NUCLEAR SCIENCE AND ENGINEERING: **161**, 78–89 (2009)

Possible Configurations for the Thorium Molten Salt Reactor and Advantages of the Fast Nonmoderated Version

L. Mathieu*

Laboratoire de Physique Subatomique et de Cosmologie, 53, avenue des Martyrs, 38026 Grenoble Cedex, France
and
Institut Laue Langevin de Grenoble, 6 rue Jules Horowitz, 38000 Grenoble, France

D. Heuer, E. Merle-Lucotte, R. Brissot, and C. Le Brun,
E. Liatard, J.-M. Loiseaux, O. Méplan, and A. Nuttin

Laboratoire de Physique Subatomique et de Cosmologie, 53, avenue des Martyrs, 38026 Grenoble Cedex, France

and

D. Lecarpentier

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

Received June 19, 2007
Accepted April 5, 2008

Abstract—*Molten Salt Reactors based on the thorium cycle were studied in the 1950 to 1960s to lead to the Molten Salt Breeder Reactor concept, which was finally discontinued prior to any industrial development. In the past few years, this concept has once again been studied in order to generalize it and seek configurations ensuring a high intrinsic safety level, an initial inventory compatible with intensive deployment on a worldwide scale, and a not-too-demanding salt chemical reprocessing scheme.*

The Thorium Molten Salt Reactor (TMSR) thus defined is studied in the Th-^{233}U cycle in various configurations obtained by modulating the amount of graphite in core to obtain a thermal, an epithermal, or a fast spectrum. In particular, configurations of a fast spectrum TMSR have been identified with outstanding safety characteristics and minimal fuel-reprocessing requirements.

I. INTRODUCTION

The Molten Salt Reactor (MSR) concept was developed in the early 1950s at Oak Ridge National Laboratory. The point was to conceive reactors whose fuel would be liquid, serving both as fuel and as coolant. The primary benefit of this concept is to allow a continuous adjustment of the fuel salt composition and thus ensure reactor operation over time with no reactivity reserve, which means improved safety characteristics. Moreover, very high temperatures and large power densities can be reached without large internal pressures, hence without severe constraints for the structural elements. The first experimental MSR stemmed from an American military program concerning plane propulsion that led to the Aircraft Reactor Experiment[1] in 1954. This 2.5-MW(thermal) reactor ran satisfactorily during 100 h. The continuation of this work led to the commissioning of the 8-MW(thermal) Molten Salt Reactor Experiment.[2] This reactor, based on a lithium, beryllium, and zirconium fluoride salt, was operated with 30% ^{235}U enriched uranium from 1965 to 1968, with ^{233}U from 1968 to 1969, and finally with plutonium mixed with ^{233}U in 1969. These successful experiments

*E-mail: mathieu@ill.fr

warranted the study of a thorium power breeder reactor coupled to a reprocessing unit for the online extraction of fission products (FPs): the Molten Salt Breeder Reactor[3,4] (MSBR). Although the concept looked promising, the project was stopped in 1976.

Studies on this type of reactor were resumed in the 1980s in Japan with the Thorims-NES project[5] and then the FUJI-AMSB project,[6] as well as in France with studies on the MSBR by Electricité de France (EDF) and the Commissariat à l'Energie Atomique (CEA). In the 1990s, the concept was taken up again with a view to incinerating nuclear wastes in subcritical reactors such as the TIER-1 project,[7] proposed by C. Bowman to transmute the plutonium of pressurized water reactors (PWRs); the CEA TASSE project[8]; or the EDF AMSTER project.[9]

Members of the Centre National de la Recherche Scientifique (CNRS) involved in the TIER-1 project[10] have continued to work on this type of reactor, and starting in 1999, more complete studies were carried out to reevaluate the MSBR in view of specifying a critical reactor based on the thorium cycle for energy production.[11,12] This reevaluation led to at least two important results that questioned the MSBR concept. On one hand, the feedback coefficient (or temperature coefficient) proved to be globally positive so that the reactor is intrinsically unstable, and on the other hand, the reprocessing unit appears unrealistic from the operational point of view and probably too complex[13] to ensure economic cost-effectiveness. Taking this into account, we have undertaken a systematic study of MSRs to resolve these problems and optimize the reactor in the framework of the deployment of a thorium-based reactor fleet on a worldwide scale.[14–19]

This work is based on the coupling of the MCNP neutron transport code[20] with the REM materials evolution code[12,14] developed at the CNRS. The former calculates the neutron flux and the reaction rates in the cells while the latter solves the Bateman equations for the evolution of the materials composition in the cells. These calculations take into account input parameters such as the power released, the criticality level, the initial composition of the salt, and the reprocessing performance while regularly adjusting the neutron flux and the materials composition. Our simulations are based on a precise description of the core geometry and consider hundreds of isotopes individually with their neutron interactions and radioactive decays. They allow a thorough and detailed interpretation of the results.

II. THE THORIUM MOLTEN SALT REACTOR AND ITS NEUTRON SPECTRA

II.A. General Description

The Thorium Molten Salt Reactor (TMSR) concept consists of a 1-GW(electric) reactor moderated by graphite and based on the $^{232}Th/^{233}U$ fuel cycle. Its operating temperature is 630°C,[a] which corresponds to a 40% thermodynamic efficiency. The moderating matrix is composed of a lattice of 15-cm side hexagonal graphite bars with hollow cylindrical channels in their center to allow fuel salt circulation. This geometry is shown in Fig. 1. The density of this nuclear-grade graphite is 1.86 g/cm^3.[b] The fuel salt used is a lithium, thorium, and uranium fluoride ($LiF–ThF_4–UF_4$). The reference proportion of heavy nuclei (ThF_4 and UF_4) is chosen equal to 22% molar, corresponding to a eutectic whose melting temperature is 565°C. The fissile matter (^{233}U) proportion in the heavy nuclei is ~3%. The density of the reference salt at 630°C is 4.3 g/cm^3 with an expansion factor of 10^{-3}/°C (Ref. 21).[c] A third of the 20 m^3 of the salt (6.5 m^3) flows through circuits that are external to the nuclear core and therefore not in the neutron flux. Actually, the heat exchangers are placed within the vessel and enclosed in neutronic protections in order to avoid fuel salt leaks to the environment. Two-thirds of the irradiated salt (9 m^3) is located in the moderator channels, and one-third (4.5 m^3) is located in the plena above and below the graphite matrix.

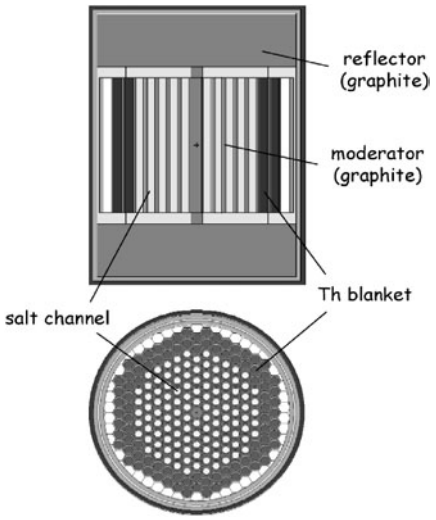

Fig. 1. Representation of the standard MSR (epithermal configuration).

