason we simulated a size 2 reactor which zones are inverted and called afterwards “inverted size 2”. This reactor has 198 channels in the central fertile zone and 222 in the peripheral fissile zone. Another reactor called “upgraded inverted size 2” was simulated. In this system, the reprocessing time is twice shorter (6 months) and the zone repartition is slightly changed.

Figure 6 shows the stockpile of ^{233}U in the inverted size 2 and upgraded inverted size 2 cases in comparison with the usual size 2 reactor (with a 1 year minimum reprocessing). Table 9 gives the total temperature coefficient and the breeding ratio of these systems.

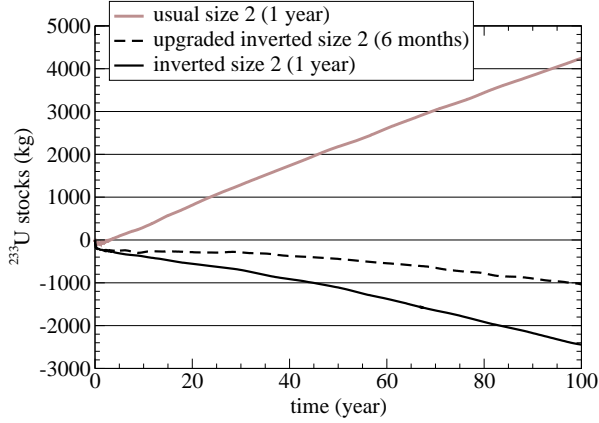


Figure 6: Stockpile of ^{233}U for inverted size 2, upgraded inverted size 2 and usual size 2 with minimum reprocessing (1 year)

Table 9: Temperature coefficient for inverted size 2, upgraded inverted size 2 and usual size 2 with minimum reprocessing (1 year)

	$\left(\frac{dk}{dT}\right)$	$\tau_{breeding}$
usual size 2	+0.74	1.042
inverted size 2	-0.44	0.971
upgraded inverted size 2	-0.68	0.988

Even if the zone inversion costs about 22 kg/year of ^{233}U instead of a breeding of 40 kg/year, this solution gives a negative total temperature coefficient. Even if it is not greatly negative, the advantage of this solution is that it only plays on core geometry and does not need any feeding or poisoning. Moreover, the reprocessing and the geometry can be adjusted to improve the reactor performances as shown with the upgraded inverted size 2.

Among the different solutions tested, improving safety often degrades breeding : erbium is a poison which capture neutrons, zone inversion puts in the maximum power density area large channels which associated breeding ratio is low (Figure 5), and decreasing reactor size increases neutron leakage and favors safety but reduces breeding.

A negative total coefficient implies more capture when temperature rises than at operating temperature. A solution is to put additional capture (poison like erbium) which evolves in

the right way. But these captures degrade the neutron balance and the breeding. So a compromise must be found. A strongly negative coefficient could be obtained if needed by giving up the breeding mode.

However, if the captures were made in a fertile material, it could improve both aspects. Another solution is then to use thorium instead of erbium.

4.4 Thorium blanket

Instead of losing neutron in a sterile poison, we can use a fertile element like ^{232}Th in place of ^{167}Er . As ^{232}Th cross section does not have useful resonance, this poison does not act the same way than ^{167}Er . Considering that neutron passing through the blanket are completely absorbed, this solution is not affected by dilatation. So it improves the density temperature coefficient.

Some thorium can be placed as a blanket in the graphite, provided that the Th concentration in the salt is adjusted to compensate captures on Th in the blanket. A solution is to replace a part of the reflector (axial and/or radial reflector) with a salt containing thorium.

