THE NON-MODERATED TMSR, AN EFFICIENT ACTINIDE BURNER AND A VERY PROMISING THORIUM-BASED BREEDER

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Starting from the MSBR concept, some parametric studies correlating the core arrangement and composition, the reprocessing set up and performances and the salt composition have shown that the reactor design where there is no graphite moderator inside the core appears to be the most promising in terms of safety coefficients, reprocessing requirements, and breeding capabilities. The core is a single large cylinder where the nuclear reactions occur within the flowing salt. Then the molten salt flows through the pumps, the gaseous extraction system which removes the gaseous and non soluble fission products and the heat exchangers before returning to the core. The core structures are protected by reflectors which absorb 80% of the residual neutron flux. With the on-line control and adjustment of the salt composition, the reactivity can be kept equal to one. A fraction of salt is periodically withdrawn and reprocessed off-line in order to extract the lanthanides before being sent back into the core. The actinides are returned to the core as soon as possible in order to be burnt.

A systematic study has shown that the salt composition has an effect on neutron energy moderation, actinide solubility, and initial fuel inventory. With a salt composition of 17.5 mole% of heavy nuclei (^{233}U: 4 650 kg and Th: 37830 kg), very good safety conditions have been found with a safety coefficient equal to -7 pcm/K. A good breeding ratio is obtained with the daily off-line reprocessing of a quantity of this salt containing 200 kg of heavy nuclei, i.e. a reasonable reprocessing rate.

The main asset of this reactor is that its initial load can be a mixture of thorium and, for its fissile material, the transuranic elements TRU (Pu, Np, Am and Cm) produced in today's water cooled reactors. The amount of TRU needed for the initial load is 6 390 kg. The results remain the same as those of a reactor started with ^{233}U for the reactivity coefficients, they are better for the production of ^{233}U during the first years of operation and the performances concerning the production of TRU elements at equilibrium are the same.

Under these conditions the TMSR appears to be a very appealing concept; building the reactor should not be very complicated and the reprocessing requirements are not too constraining. The main advantage of the concept is its safety performance; the reactivity coefficients are excellent, the void coefficient is negative thanks to the liquid fuel and the reaction can be easily stopped by pouring the liquid salt in dedicated tanks; the same design acts as thorium-based breeder and as actinide burner for the elements that it produces or for those produced in today's water cooled reactors.

As the Lithium salts have a very high boiling point, very high temperatures, useful for example for the production of hydrogen, may be reached without pressure and with fluids having very good heat conductivity properties. Due to the liquid fuel, the fuel manipulations are simple: no solid fuel fabrication with actinides, no transportation of spent fuel, and no solid fuel head-on difficulties for the reprocessing plant. The main constraints are the on-line control of the salt composition and of its chemical and physical properties. However, the aluminium industry, for example, is used to dealing with large salt quantities at very high temperatures.

All these properties put the TMSR in a very favourable position to fulfil the conditions defined by the GEN IV International Forum and to produce the large amount of nuclear energy that we will need in the near future with a small production of long lived nuclear wastes.

I. INTRODUCTION

As one of the six candidate systems retained by the Generation IV International Forum for the next generation of nuclear reactors, Molten Salt Reactors (MSRs) have to fulfill the general following criteria: durability, safety and reliability, waste minimization, economic competitiveness and proliferation resistance… The Molten Salt Reactor is a very attractive concept especially for the Thorium fuel cycle which allows nuclear energy production with a very low production of radiotoxic minor actinides and to reduce the existing transuranic element (TRU) amounts produced by today's water cooled reactors. Studies on molten salt reactors done at ORNL during the sixties led to the Molten Salt Breeder Reactor (MSBR)[1] project. Recent calculations[2] have been done to re-evaluate this concept. They have shown that the MSBR suffers from major drawbacks concerning safety and reprocessing, making it incompatible with any industrial development. On the other hand, the advantages of the Thorium fuel cycle were too attractive not to look further into it. So in order to take into account the advantages of the thorium fuel cycle which fit very well with the use of molten salts, we have started a parametric study of a thorium molten salt reactor where the core geometry and composition allow to extensively vary the neutron spectrum and the safety performances. The salt processing requirements have been simplified with a two step process: an on-line helium bubbling system dedicated to the extraction of gaseous and non soluble fission products (FP) and a separate pyrochemical unit dedicated to the extraction of the lanthanides, the actinides being re-injected in the core as soon as possible.

