THE THORIUM MOLTEN SALT REACTOR: LAUNCHING THE THORIUM CYCLE WHILE CLOSING THE CURRENT FUEL CYCLE

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ABSTRACT

Molten salt reactors, in the configuration presented here and called Thorium Molten Salt Reactor (TMSR), are particularly well suited to fulfil the criteria defined by the Generation IV forum, and may be operated in simplified and safe conditions in the Th/233U fuel cycle with fluoride salts. The characteristics of TMSRs based on a fast neutron spectrum are detailed in this paper, focusing on their excellent level of deterministic safety. We aimed at designing a critical TMSR able to burn the Plutonium and the Minor Actinides produced in the currently operating reactors, and consequently to convert this Plutonium into ²³³U. This leads to closing the current fuel cycle thanks to TMSRs started with transuranic elements on a Thorium base, i.e. started in the Th/Pu fuel cycle. We study the transition between the reactors of second and third generations to the Thorium cycle in a European frame.

1. Introduction

The Generation-IV International Forum for the development of new nuclear energy systems has established a set of goals as research directions for nuclear systems: enhanced safety and reliability reduced waste generation, effective use of uranium or thorium ores, resistance to proliferation, improved economic competitiveness. The Molten Salt Reactor (MSR) is one of the candidates retained by Generation IV. MSRs are based on a liquid fuel, so that their technology is fundamentally different from the solid fuel technologies currently in use. Some of the advantages specific to MSRs (in terms of safety/reliability, for example) result directly from this characteristic. Furthermore, this type of reactor is particularly well adapted to the Thorium fuel cycle (Th-²³³U) which has the advantage of producing less minor actinides than the Uranium-Plutonium fuel cycle (²³⁸U-²³⁹Pu) [1].

We have reassessed the MSR concept to propose an innovative reactor called Thorium Molten Salt Reactor (TMSR). Many parametric studies of the TMSR have been carried out [2,3,4,5], correlating the core arrangement and composition, the reprocessing performances, and the salt composition. These studies have shown that the reactor design in which there is no graphite moderator inside the core appears to be the most promising. It uses a fast neutron spectrum, as described in section II of this paper. The characteristics (initial fissile inventory, salt composition, safety parameters, deployment capabilities) of such a TMSR configuration are detailed with the reactor operating either as a Thorium-based breeder or as an actinide burner.

Finally, the full transition between the second and third generation reactors to the Thorium cycle is optimized in the third section by considering the deployment capacities of the TMSR concept in a European frame, in association with the currently operated light water reactors.

This work made use of the MCNP neutron transport code [6] coupled with an in-house materials evolution code REM [4]. The former evaluates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition within the cells. These calculations take into account the input parameters (power released, criticality level, chemistry ...), by continuously adjusting the neutron flux or the materials composition of the core. Our calculations rest on a precise description of the geometry and consider several hundreds of nuclei with their interactions and radioactive decay.

2. The Non-Moderated Thorium Molten Salt Concept

2.1 Neutronic Core Description

The TMSR concept is a 2500 MWth (1GWe) reactor operated in the Thorium fuel cycle, using either ²³³U or Pu as its initial fissile fuel. As shown in Fig. 1, the core is a simple cylinder (1.25m radius and 2.60m height). The nuclear reactions occur within the up-flowing molten salt. The operating temperature is 630°C, corresponding to a thermodynamic efficiency of 40 %. At any moment, one third of the 20 m³ of fuel carrying salt is outside of the core, flowing through pipes, pumps, heat exchangers and the extraction systems aimed at removing the gaseous and insoluble fission products.

The core structures are protected by reflectors which ensure that 80% of the neutron flux are absorbed. To avoid thermalization of the reflected neutrons, the axial reflectors are made of ZrC. The radial reflector consists in graphite channels containing a binary fluoride salt LiF-ThF₄ with 28%- mole ²³²Th. This reflector is a fertile blanket, increasing the breeding ratio via a biannual extraction of ²³³U.

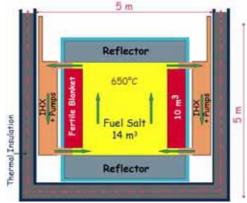


Fig. 1. Schematic vertical section of the TMSR, including pumps and heat exchangers (IHX)

2.2 Salt Reprocessing

Here, we consider only the reprocessing aspects relevant to the reactor operation. This reprocessing first involves a control and adjustment of the salt composition (redox potential measurement, reactivity kept equal to one...). The reprocessing itself is done in two steps. First, an on-line gaseous extraction system with helium bubbling removes all gaseous fission products (neutron poisons). Our simulations assume that helium bubbling extracts the gaseous fission products and the noble metals within 30 seconds. In fact, a less efficient extraction would not affect core behaviour much. Indeed up to an extraction time of a few days, the breeding ratio stays almost unchanged.