Considering that cross sections do not significantly vary, the amount of Th which has to capture can be calculated from the reactivity definition. In the following equations, c , a_{in} and a_{out} are neutron creation rate and absorption rate in and out of the salt, and ϵ the dilatation coefficient which value is 8.10^{-4} for our salt [8] :

$$k = \frac{c}{a_{in} + a_{out}} \quad (5)$$

$$k + dk = \frac{c \cdot (1 - \epsilon)}{a_{in} \cdot (1 - \epsilon) + a_{out}} \quad (6)$$

Supposing that $k = 1$ and that $\epsilon \ll 1$ the formula becomes :

$$k + dk = 1 - \frac{a_{out} \cdot \epsilon}{c} \quad (7)$$

So we can deduce :

$$a_{out} = -\frac{c \cdot dk}{\epsilon} = -\frac{n_f \cdot v \cdot dk}{\epsilon} \quad (8)$$

with n_f the number of fission and v the number of neutrons emitted per fission (which values are respectively 950 kg/year and 2.4 n/fission). It means that a temperature coefficient of $-1 \text{ pcm}/^\circ\text{C}$ corresponds to a capture rate of 28 kg/year. The safety improvement is however limited by the total amount of neutrons passing through the reflectors : 78 kg/year for both reflectors (80% in radial and 20% in axial reflector), which allows a improvement of about -2 to $-3 \text{ pcm}/^\circ\text{C}$. The amount of salt involved in such tanks is some tens of tons, considering the same salt composition than the fuel. So ^{233}U produced must be extracted in several months (6 months to 1 year) in order to be injected in the fuel and possibly compensate the lower ^{233}U formation rate in fuel.

Such a solution seems promising, since it can improve the density temperature coefficient without degrading breeding.

4.5 Conclusion for safety studies

Three solutions have been evaluated to obtain a negative total temperature coefficient. The erbium solution corresponds to a neutron loss and the erbium inventory must be managed. In the same way, the zone inversion corresponds to a neutron loss but is interesting as a completely passive solution. Eventually, the thorium blanket solution is the only one in which captures improving safety participate also to the breeding, providing that the thorium inventory is managed. Studies are underway and more results will be available soon.

Conclusion

New reprocessing schemes for the Thorium Molten Salt Reactor started with ^{233}U have been studied, in order to examine the viability of such reactor, for which the reprocessing characteristic time is of the order of one year, instead of 10 days in the original MSBR studies. The general constraint is that the system has to be at least self-regenerator as regards the fissile material.

It has been shown that the concept simplification is possible, firstly by slowing down the process and secondly by modifying extraction scheme, while keeping the system self-regenerator.

The modification of the power density has been studied too. Lowering the power density by increasing the reactor size improves the neutron balance and gives a greater latitude, either to simplify the reprocessing, or to improve the temperature coefficients.

Concerning safety, we proposed three realistic solutions to face the temperature coefficients issue.

We can conclude that the drastic simplification of the reprocessing scheme makes the TMSR a quite attractive concept for generation-IV reactors.

References

- [1] J.F. BRIESMEISTER : “MCNP4B-A General Monte Carlo N Particle Transport Code”, Los Alamos Laboratory report LA-12625-M, 1997
- [2] A. NUTTIN et al. : “Thorium fuel cycles : a graphite-moderated molten salt reactor versus a fast spectrum solid fuel system”, Global, Paris, 2001
- [3] D. LECARPENTIER : “Le concept AMSTER, aspects physiques et sûreté”, PhD thesis, Conservatoire Nationale des Arts et Métiers, Paris, 2001
- [4] A. NUTTIN : “Potentialités du concept de réacteur à sels fondus pour une production durable d’énergie nucléaire basée sur le cycle thorium en spectre épithermique”, PhD thesis, University Joseph Fourier, Grenoble, 2002

- [5] EDF/DER : “Analyse critique du projet MSBR”, HT-12/24/77
- [6] E. WALLE, J. FINNE, G. PICARD, S. SANCHEZ, O. CONOCAR, J. LACQUEMENT : “Molten Salt Reactors : Chemistry of Fuel Salt and Fuel Salt Cleanup”, Global, New Orleans, 2003
- [7] D. LECARPENTIER, C. GARZENNE, D. HEUER, A. NUTTIN : “Temperature Feedbacks of a Thermal Molten Salt Reactor: Compromise between Stability and Breeding Performances”, Proceedings of ICAPP '03, Paper 3287, Córdoba, 2003
- [8] E.WALLE : “Densité et coefficient de dilatation thermique des mélanges de fluorures fondus utilisables dans AMSTER”, EDF internal report, Paris, 2003