The main results obtained on the general TMSR concept concerning the safety coefficients and the breeding ratio as a function of the neutron moderation and of the processing performances of the chemical unit show that various configurations are possible which are briefly outlined here and are detailed in the reference[3]. We will focus our presentation on the TMSR with no moderator in the core which presents the best characteristics and mainly the capability to be started with the TRU produced in Light Water cooled Reactors (LWRs) mixed with a thorium base without losing the advantages of the thorium cycle

II. RE-EVALUATION OF THE THORIUM MOLTEN SALT REACTOR CONCEPT

This work is based on the coupling of a neutron transport code called MCNP[4] with the materials evolution code REM[1,5]. 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 within 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. Our calculations rest on a precise description of the core geometry and consider several hundreds of nuclei with their interactions and radioactive decay; they allow a thorough interpretation of the results.

We have reassessed the molten salt reactor concept using the thorium cycle to propose an innovative reactor called Thorium Molten Salt Reactor (TMSR). Many parametric studies of the TMSR have been carried out[3, 5, 6, 7], correlating the core arrangement and composition, the reprocessing performances, and the salt composition. In particular, by changing the moderation ratio of the core, the neutron spectrum can be modified and placed anywhere between a very thermalized neutron spectrum and a relatively fast spectrum. These studies are detailed in section II.A. A promising reactor configuration is then presented in section II.B and studied in the rest of the paper.

II.A. Description of the general TMSR

The general concept of the Thorium Molten Salt Reactor (TMSR) is a 2500 MWth (1 GWe) graphite moderated reactor based on the $^{232}Th/^{233}U$ fuel cycle. The graphite matrix comprises a lattice of hexagonal elements with 15 cm sides. The density of this nuclear grade graphite is set to 1.86. The salt runs through the middle of each of the elements. In this section, the salt used for the systematic studies is a binary salt, LiF - (Heavy Nuclei)F_4, whose (HN)F_4 proportion is set at 22 mole % (eutectic point), corresponding to a melting temperature of 565°C. The ^{233}U proportion in HN is about 3 %.

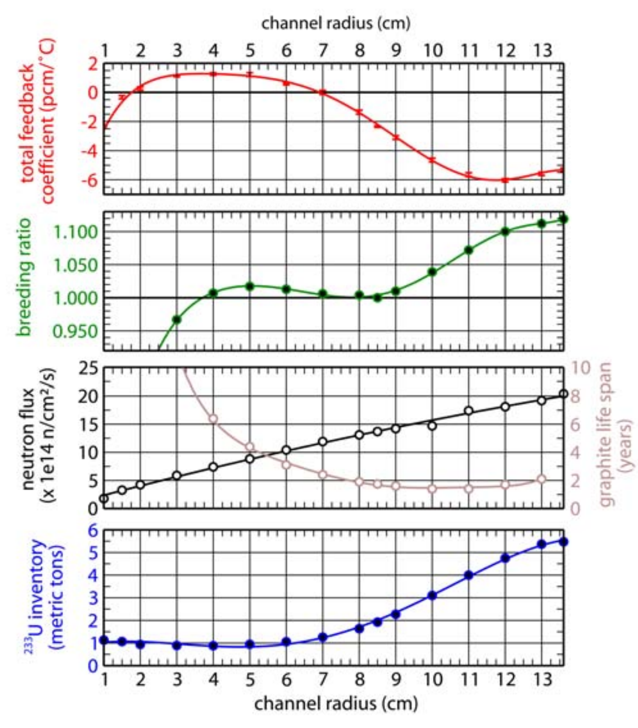

Fig. 1. Influence of the channel radius on the TMSR's behavior

A graphite radial blanket containing a fertile salt (LiF 78 mole% - ThF4) surrounds the core to protect the external structures, so that approximately 80 % of the escaping neutrons are stopped, and the regeneration is improved. We assume that helium bubbling in the salt circuit is able to extract the gaseous Fission Products (FP) and the noble metals within 30 seconds. For the first study presented, we consider a delayed reprocessing of the total salt volume over a 6 month period with a complete extraction of the FPs and of the transuranic elements (TRU). We assume that the ^{233}U produced in the blanket is also extracted within a 6 month period.