As shown in Fig. 1, a radial blanket containing a fertile fluoride salt LiF-(^{232}Th)F_4 surrounds the core to

[a]The temperature chosen for our simulations is quite low compared to the melting point temperature of the salt. The mean operating temperature certainly has to be slightly higher. Nevertheless, a small gap of the absolute temperature has no consequence on the results.

[b]The simulated graphite is pure (does not contain any contaminant like boron) and is considered an assembly of perfect microcrystals.

[c]The expansion factor is defined as the variation of the density with the temperature.

improve the system's breeding capacity. This blanket was estimated to capture ~80% of the neutrons that escape the core radially. It contributes to protecting the external structure from irradiation effects while improving the breeding. The ^{233}U produced in the blanket is removed within 6 months by effective fluorination.

A key characteristic of MSRs is the possibility to reprocess the fuel during reactor operation. This allows on one hand limiting parasitic captures due to FPs and on the other hand avoiding having a reactivity reserve to ensure criticality over time. A possible fuel salt reprocessing scheme is shown in Fig. 2. The gaseous and noble metal FPs (nonsoluble in the salt) are extracted within a characteristic time shorter than 1 min by a helium bubbling placed in the external part of the fuel salt circuit. Figure 2 does not show the reprocessing of the salt blanket, which is much simpler.

The fuel salt is reprocessed off-line in order to extract the remaining FPs and optionally the transuranic elements (TRUs). With daily batches of ~100 ℓ, the total core volume is reprocessed over a 6-month period. Although it is still ill defined, this off-line reprocessing is subdivided into four phases, which can be grouped or interchanged depending on the chemical options chosen. First, the uranium is extracted by fluorination and quickly reinjected into the core, thus minimizing the amount of fissile matter in the fuel cycle. In the second stage, the TRUs are extracted. They can be either reinjected into the core or stored for further reprocessing. In the latter case, the ^{233}U resulting from the decay of ^{233}Pa ($T_{1/2}$ = 27 days) will be extracted by fluorination after a few months of storage, to be reintroduced into the core. It is important to point out that storing the ^{233}Pa outside of the flux brings a significant contribution to the breeding capacities of the system when the reprocessing is fast (as for the MSBR). With a slow reprocessing, the contribution is much smaller so that this option does not necessarily counterbalance the proliferation issues associated with such storage.

The third step consists of extracting the thorium, allowing, in the fourth step, removal of the FPs without taking too much thorium along. All these operations rest on fluoride pyrochemistry and are akin to those contemplated for the solid fuel reprocessing of the future fast neutrons reactors (FNRs).

II.B. Impact of the Moderation Ratio

The neutron spectrum of a nuclear reactor depends on its moderation ratio, i.e., the ratio of the amount of fuel to that of the moderating matter. In the TMSR, the neutron spectrum can be controlled by changing the size of the salt channels, extending from an extremely thermalized to a relatively fast spectrum. While changing the size of the channels, we decided to adjust the core size to keep a constant 20-m^3 volume of fuel salt. Indeed, the salt volume has a direct and strong influence on the reactor behavior, and this volume would vary by a factor of 200 between the most extreme configurations at constant reactor size. Studies of the influence of the salt volume for a constant moderation ratio were also carried out, and results are summarized in Sec. IV. For large-channel configurations, the axial graphite reflectors are replaced with ZrC reflectors for reasons that are explained in Sec. V.A.

This study is presented in full detail in Ref. 16; its most important results are reviewed here. Figure 3 illustrates the influence of the channel size, thus of the moderation ratio, on the behavior of the TMSR. Table I presents the quantitative values associated with Fig. 3.

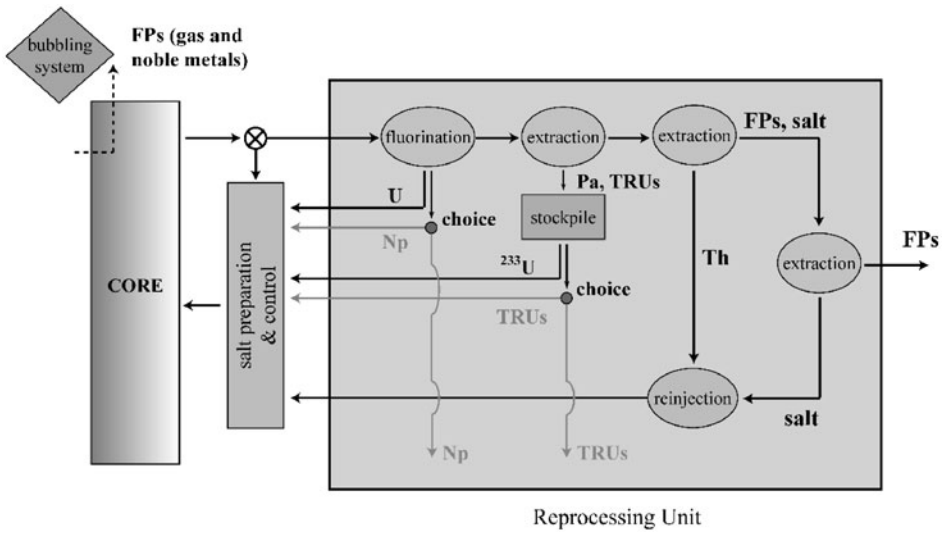

Fig. 2. Diagram of the fuel salt reprocessing (not showing the salt blanket reprocessing).

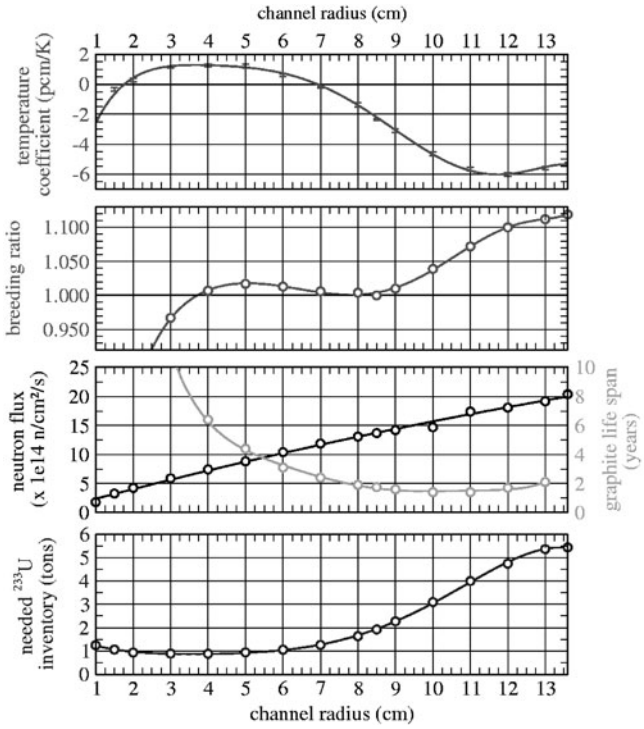

Fig. 3. Influence of the channel radius on the behavior of the TMSR with a 6-month fuel reprocessing time.