Secondly, foror the extraction of the other fission products, mainly lanthanides, a fraction of salt is periodically set aside to be reprocessed off-line (batch). The fissile matter (uranium) is quickly extracted by fluorination and sent back to the core. The other actinides and lanthanides are assumed to be separated via various methods such as electrolysis, reduction into metallic solvents, solid precipitation, or any other method studied in the frame of pyrochemistry reprocessing. Finally, the actinides are sent back to the reactor core to be burnt, while the lanthanides are evacuated as waste. The calculations below have been done assuming that reprocessing rates remain within a technologically realistic range taken as [50kg, 200 kg] of HN reprocessed per day.

2.3 Fuel Salt Composition

The core contains a fluoride fuel salt, composed of LiF enriched in ⁷Li (99.999 %) and heavy nuclei (HN) amongst which the fissile element, ²³³U or Pu. We have done parametric studies of this salt composition by varying the proportion of heavy nuclei in the fuel salt, and the reprocessing [7]. This has an influence on neutron energy moderation, actinide solubility, and the necessary initial fuel inventory. For HN proportions ranging from 20 to 30 mole%, a binary salt LiF-(HN)F₄ has been chosen whose melting point is around 570°C. For lower proportions of HN, the calculations have been done with a salt containing 80 mole% of LiF completed with BeF₂ to lower the eutectic point temperature and to allow operation at 630°C. Thanks to these parametric studies, we have selected for this presentation a typical fuel salt containing 17.5 mole % of heavy nuclei, with a reprocessing of 200

kg of heavy nuclei per day. This composition corresponds to a fast neutron spectrum. The salt density is equal to 3.8, with a dilatation coefficient of 10⁻³/°C [8].

As already stated, TMSRs can be operated in the Thorium fuel cycle, using either ²³³U (²³³U-started TMSR) or Pu (transuranic-started TMSR or TRU-started TMSR) as initial fissile matter. The ²³³U-started TMSR with 17.5 mole % of heavy nuclei requires an initial fissile load of 4.6 tons of ²³³U mixed with around 37 tons of Thorium.

Concerning the TRU-started TMSR, we have considered as initial load a mixture of thorium (29 tons) 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, these TMSRs are started with the mix of 87.5% of Pu (238Pu 2.7%, 239Pu 45.9%, 240Pu 21.5%, 241Pu 10.7%, and 242Pu 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 Light Water Reactor followed by five years of storage [9]. For the TMSR configuration considered here (17.5 mole% of HN in the salt), an amount of 7.3 tons of fissile elements is needed initially, corresponding to 4.5 mole % of Plutonium.

Finally, concerning safety, the feedback coefficients of the TMSR are negative for all HN proportions, thus ensuring in both cases a very good level of deterministic safety [10]. For the TMSR configurations considered here, the total feedback coefficient is equal to -7 pcm/°C at equilibrium for both the ²³³U-started and TRU-started TMSRs. The sub-coefficient called density coefficient, which can be viewed as a void coefficient, is also largely negative, around -3 pcm//°C.

2.4 TMSR as Actinide Burner

We have analyzed the evolution of a typical fuel salt composition during the operation of a TMSR started with ²³³U fuel and with a TRU initial fuel. As shown in Fig. 2, the TRU inventory of a TMSR started with TRU elements becomes equivalent to that of ²³³U started TMSRs after about forty years for our fuel salt with 17.5% of heavy nuclei: more than 85% of the initial TRU inventories are burned so that the assets of the Thorium fuel cycle are recovered for these TRU-started TMSRs within 40 years of operation.

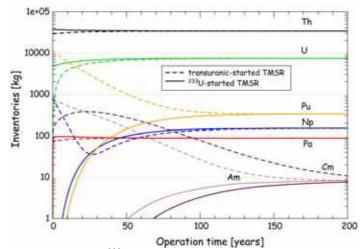


Fig.2. Heavy nuclei inventory for the ²³³U-started TMSR (solid lines) and for the TRU-started TMSR (dashed lines)

2.5 Deployment Capacities of TMSRs

Concerning ²³³U-started TMSRs, the deployment capacities are based on the amount of ²³³U produced compared to the initial fissile (²³³U) inventory necessary to start such a reactor. The amount of ²³³U produced and extracted all along the lifespan of the selected TMSR configuration is presented in Fig. 3 (purple line): for TMSRs directly started with ²³³U, the ²³³U extraction follows a linear growth, of 97 kg per year in the case shown. This production is directly related to the breeding ratio of the configuration.