The moderation ratio can be altered by changing the channel radius. This modifies the neutron spectrum of the core, placing it anywhere between a very thermalized neutron spectrum and a relatively fast spectrum. The core size is adjusted to keep the whole salt volume constant and equal to 20 m^3. Fig. 1 shows the influence of the channel radius on the neutronic behavior. In order to evaluate the performance of a reactor configuration, we check a number of parameters: total reactivity feedback coefficient, breeding ratio, graphite life span and initial fissile inventory.

A wide variety of neutronic behaviors is available by changing the moderation ratio. We define three types of configurations: thermal, epithermal and fast spectrum. Each one has advantages and drawbacks: a thermal spectrum leads to a low fissile inventory but slightly positive feedback coefficient due to the graphite, an epithermal spectrum like that of the MSBR requires an efficient salt processing and leads to safety coefficients that are too limited while a fast spectrum implies a high breeding ratio and excellent safety coefficients but requires a large fissile inventory. Extensive studies have been carried out on these three fields of research to improve these configurations and to find relevant solutions [5, 6]. In this paper we will focus only on the fast neutron spectrum since it appears to be the most promising in terms of safety coefficients, reprocessing requirements, breeding and deployment capabilities. Moreover, a fast neutron spectrum opens the possibility of using fissile matter such as Plutonium, while it is hardly feasible with a thermal neutron spectrum[5].

II.B. The Non-Moderated TMSR

II.B.1. Core Description

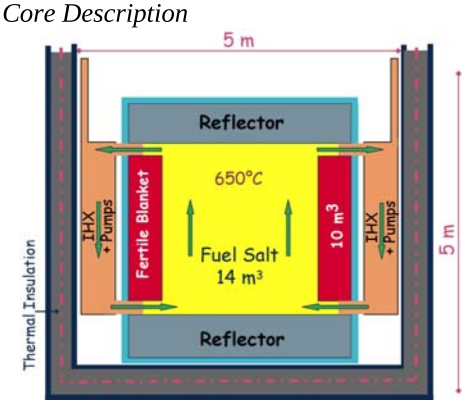

Fig.2. To scale vertical representation of the TMSR, including pumps and heat exchangers (IHX)

As shown in Fig. 2, the core is a single cylinder (1.25 m radius and 2.60 m height) where the nuclear reactions occur within the flowing salt. One third of the 20 m^3 of fuel salt circulates outside of the neutron flux in external circuits flowing through the pumps, the gaseous extraction system which removes the gaseous and non soluble fission products and the heat exchangers before coming back into the core.

II.B.2. Salt Composition

The salt composition (heavy nuclei content) has an influence on neutron energy moderation, actinide solubility, and initial fuel inventory. The actinide solubility has to be compatible with the salt composition and specially the heavy nuclei (HN) concentration. For HN proportions ranging from 20 to 30 mole%, a binary salt whose melting point is around 570°C has been chosen (see Fig. 3), that allows operation at 630°C.

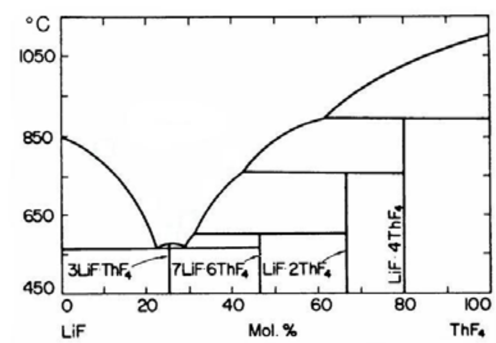

Fig. 3. LiF - ThF_4 phase diagram

For lower proportions of HN, we have either to increase the operating temperature or to add another fluoride to decrease the eutectic point temperature as shown in Fig. 3. The salt density ranges from 3.1 to 4.6 according to the HN proportion, with a dilatation coefficient[7] of 10^{-3}/°C. The calculations here have been made with a salt containing 80 mole% of LiF completed with BeF_2 but some other possible components (CaF_2 for example) are being studied.