In order to evaluate precisely each reactor configuration, we have calculated the temperature coefficient dk/dT (in pcm/K[d]) for the safety aspects, the breeding ratio with the core and the blanket taken as a whole to ensure a long-lasting use of the resource, the neutron flux averaged on the salt in moderator channels and then the graphite life span to assess the operating difficulties, and the initial needed fissile inventory[e] to determine fast deployment capabilities.

Table II gives the geometry parameters for three typical reactor configurations (channel radii of 4, 8.5, and 13.6 cm). Tables III and IV present in-core and removal inventories of heavy nuclei for the two TRU handling options (autoincineration in Table III or extraction of the TRUs in Table IV). Tables III and IV allow us to compare the production of highly radiotoxic elements/nuclear wastes for the two options. The in-core inventories are the amounts of matter present inside the reactor, while the removal inventories are the amount of matter extracted during each reprocessing. The two quantities are given after 100 yr of reactor operation. The removals are normalized to the amount of energy produced on the basis of 7 TW·h(electric)/yr, and they do not take into account losses incurred during FP extraction. That is why the output flow of the autoincinerated MSRs is set to zero in Table IV.

Many other quantities were observed and are fully developed in Ref. 14. Various neutron behaviors are available according to the moderation ratio chosen. The properties of the three configuration types[f] considered are summarized:

1. thermal spectrum configuration for 4-cm channel radii. Except for the temperature coefficient, this configuration has attractive characteristics, in particular its small initial inventory and its low production of light TRUs (Np and Pu).

2. epithermal spectrum configuration for 8- to 9-cm channel radii, which draws nearer to the MSBR characteristics. In that case, the operating parameters can be considered acceptable, except for the short life span of the moderating graphite.

3. fast spectrum configuration for 13.6-cm channel radii, which corresponds to a core with no moderating graphite (this configuration will be referred to as the "single salt channel" configuration). As opposed to other fast spectrum configurations, this one has the obvious advantage that it requires no control of the graphite. Moreover, its very negative feedback coefficient ensures a high deterministic safety level,[22] and its production of heavy TRUs (Am, Cm, or Cf) is particularly low. The only problem is the initial inventory of 5 tons of ^{233}U. It is smaller than that of any solid-fuel fast reactor started with Pu, but ^{233}U is not available in such large quantities. This inventory seems too large to ensure a satisfactory intensive deployment on a worldwide scale.[23] Studies to optimize the transition to the thorium cycle with such reactors[19] are under way.

The neutron spectrum of each of these three configurations is shown in Fig. 4. In Fig. 4, both the thermal and fast spectra are compared to generic spectra of the same kind: a PWR and a type BN800 sodium-cooled FNR. As one can see, the spectrum of a PWR, which is ordinarily qualified as thermal, is less thermalized than that of the 4-cm channel radius configuration. On the contrary, the spectrum obtained for a TMSR with no graphite is not as fast as the one obtained for the BN800 FNR. Indeed, the ^{19}F component in the salt has a large inelastic scattering cross section at high energies, causing a dip in the spectrum between 100 keV and 1 MeV.

[d]The unit of measure pcm/K represents 10^{-5} of the multiplication factor per kelvin.

[e]The initial needed fissile inventory is the sum of the inventory necessary to reach criticality and the amount of ^{233}U added in the first weeks to compensate the delayed ^{233}Pa decay.

[f]The neutron spectra depend on the materials used. Specifically, the configuration without graphite has a fast spectrum thanks to its poorly moderating heavy nuclei–rich fuel salt.

TABLE I

Influence of the Channel Radius on the Behavior of the TMSR with a 6-Month Reprocessing

Channel Radius (cm)	Temperature Coefficient (pcm/K)	Breeding Ratio	Neutron Flux ($n/cm^2 \cdot s$)	Graphite Life Span (yr)	Needed ^{233}U Inventory (tons)
1	-2.29 ± 0.07	0.56	1.8×10^{14}	94	1.25
1.5	-0.33 ± 0.11	0.73	3.3	40	1.07
2	0.27 ± 0.11	0.85	4.2	23	0.94
3	1.13 ± 0.06	0.967	5.9	11	0.89
4	1.26 ± 0.09	1.007	7.4	6.4	0.88
5	1.22 ± 0.13	1.017	8.8	4.4	0.94
6	0.59 ± 0.07	1.013	10.4	3.1	1.06
7	-0.21 ± 0.05	1.006	11.9	2.4	1.26
8	-1.36 ± 0.14	1.004	13.1	1.9	1.64
8.5	-2.35 ± 0.05	1.000	13.7	1.75	1.92
9	-3.1 ± 0.12	1.010	14.2	1.6	2.27
10	-4.65 ± 0.13	1.039	14.7	1.4	3.10
11	-5.65 ± 0.13	1.072	17.4	1.4	4.00
12	-6.03 ± 0.11	1.100	18.1	1.7	4.75
13	-5.59 ± 0.11	1.112	19.2	2.1	5.37
13.6	-5.3 ± 0.10	1.119	20.4	—	5.44

TABLE II

Geometry Parameters for Three Typical TMSR Configurations

	$r = 4$ cm	$r = 8.5$ cm	$r = 13.6$ cm
Core radius	2.65 m	1.6 m	1.25 m
Core height	5.3 m	3.3 m	2.6 m
Number of channels	336	120	—
Mass of graphite (moderator only)	175 tons	15 tons	0 ton

Possible improvements to the thermal and epithermal spectrum configurations are discussed below, as well as the excellent potential of the fast spectrum configuration.

III. THE THERMAL THORIUM MOLTEN SALT REACTOR

The thermal spectrum version of the TMSR has a positive temperature coefficient, which means an intrinsic instability of the core. There are several solutions to

TABLE III

Heavy Nuclei in Core and Removal Inventories for Three Typical Configurations After 100 yr Operation in the Case Where the TRUs Are Extracted

	In-Core Inventories			Removal Inventories (Output Flows)		
	$r = 4$ cm	$r = 8.5$ cm	$r = 13.6$ cm	$r = 4$ cm	$r = 8.5$ cm	$r = 13.6$ cm
Th	48.4 tons	46.2 tons	43.8 tons	0	0	0
Pa	77 kg	78 kg	85 kg	0	0	0
U	1.6 tons	3.8 tons	8.3 tons	0	0	0
(^{233}U)	(770 kg)	(2.1 tons)	(5.3 tons)	(0)	(0)	(0)
Np	7.0 kg	15 kg	9.7 kg	2.0 kg/TW·h(electric)	4.3 kg/TW·h(electric)	2.8 kg/TW·h(electric)
Pu	1.9 kg	2.8 kg	600 g	540 g/TW·h(electric)	800 g/TW·h(electric)	170 g/TW·h(electric)
Am	390 mg	515 mg	750 μg	110 mg/TW·h(electric)	150 mg/TW·h(electric)	215 μg/TW·h(electric)
Cm	1.0 g	105 mg	22 μg	285 mg/TW·h(electric)	30 mg/TW·h(electric)	6 μg/TW·h(electric)
Bk	50 ng	15 ng	0	15 ng/TW·h(electric)	4 ng/TW·h(electric)	0
Cf	600 ng	50 ng	0	170 ng/TW·h(electric)	15 ng/TW·h(electric)	0

TABLE IV

Heavy Nuclei In-Core and Removal Inventories for Three Typical Configurations After 100 yr Operation, in the Case Where the TRUs Are Autoincinerated*

	In-Core Inventories			Removal Inventories (Output Flows)		
	r = 4 cm	r = 8.5 cm	r = 13.6 cm	r = 4 cm	r = 8.5 cm	r = 13.6 cm
Th	48.4 tons	45.3 tons	43.3 tons	0	0	0
Pa	75 kg	74 kg	85 kg	0	0	0
U	1.6 tons	4.2 tons	8.3 tons	0	0	0
(^{233}U)	(790 kg)	(2.1 tons)	(5.2 tons)	(0)	(0)	(0)
Np	29 kg	110 kg	150 kg	0	0	0
Pu	38 kg	260 kg	270 kg	0	0	0
Am	3.1 kg	7.1 kg	4.8 kg	0	0	0
Cm	14 kg	18 kg	2.4 kg	0	0	0
Bk	5.1 g	40 g	170 mg	0	0	0
Cf	60 g	110 g	730 mg	0	0	0

*In that case, the configuration with r = 13.6 cm has not yet reached equilibrium.

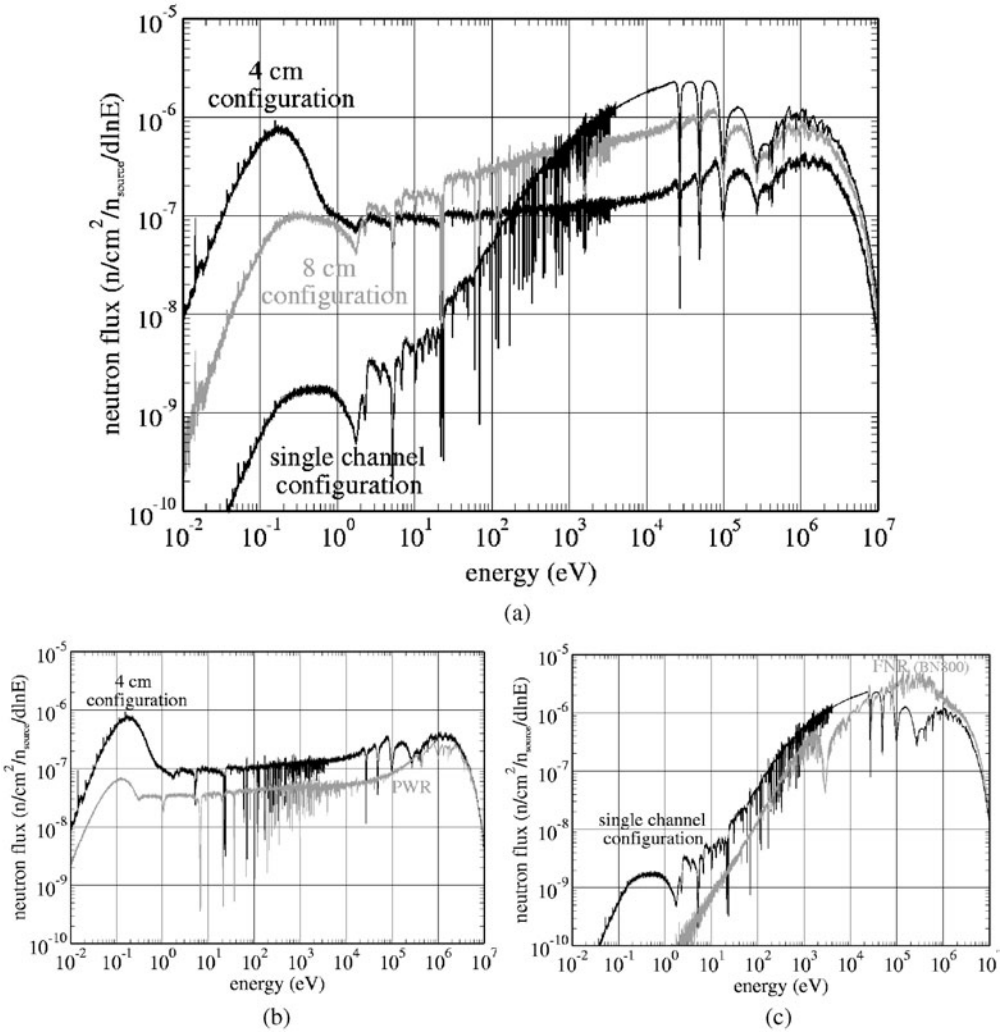

Fig. 4. (a) Neutron spectra of the three sorts of configuration of the TMSR and comparisons of (b) the thermal spectrum with the generic spectrum of a PWR and (c) the fast spectrum with that of a sodium-cooled–type BN800 FNR.

correct this effect, but we will not discuss the most classical ones. The study presented above was carried out by changing the radius of the channels inside constant-sized hexagons with 15-cm sides. However, the size of the hexagons also has an influence on neutron behavior in the core so that we studied how other hexagon sizes, 5-, 10-, and 20-cm sides, affect the reactor's performance. Figure 5 illustrates the transition to small hexagons (size reduction by a factor of 3) with a constant moderation ratio. Note that the total salt volume, which is a key parameter of the core, is kept constant by adjusting the number of hexagons in the core.

The evolution of the temperature coefficient for different hexagon sizes is shown in Fig. 6. As the range within which the channel radius can vary is directly linked to the hexagon size, the results are plotted versus the ratio between these two lengths so that the different configurations can be compared. The equivalent radius for 15-cm hexagons is given at the top of Fig. 6.

Although the three types of neutron spectrum are covered in Fig. 6, we will discuss only the thermal domain. The feedback coefficients are significantly improved when the hexagon size is reduced, i.e., when the salt and graphite are closely linked. Thus, a single moderation ratio can lead to very different neutron behaviors.

For the thermal configurations, the feedback coefficients are improved thanks to two effects. First, the global homogenization of the core leads to smaller channels and reduces the self-protection of the resonances. As the capture resonances are predominant, this effect induces a lower temperature coefficient. Second, the influence of the graphite is reduced as the core becomes more homogeneous. In fact, the core becomes less of a reflector and neutron storage area. Since the graphite has an adverse impact on the safety, this effect improves the temperature coefficient.