II.B.3. Salt Reprocessing

Reprocessing at first deals with the on-line control and adjustment of the salt composition and properties (redox potential measurement, reactivity, temperature…).

At the present stage, we present a reprocessing scheme split in two parts: an on-line gaseous extraction system with helium bubbling is dedicated to the removal of gaseous fission products which are strong neutron poisons. As shown by ORNL data, this bubbling also extracts at least a part of the noble metals and non soluble fission products. In our simulations, we assume that this helium bubbling is able to extract the gaseous fission products and the noble metals within 30 seconds, a less efficient extraction having little effect on core behavior: up to an extraction time of a few days, the breeding ratio remains nearly constant. The extraction efficiency, which depends on the interaction between the liquid salt, the metallic clusters and the gas bubbles, needs scientific investigations and dedicated measurements to be optimized. Studies are also needed to determine the means to separate the fission products from the gas, to store them and to purify the gas.

For the extraction of the other fission products, mainly lanthanides, a fraction of salt is periodically set aside to be reprocessed off-line (batch mode). The fissile matter (uranium) can be extracted quickly by fluorination and sent back in the core. The other actinides and lanthanides can be separated via several methods like electrolysis, reduction into metallic solvents, solid precipitation, and any other method studied in the frame of pyrochemistry reprocessing. Finally the actinides are sent back into the reactor core to be burnt, while the lanthanides are stored apart. The performances of the reactor, in terms of breeding and deployment capacities, directly depend on the rate at which this off-line reprocessing is done (fig. 4).

III. THE THORIUM MOLTEN SALT REACTOR AS THORIUM-BASED BREEDER

III.A. Study as a function of the off-line reprocessing

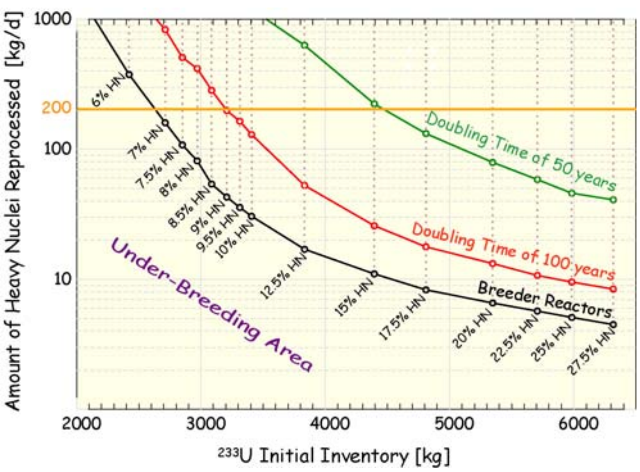

Fig. 4. Amount of heavy nuclei reprocessed per day versus initial fissile (^{233}U) inventory, for different heavy nuclei proportions in the fuel

For each proportion of heavy nuclei in the salt, ranging from 6 mole % to 27.5 mole %, we evaluate by simulation the breeding ratio of each reactor configuration as a function of the amount of heavy nuclei reprocessed per day. The result of this study is displayed in Fig. 4 which presents the reprocessing capacity required for a TMSR system. The ordinate is the weight of heavy nuclei reprocessed per day while the abscissa corresponds to the initial fissile (^{233}U) inventory. Heavy nuclei proportions in the MSR fuel are indicated (x %mole HN). The black line separates breeding and non-breeding zones. In the breeding zone, the red and green lines indicate the reprocessing requirements which allow the generation of the ^{233}U needed for a new TMSR reactor with the same inventory over 100 years and 50 years respectively.

Under-breeder reactor configurations, which are located under the black line (bottom of the figure), will not be considered in the following since they do not allow sustainable reactor deployment.