The breeding ratio is another important factor. Its evolution is displayed in Fig. 7. As previously stated, these results are obtained for a reprocessing of the whole salt volume in 6 months. Reducing the size of the hexagons improves the breeding ratio for the thermal configuration since it limits unwanted scattering in the moderator. Indeed, the abrupt dip in the breeding ratio for the thermal configuration with 15-cm hexagons is due to parasitic captures in the voluminous moderating graphite. The reduction of the parasitic captures obtained with smaller hexagons leads directly to an improved breeding ratio.

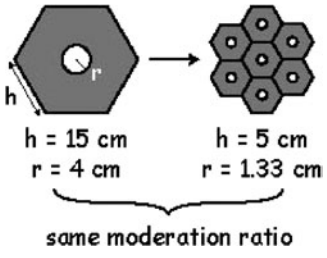

Fig. 5. Example of a change in size of the hexagons.

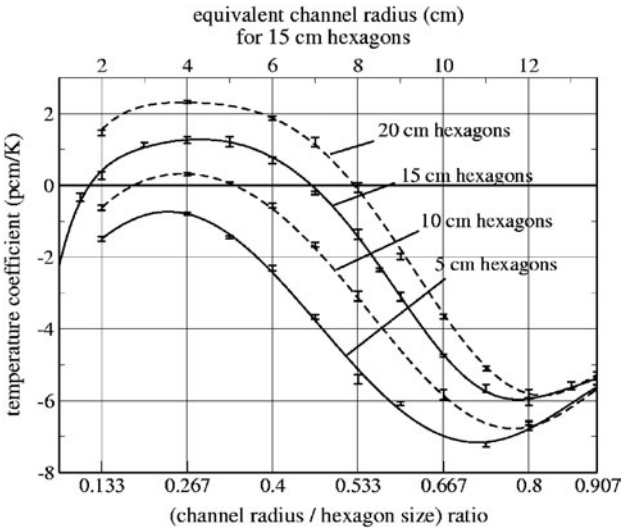

Fig. 6. Evolution of the temperature coefficient as a function of the channel radius for several hexagon sizes.

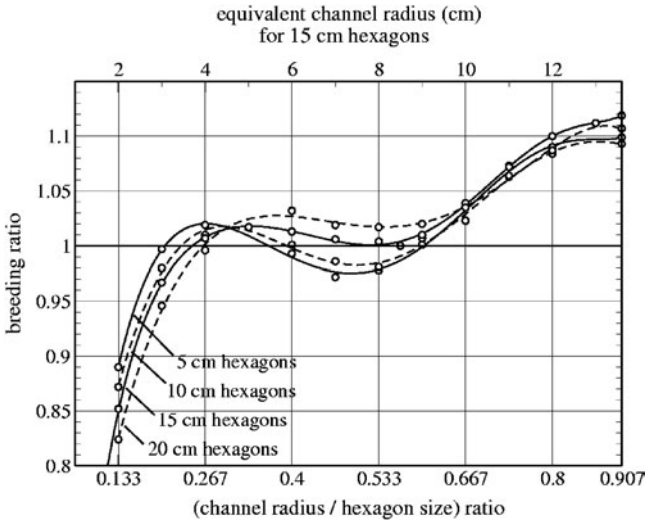

Fig. 7. Evolution of the breeding ratio as a function of the channel radius for several hexagon sizes.

The other characteristics of the core do not change much so that this homogenized thermal configuration leads to a reactor with a slightly negative feedback coefficient, a breeding ratio somewhat larger than 1.00, and a graphite life span of several years, all this for an initial fissile matter inventory <1 ton.

Additional studies[22] have shown that a slightly negative temperature coefficient is not enough so that it would

be useful to further improve this thermal configuration with small hexagons. A possibility would be a smaller reactor, containing for example 10 m^3 of salt releasing a power of 500 MW(electric) [instead of 20 m^3 of salt producing 1 GW(electric)]. The resulting increase in neutron escapes improves the feedback coefficient without deteriorating the system's breeding capacities too much thanks to the fertile blanket.

The performance of such a reactor would be the following: a feedback coefficient of -1.4 pcm/K instead of -0.8 pcm/K, an unchanged breeding ratio of ~ 1.020, and an average graphite life span of >5 yr for an initial inventory of 0.5 ton of ^{233}U for 500 MW(electric) [that is to say 1 ton to produce 1 GW(electric)].

IV. THE EPITHERMAL THORIUM MOLTEN SALT REACTOR

An epithermal spectrum is obtained for example with salt channels whose radius is 8.5 cm in 15-cm hexagons. The characteristics of such a reactor (see Fig. 3) are acceptable, without ensuring a wide operating margin. In particular, the -2.4 pcm/K feedback coefficient is close to the acceptable limit, and the breeding ratio, extremely close to 1.000 in the simulations, does not guarantee that isobreeding will be reached in a real reactor. Moreover, the average graphite life span is <2 yr, and this would certainly complicate reactor operation significantly.

Figure 6 shows that an epithermal configuration with small hexagons (equivalent radius of 8.5 cm) has much better safety characteristics, with a feedback coefficient of -5.5 pcm/K instead of -2.4 pcm/K. This improvement is due to a reinforcement of the epithermal part of the neutron spectrum where all the resonance cross sections lie. For captures taking over fissions, this domain of the spectrum has a stabilizing effect and makes the temperature coefficient more negative.

From the breeding point of view, there is a slight dip in Fig. 7 for the epithermal configurations. The reinforcement of the epithermal part of the neutron spectrum induces a deterioration of the $\alpha_{^{233}U}$ ratio (ratio of capture to fission cross sections for ^{233}U), which affects the breeding ratio on the first order. This loss in breeding must be compensated, for example with faster reprocessing.

Another solution to improve the performance of the epithermal TMSR (for whatever hexagon size) would be to increase the size of the core and, as a consequence, the fuel salt volume, while keeping the core power constant [i.e., 1 GW(electric)]. The findings in the study of the salt volume impact (discussed in Ref. 16) are only summarized in this paper (see Fig. 8). From these results, it seems interesting to double the salt volume from 20 to 40 m^3, increasing the breeding ratio