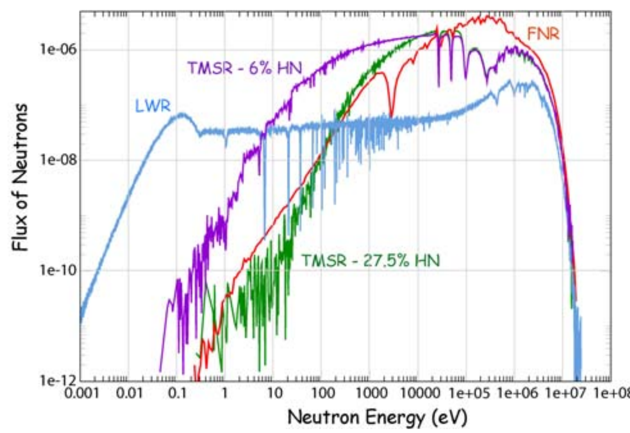

Fig.5. Neutron spectrum of two TMSR configurations (6% and 27.5% of heavy nuclei in the salt) compared to the neutron spectrum in a Light Water cooled Reactor (LWR) and in a Fast Neutron Reactor (FNR)

The ^{233}U initial inventory ranges from 2400 kg for a HN proportion in the salt of 6 mole % to 6300 kg for a HN proportion of 27.5 mole %. This corresponds to a variation of the neutron spectrum from an epithermal to a fast spectrum, as shown in Fig. 5.

We chose a reasonable reprocessing rate of 200 kg of HN per day, indicated by the orange horizontal line on Fig. 4. This choice disqualifies TMSRs with HN proportions lower than 7%, since they cannot then be breeder reactors (see Fig. 4).

III.B. Safety level

The total feedback coefficient at equilibrium is displayed in Fig. 6, together with its components, the contributions of the salt heating and salt dilatation, as a function of the HN proportion in the salt. All these safety coefficients are significantly negative for all HN proportions, including the density coefficient which can be viewed as a void coefficient. The total feedback coefficient ranging from -10 pcm/K to -5 pcm/K and thus ensuring a very good level of deterministic safety[9] in all these ^{233}U-started TMSR configurations, safety is not a discriminating factor in our case to choose the optimal salt composition.

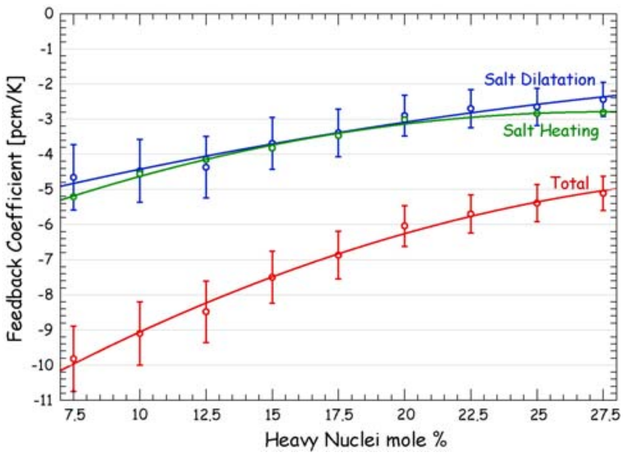

Fig.6. Feedback Coefficients of ^{233}U-started TMSRs at equilibrium as a function of the HN proportion

The uncertainties indicated are a quadratic combination of the statistical and systematic uncertainties on the determination of the sub-coefficients. The statistical errors are precisely estimated by simulation, the quantification of the systematic errors is more difficult. Specifically, concerning the systematic uncertainties on the contribution of salt heating, the cross-sections involved are well known, inducing only negligible uncertainties. The uncertainties on the salt density and its dilatation lead to systematic errors lower than 20% on the contribution of salt dilatation.

IV. THE THORIUM MOLTEN SALT REACTOR AS ACTINIDE BURNER

IV.A. Definition

The initial load of the non-moderated TMSR presented in section 1 can also be a mixture of thorium and, for its fissile material, the transuranic elements (Pu, Np, Am and Cm) produced in the water moderated reactors fed at present with natural or slightly enriched uranium. Actually, to be more realistic in the calculations, these TMSRs are started with the mix of 87.5% of Pu (^{238}Pu 2.7%, ^{239}Pu 45.9% , ^{240}Pu 21.5%, ^{241}Pu 10.7%, and ^{242}Pu 6.7%), 6.3% of Np, 5.3% of Am and 0.9% of Cm, corresponding to the transuranic elements of an UOX fuel after one use in a standard LWR and five years' storage[10].