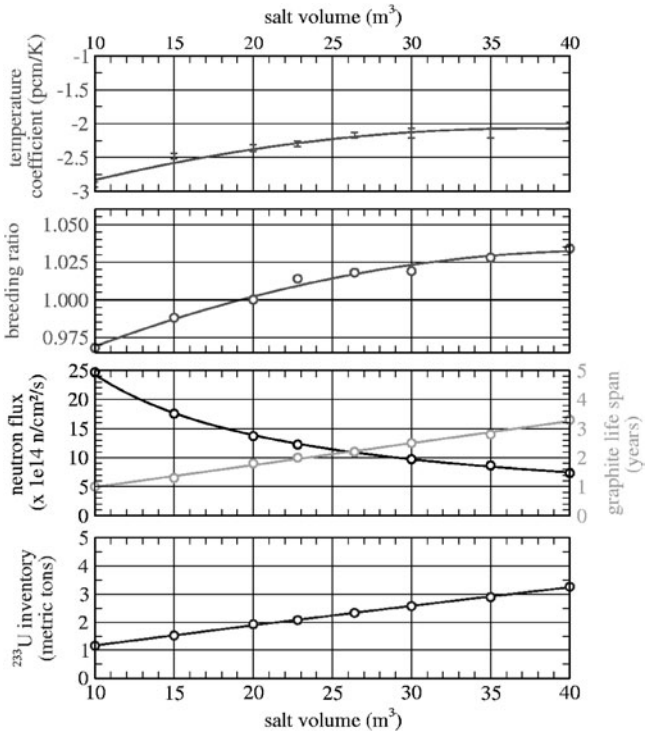

Fig. 8. Influence of the salt volume on the behavior of an epithermal TMSR with 15-cm hexagons. The reprocessing time is adjusted for a constant amount of heavy nuclei processed daily.

from 1.000 to 1.034. This leaves a sufficient margin to palliate the database uncertainties. Moreover, the average life span of the moderating graphite is almost doubled to reach 3.5 yr, representing a substantial increase, if not an ideal solution. On the contrary, the temperature coefficient is slightly worse (-2 pcm/K instead of -2.4 pcm/K), and the initial inventory is now 3.3 tons of ^{233}U instead of 1.9 tons. It is also important to note that the flux of irradiated graphite that has to be reprocessed or stored each year (not represented here) is not reduced: There is twice as much graphite in the core so that the increase of its life span is offset by the larger volume.

Combining these two options could produce a configuration with a very negative feedback coefficient and a graphite life span slightly larger than 3 yr for an initial inventory of ~ 3 tons of ^{233}U.

V. THE FAST THORIUM MOLTEN SALT REACTOR

V.A. Reflecting Materials

As previously mentioned (see Sec. II.B), in the fast spectrum configuration with no moderating graphite, the

axial reflectors are made of ZrC and not of graphite. Indeed, the presence of a large amount of a moderating material like graphite locally thermalizes the neutron spectrum. In a reactor with no moderating matrix, such a thermalization strongly decentralizes fissions, which then occur exclusively on the reactor's borders. With ZrC axial reflectors, because they do not moderate neutrons much, the fissions remain localized in the center of the core.

As shown in Fig. 3, in this configuration the temperature coefficient is very negative, and the breeding ratio is significantly larger than 1. This leaves a margin for several improvements.

V.B. Simpler Geometry

With the fertile blanket (fertile salt without ^{233}U), the neutrons that escape the core can be used for breeding. Removing this blanket or replacing it with a graphite or ZrC reflector directly reduces the breeding ratio. This is shown in Fig. 9 where the breeding ratio with (results identical to those in Fig. 3) and without a fertile blanket for a 6-month reprocessing time is plotted.

All the neutron spectrum types are spanned in Fig. 9, and it appears that the single salt channel configuration continues to breed even without a fertile blanket. However, the loss in breeding is larger for this configuration because the smaller core volume allows more neutron escapes. Removing the fertile blanket simplifies the core geometry drastically as well as the reactor's industrial operation. The negative aspect is that the initial inventory of such a configuration is 5.5 tons of ^{233}U, making intensive deployment on a global scale difficult.

V.C. Simplified Reprocessing

The reprocessing described in Sec. II.A is often considered as complex even after the simplifications already introduced since the MSBR (Ref. 16). We had indeed considerably reduced the speed of FP extraction from 10 days to 6 months. This slower reprocessing leads to a reduction of the reactor's breeding ratio because of core poisoning due to FP accumulation. The single salt channel configuration is much less sensitive to this poisoning thanks to its fast neutron spectrum. This is shown in Fig. 10 where the deterioration of the breeding ratio as a function of the reprocessing time is compared for the fast and epithermal configurations (with a fertile blanket).

The 6-month reprocessing time corresponds to iso-breeding for the epithermal configuration, but it is not the optimum of a fast configuration. We find that this configuration continues to breed, even with a reprocessing time of 10 yr.

It is also important to evaluate the system's dynamics in relation to long reprocessing times. Figure 11 shows the ^{233}U stockpile that we recover (if the reactor is a breeder reactor) or that we need to feed into the reactor to maintain criticality (if the reactor is under-breeder). This stockpile would be constant for an iso-breeder reactor.

We normally calculate the breeding ratio after 100 yr of evolution (that is to say, generally at equilibrium). However, this ratio decreases as and when the FPs accumulate in the salt. This explains not only the shape of the curves in Fig. 11 but also the fact that very long reprocessing times allow several years' operation without any external input of fissile matter, contrary to what Fig. 10 alone implies.

A reactor with no reprocessing unit can run for $>$20 yr with no external fissile matter input. It is worth pointing out that there is a physico-chemical limit to FP accumulation that is evaluated to be $\sim$20 yr at this production rate.[15] Such a configuration seems particularly attractive because of its extreme simplicity.

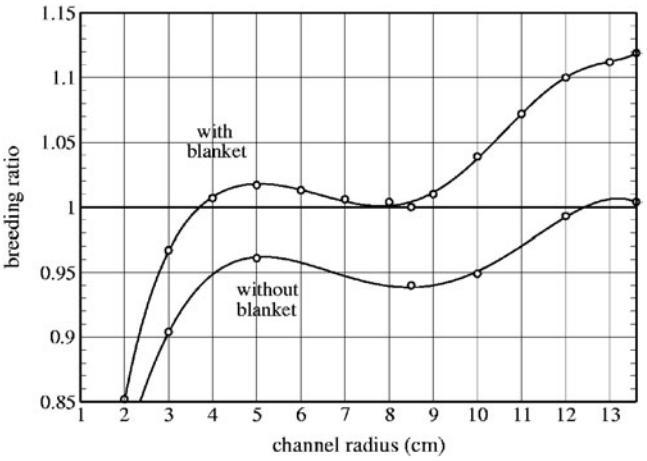

Fig. 9. Evolution of the breeding ratio as a function of the channel radius with and without a fertile blanket.

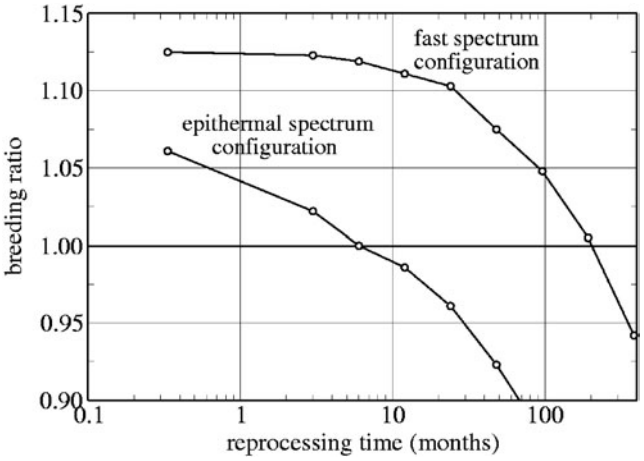

Fig. 10. Evolution of the breeding ratio as a function of reprocessing time, for the fast and epithermal configurations.