For a typical TMSR configuration with 17.5 mole% of HN in the salt, an amount of 7300 kg of fissile elements is needed initially, that corresponds to 4.5 mole % of Plutonium and is of the order of magnitude of the amount needed to start a FNR in the U- Pu cycle.

IV.B. Safety level

The feedback coefficients for Pu-started TMSRs have been evaluated after one year of operation and at equilibrium. The values after one year of operation are displayed in Fig. 7 as a function of the HN proportion, together with their components, the contributions of salt heating and salt dilatation. They correspond to the initial safety behavior of the reactor, since the inventories after one year of operation are quite similar to the initial inventories, but we also take into account the effects of the fission products which are not present in the initial load of the core.

All these initial safety coefficients are still largely negative for any HN proportion, even if less negative (~25%) than for the ^{233}U-started TMSRs (see Fig. 6). The salt density contribution (equivalent to a void coefficient) is comparable, while the salt heating contribution is less negative because of the presence of Pu instead of ^{233}U.

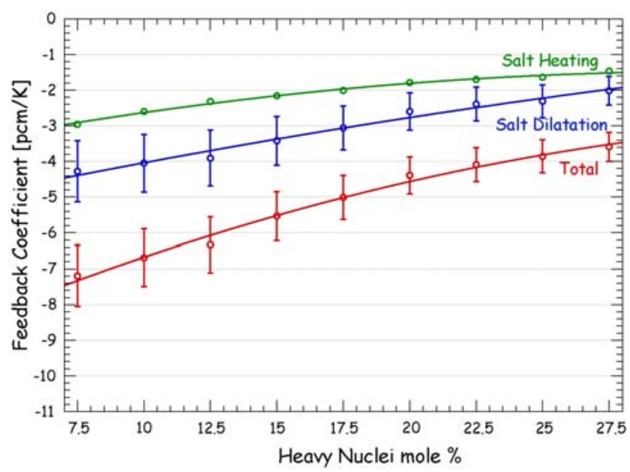

Fig.7. Feedback Coefficients of Pu-started TMSRs after one year of operation as a function of the HN proportion

The feedback coefficients evaluated at equilibrium are equivalent to the results presented in Fig.6 for the ^{233}U-started TMSRs, since these reactors are identical at equilibrium (see section IV.B).

IV.C. Heavy Nuclei inventories

Fig. 8 illustrates the evolution of a typical fuel salt composition (17.5% mole HN) all along the operation of this reactor, for the ^{233}U-started (solid lines) and for the transuranic-started (dashed lines) TMSR.

In terms of transuranic inventory, as shown in Fig. 8, the TMSRs started with transuranic elements become equivalent to TMSRs directly started and operated with ^{233}U after about forty years for a fuel salt with 17.5% of heavy nuclei, where more than 85% of the initial TRU inventories are burned. More generally, the assets of the Thorium fuel cycle are finally recovered for these transuranic-started TMSRs after 25 to 50 years for HN proportions ranging from 7 to 27.5 mole % (fig.9), except for minor actinides where longer times are needed.

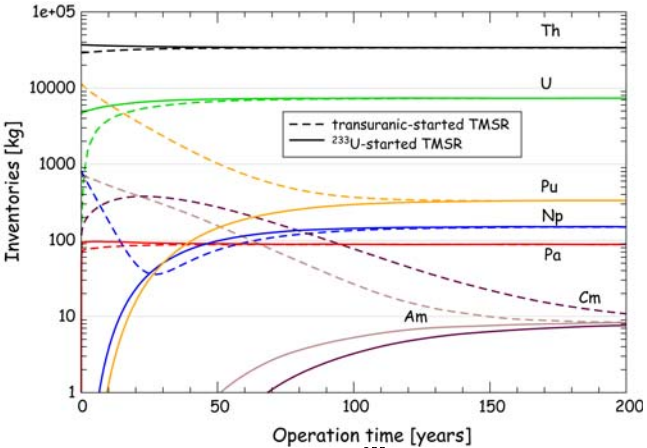

Fig.8. Heavy nuclei inventory for the ^{233}U-started TMSR (solid lines) and for the transuranic-started TMSR (dashed lines) with 17.5 mole% of heavy nuclei in the salt

At equilibrium, isotopic analysis shows that the $^{232}U/^{233}U$ ratio in the fuel salt is around 0.1 to 0.3%, corresponding to 2.5 to 20 kg for HN proportions ranging from 7 to 27.5 mole %. This ^{232}U is a major asset against proliferation in the case of the Th-^{233}U cycle, while presenting no major disadvantage during reprocessing thanks to the liquid fuel.

IV.D. Waste reduction

As the TMSRs started with transuranic elements are studied for their ability to close the current fuel cycle, we have to estimate the reduction of radiotoxicity reached at equilibrium. We aim at burning all TRUs introduced to start the reactor, so as to have TRU inventories at equilibrium identical to the TRU inventories of ^{233}U-started TMSRs, and this is verified (see fig 8).

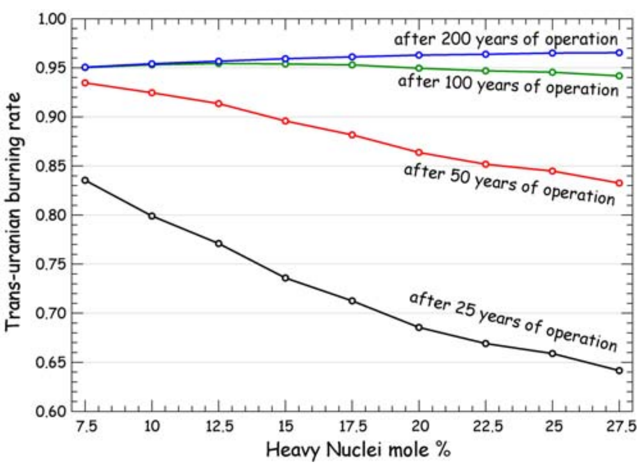

Fig.9. Burning rate of all the transuranic elements after different operation times, as a function of the HN proportion

Fig.9 shows the burning rate obtained for all the transuranic elements after 25, 50, 100 and 200 years of operation. Operation times greater than 60 years (a reactor lifespan) are obtained by transferring the salt contained in the closing TMSR into a new TMSR. This is made easier thanks to the fact that the fuel is liquid.

The lower HN proportion configurations allow faster reductions, since more than 80% of the TRUs are burned after only 25 years of operation, and nearly 95% after 50 years of operation. The larger HN proportion configurations allow higher burning rates, up to 97%, but on the longer run, after more than 100 years of operation.

IV.E. Deployment Capacities

Along with other reactors, the TMSR is among the systems with the capacity to incinerate a significant fraction of the GEN-2 TRU. The next issue is whether, given the absence of naturally available ^{233}U, a fleet of TMSRs can be deployed. As a matter of fact, the same question could be asked for any other GEN-4 reactor since ^{239}Pu is not available in nature either. The only difference is that ^{239}Pu (along with other TRUs) is present in LWR waste. On the other hand, the results presented in Section II establish that a TMSR not only is able to run in the Th-U cycle but can also start its operation using the TRU waste of LWR reactors destroying most of these TRU in the course of a transition towards the Th-U cycle operation.