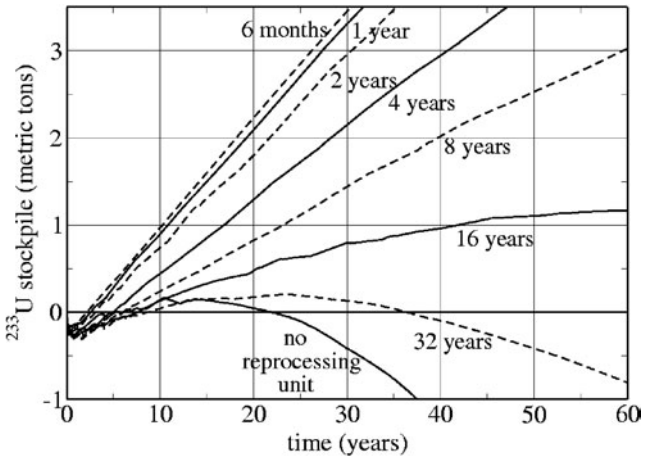

Fig. 11. Time evolution of the fissile matter stockpile of the fast configuration for several reprocessing times.

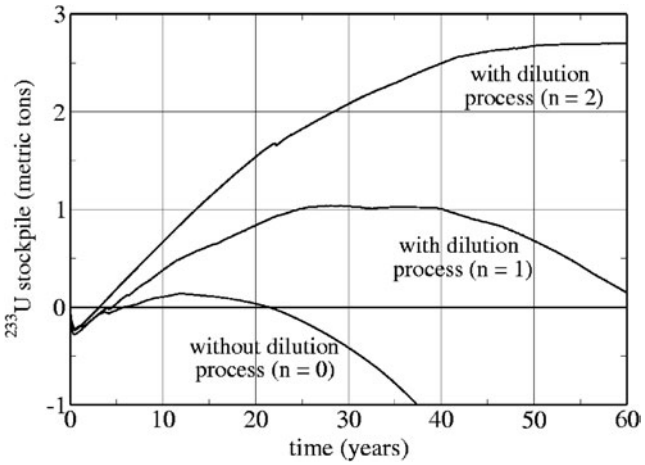

Fig. 13. Time evolution of the fissile matter stockpile of the fast configuration for several dilution factors.

Note that this discussion deals with the reprocessing unit alone and not with the bubbling system, which extracts nonsoluble gaseous FPs and noble metals. However, because it is more flexible, the single salt channel configuration requires a less efficient bubbling system. Such a reactor can run during ~10 yr without requiring a massive input of ^{233}U, in spite of the lack of reprocessing unit and with gaseous FP and noble metal extraction within a month instead of 30 s.

V.D. Dilution Process

It is possible to further improve the no-reprocessing-unit single salt channel configuration by means of a dilution process. This process consists of diluting the FPs and the TRUs in a bigger salt volume, according to the principle illustrated in Fig. 12. Here, n represents the ratio of the salt volume in the tank to that in the core. It is important to point out that this process is useful only as a replacement of the reprocessing unit and not to back it up.

The salt is extracted from the core and injected into the tank, which initially contains fresh salt (with no fissile matter, FPs, or TRUs) so that the initial ^{233}U inventory is not increased. Thanks to an immediate fluorination step, the fissile matter is returned to the core to avoid increasing its inventory unnecessarily. The tank's salt is itself reinjected in core in order to conserve the flow of matter. Thus, the tank salt is loaded with FPs as they are formed in the core. Although the same amount of FPs is being formed in core, they are now diluted in an $n + 1$ bigger volume, and their capture rate is decreased by the same $n + 1$ factor.

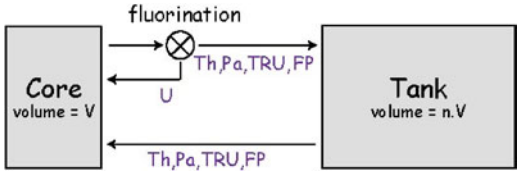

Fig. 12. Diagram of the dilution process.

This results in improved breeding capabilities and a larger fissile matter accumulation as shown in Fig. 13. With the no dilution process, the accumulation is identical to the one presented in Fig. 11 (with no reprocessing unit). If the reservoir is the same size as the core, i.e., $n = 1$, the system can run with no external input of fissile matter during ~60 yr. The physico-chemical limit is also increased by an $n + 1$ factor since it takes longer for the FPs to reach their saturation concentration. This limit, which is assumed to be 20 yr with no dilution process, is extended to 40 yr for $n = 1$, to 60 yr for $n = 2$, etc.

The dilution process is extremely interesting since it allows doing without a processing unit, except for the bubbling and the fluorination step, while ensuring satisfactory breeding capacities. Moreover, the cost of such a process is very low since it requires only one fluorination step and an adequate volume of fresh salt (containing Th and ^{7}Li in particular). Finally, the improved breeding capabilities mitigate the constraints concerning the bubbling system. In fact, with gaseous FP and noble metal extraction within 1 month instead of 30 s, the reactor can still run during 40 yr with no fissile matter input thanks to the dilution process ($n = 1$).

VI. CONCLUSION

We analyzed the behavior of three types of TMSRs, according to their operation with a thermal, epithermal, or fast neutron spectrum. We have attempted to find solutions to the various problems raised for each type of

reactor in order to obtain acceptable configurations. It appears from these studies that the TMSR can be operated with those three neutron spectrum types.

In the thermal spectrum, the safety issues are alleviated by reducing the distance between the salt channels in the graphite matrix. It is then possible to obtain a reactor with satisfactory properties although its feedback coefficient is only slightly negative and the irradiated graphite has to be reprocessed every few years.

In the epithermal spectrum, the safety is improved thanks to a very negative feedback coefficient, but the drawback is more frequent processing of the irradiated graphite and a larger fissile matter inventory.

The fast spectrum configuration with a single salt channel and without moderating graphite is the most promising. It has very negative feedback coefficients. This is true not only for the global temperature coefficient but also for the partial coefficients that characterize the dilatation or the heating of the salt, and the void effect. The breeding capabilities are large, and the constraints on the management of the irradiated graphite are nonexistent. However, its initial inventory is larger than that of the other configurations with >5 tons of fissile matter. Thanks to its good breeding capacities, the reactor, or more particularly its reprocessing, can be drastically simplified. It is even possible to design a TMSR that requires no coupling to an online fuel salt reprocessing unit.