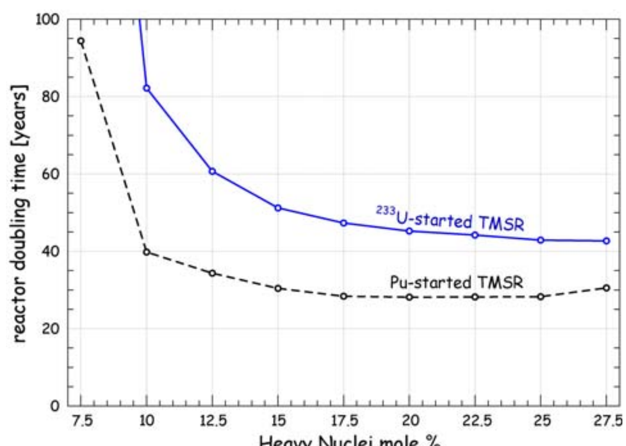

Fig.10. Reactor doubling time of ^{233}U-started (solid line) and Pu-started TMSRs (dashed line) for different HN proportion configurations and with a reprocessing of 200 kg / day

Finally a question can be raised as to whether the smaller breeding potential of the Th-U cycle (both in the thermal and fast spectrum) as compared to that of the U-Pu cycle (in fast reactors only) would allow a smooth transition from GEN-2-(GEN-3) reactors to a TMSR fleet. Before considering an evaluation of the breeding capacity of the TMSR it is worth recalling that to start a Fast

Neutron Reactor (FNR) using solid fuel one must have the TRU amount for two cores (to take into account the necessary cooling period before reprocessing). On the other hand, only one core load is needed for a TMSR since the reprocessing is performed as a (semi)-continuous batch procedure. This fact could compensate for the difference in the breeding potential of the two cycles. The transition from a GEN-2 to GEN-4 fleet can be initiated faster with TMSRs.

The quantity of interest for an evaluation of the deployment possibility is the doubling time, defined as the period of time over which the TMSR must be operated to produce the ^{233}U inventory for another TMSR. The results of such an analysis are given in Fig.10 for a range of values for the heavy nuclei proportion in the salt. Not surprisingly the cleaner the salt, the shorter the doubling time. It is also seen that in TRU-started TMSRs significantly larger amounts of ^{233}U can be extracted during their first 20 years of operation when the incineration of the initial TRUs allows most of the ^{233}U produced in the core to be saved. Optimal reactor doubling times around 30 to 40 years could be reached for heavy nuclei concentrations close to 17.5%. For ^{233}U-started TMSRs operated with the same HN proportion in the salt, the doubling time is about 50 years. Smaller values are obtained with higher concentrations. It thus appears that TMSRs are able to cope with energy strategies in which nuclear energy production is expected to grow at an annual rate of 1.5 to 2.2%.

V. CONCLUSIONS

The Thorium Molten Salt reactor (TMSR) presented here without moderator in the core appears to be a very promising, simple and viable molten salt reactor concept.

TMSRs, as any molten salt reactor, benefit from several specific advantages, mainly due to the liquid fuel and to the Thorium cycle. The liquid fuel does not have to be kept under high pressure even at high temperature; it is very stable vis-à-vis irradiation and allows to avoid the solid fuel preparation problems in the presence of actinides. Moreover, using a liquid fuel allows on-line fuel control. The amounts of fissile and fertile matter can be adjusted without unloading the core, doing away with the need for any initial reactivity reserve.

The non-moderated TMSR configurations with high HN proportions, leading to a rather fast neutron spectrum, present particularly interesting characteristics concerning their safety performances and their ability to be first loaded with transuranic elements produced by today's water cooled reactors. Finally, their rather large initial fissile inventory does not inhibit their capability for a fast deployment thanks to their very good ^{233}U breeding ratio during the TRU burning and they present very good arguments against proliferation due to the significant production of ^{232}U.

Under these conditions the TMSR appears to be a very appealing concept; building the reactor seems to be not very complicated, and our calculations do not indicate a major reprocessing constraint, allowing batch mode reprocessing in the vicinity of the reactor. The main additional studies needed to demonstrate the scientific feasibility of the concept deal with the on-line control of the salt composition and of its chemical and physical properties. Such studies are in progress in the frame of the French concerted research program 'Molten Salt Reactor' (PCR-RSF). Finally we want to point out the hardiness and the flexibility of this TMSR concept, allowing it to be adjustable without loosing its advantages in the event of any technologic problem.

All these properties put the TMSR in a very favourable position to fulfil the conditions defined by the GEN IV International Forum and to produce the large amount of nuclear energy that the world will need in the near future.

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