The studies we carried out are not exhaustive, and other routes could be investigated. Meanwhile, these studies clarify the possibilities offered by TMSRs and demonstrate the inherent advantages of the fast neutron spectrum and of removing the moderating graphite. Continuing work has shown that this configuration can be further improved. The fuel salt composition [78% LiF–22%$(HN)F_4$] is rich in heavy nuclei so that a large fissile matter inventory is necessary. Studies with a smaller proportion of heavy nuclei have been initiated[16] and are continuing. Similarly, studies are under way in which the reactor is started with fissile nuclei other than ^{233}U, namely, Pu and minor actinides, and the results are very encouraging.[18,19]

ACKNOWLEDGMENTS

The studies described in this paper rest partly on work done with M. Allibert and on discussions in the frame of the European contract MOST (MOlten SalT: FIKW-CT-2001-20183). We thank A. Grockowiak and E. Huffer for their translation (from French to English).

REFERENCES

1. R. C. BRIANT and A.M. WEINBERG, "Aircraft Nuclear Propulsion Reactor," *Nucl. Sci. Eng.*, **2**, 795 (1957).

2. P. N. HAUBENREICH and J. R. ENGEL, "Experience with the Molten Salt Reactor Experiment," *Nucl. Appl. Technol.*, **8**, 107 (1970).

3. E. S. BETTIS and R. C. ROBERTSON, "The Design and Performance Features of a Single-Fluid Molten Salt Breeder Reactor," *Nucl. Appl. Technol.*, **8**, 190 (1970).

4. M. E. WHATLEY et al., "Engineering Development of the MSBR Fuel Recycle," *Nucl. Appl. Technol.*, **8**, 170 (1970).

5. K. FURUKAWA et al., "Thorium Molten-Salt Nuclear Energy Synergetics," *Nucl. Sci. Technol.*, **27**, *12*, 1157 (1990).

6. K. FURUKAWA et al., "New Primary Energy Source by Thorium Molten-Salt Reactor Technology," *Electrochemistry*, **73**, *8*, 552 (2005).

7. C. D. BOWMAN, "Once-Through Thermal-Spectrum Accelerator-Driven System for LWR Waste Destruction Without Reprocessing: Tier-1 Description," ADNA/98-04, ADNA Corporation (1998).

8. V. BERTHOU, "Le concept TASSE (Thorium ADS with Simplified Fuel Cycle for Long Term Energy Production)," Doctorate Thesis, Université d'Evry–Val d'Essonne (2000).

9. D. LECARPENTIER, "Le concept AMSTER, aspects physiques et sûreté," Doctorate Thesis, Conservatoire National des Arts et Métiers (2001).

10. O. MÉPLAN, "Technical Report on Bowman's Reactor," Internal Report, Laboratoire de Physique Subatomique et de Cosmologie (1999).

11. 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," Doctorate Thesis, Université Joseph Fourier–Grenoble-I (2002).

12. A. NUTTIN, D. HEUER, A. BILLEBAUD, R. BRISSOT, C. LE BRUN, E. LIATARD, J. M. LOISEAUX, L. MATHIEU, O. MEPLAN, E. MERLE-LUCOTTE, H. NIFENECKER, F. PERDU, and S. DAVID, "Potential of Thorium Molten Salt Reactors," *Prog. Nucl. Energy*, **46**, 77 (2005).

13. E. WALLE, J. FINNE, G. PICARD, S. SANCHEZ, O. CONOCAR, and J. LACQUEMENT, "Molten Salt Reactors: Chemistry of Fuel Salt and Fuel Salt Cleanup," *Proc. Global 2003*, New Orleans, Louisiana, November 16–20, 2003, American Nuclear Society (2003) (CD-ROM).

14. L. MATHIEU, "Cycle Thorium et Réacteurs à Sel Fondu: Exploration du champ des Paramètres et des Contraintes définissant le Thorium Molten Salt Reactor," Doctorate Thesis, Institut National Polytechnique de Grenoble (2005).

15. L. MATHIEU, D. HEUER, R. BRISSOT, C. GARZENNE, C. LE BRUN, E. LIATARD, J. M. LOISEAUX, O. MEPLAN, E. MERLE-LUCOTTE, and A. NUTTIN, "Proposition for a Very Simple Thorium Molten Salt Reactor," *Proc. Global 2005*,

Tsukuba, Japan, October 9–13, 2005, Atomic Energy Society of Japan (2005).

16. L. MATHIEU, D. HEUER, R. BRISSOT, C. GARZENNE, C. LE BRUN, E. LIATARD, J. M. LOISEAUX, O. MEPLAN, E. MERLE-LUCOTTE, A. NUTTIN, E. WALLE, and J. WILSON, "The Thorium Molten Salt Reactor: Moving on from the MSBR," *Prog. Nucl. Energy*, **48**, 664 (2006).

17. D. HEUER, L. MATHIEU, and E. MERLE-LUCOTTE, "Concept de réacteurs à sels fondus en cycle thorium sans modérateur," *Revue Générale du Nucléaire N° 5* (2006).

18. E. MERLE-LUCOTTE, D. HEUER, C. LE BRUN, L. MATHIEU, R. BRISSOT, E. LIATARD, O. MEPLAN, and A. NUTTIN, "Fast Thorium Molten Salt Reactors Started with Plutonium," *Proc. Int. Congress Advances in Nuclear Power Plants (ICAPP '06)*, Reno, Nevada, June 4–8, 2006, American Nuclear Society (2006) (CD-ROM).

19. E. MERLE-LUCOTTE, D. HEUER, M. ALLIBERT, V. GHETTA, C. LE BRUN, L. MATHIEU, R. BRISSOT, E. LIATARD, and A. NUTTIN, "Optimized Transition from the Reactors of Second and Third Generations to the Thorium Molten Salt Reactor," *Proc. Int. Congress Advances in Nuclear Power Plants (ICAPP 2007)*, Nice, France, May 13–18, 2007, French Nuclear Energy Society (2007).

20. "MCNP4B—A General Monte Carlo N Particle Transport Code," LA-12625-M, J. F. BRIESMEISTER, Ed., Los Alamos National Laboratory (1997).

21. V. IGNATIEV, A. MERZLYAKOV, E. WALLE, and J.-M. BOURSIER, "Density of Molten Salt Reactor Fuel Salts," *Proc. 11th Int. Topl. Mtg. Nuclear Reactor Thermal Hydraulics (NURETH-11)*, Avignon, France, October 2–6, 2005 (2005).

22. E. MERLE-LUCOTTE, D. HEUER, L. MATHIEU, and C. LE BRUN, "Molten Salt Reactor: Deterministic Safety Evaluation," *Proc. European Nuclear Conf.*, Versailles, France, 2005.

23. E. MERLE-LUCOTTE, D. HEUER, C. LE BRUN, and J.-M. LOISEAUX, "Scenarios for a Worldwide Deployment of Nuclear Power," *Int. J. Nucl. Governance, Economy and Ecology*, **1**, *2*, 168 (2006).