TRU-started TMSRs allow the extraction of significantly larger amounts of ²³³U during the first 20 years of operation (Fig. 3, red upper line), thanks to the burning of TRUs which saves a part of the ²³³U produced in the core. This production reaches 185 kg per year in the example shown, i.e. 85% more than in the corresponding ²³³U-started TMSR. After the first 20 years, the ²³³U extraction rate is equivalent to that of the ²³³U-started TMSR.

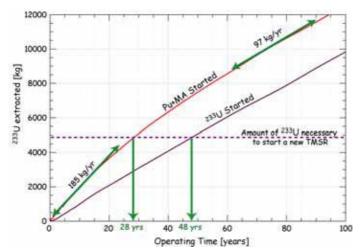


Fig.3: ²³³U extracted during the operation of TRU-started (red line) and ²³³U-started (purple line) TMSRs

The operating time necessary to produce one initial fissile (²³³U) inventory is called the reactor doubling time. On Fig. 3, the reactor doubling time of the ²³³U-started TMSR is equal to 48 years and corresponds to the crossing of the ²³³U production line (red line) with the initial ²³³U inventory (green dashed line). This reactor doubling time is reduced to 28 years when using transuranic elements as initial load. This production of significantly larger amounts of ²³³U in the TRU-started TMSRs during their first 20 years of operation is due to the burning of TRUs which saves a part of the ²³³U produced in the core. Using transuranic elements in TMSRs not only closes the current fuel cycle, but also improves the deployment capacities of Thorium based reactors.

3. European Deployment Scenarios

The deployment scenarios described below rest on the following nuclear power progression: starting near 1970, nuclear power production grows to 160 GWe.y (GigaWatt electric-year) in 2000. We postulate that nuclear power doubles between 2000 and 2050 to later increase slowly by 0.5% per year until 2100. Extrapolating up to 2100 allows us to verify that the deployment scenarios are lasting.

	Sodium cooled Fast
	neutron Reactor (SFR)
Output capacity	1.0 GWe
First operating date	2040
Reactor lifespan	50 yrs
Pu amount (per load)	6 tons
Loading periodicity	5 yrs
Number of loads	2
Breeding (per reactor-yr):	100 kg of Pu

Table 1: Characteristics of the SFRs considered

We have simulated the deployment of several nuclear reactor fleets based on different reactor technologies, to satisfy the anticipated energy demand stated above:

- The first scenario involves light water reactors (LWRs) [9] and fast neutron breeder reactors operated in the U-Pu fuel cycle, specifically Sodium cooled Fast Reactors (SFRs) (characteristics given in Table 1).

- The second scenario involves light water reactors and the TMSRs presented above: TRU-started TMSRs and ²³³U-started TMSR. As we aim at closing the current fuel cycle while lauching the Th fuel cycle, TRU-started TMSRs are started preferentially as long as transuranic elements produced in the light water reactors are available.

A standing question was whether a fleet of TMSRs can be deployed given the absence of naturally available ²³³U. In fact, the question could have been asked for any other GEN-4 reactor since ²³⁹Pu is not naturally available either. The difference is that ²³⁹Pu (along other TRU) is present in LWR wastes. On the other hand, the results presented in Section II establish that a TMSR is not only able to run in the Th-U cycle but can also start its operation using the TRU produced in LWR reactors.

These scenarios also led to an estimation of the production of heavy nuclei induced by the deployment of such a nuclear reactor fleet in Europe. We aim at evaluating the complexity of the management of these heavy nuclei stockpiles, as well as their radio-toxicity. Ultimately when this type of electricity production is replaced by a novel technology (fusion for instance) all the actinides inside reactors will become discardable waste. The possibility of eventually shutting down the reactor fleets started has thus to be studied, the heavy nuclei management being the key issue of such an option.

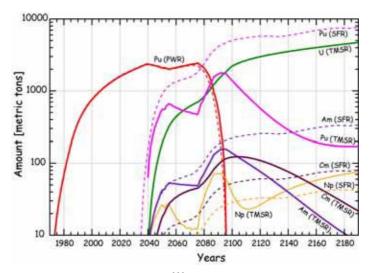


Fig. 5: Stockpiles of transuranic elements and ²³³U for two deployment scenarios: combination of LWRs and SFRs (solid lines), and combination of LWRs, TRU-started TMSRs and ²³³U-started TMSRs (dashed lines)

As shown in Fig 5, the scenario based on TMSRs allows a significant reduction of the transuranic element stockpiles. For example, the Plutonium inventory of the fleet composed of LWRs and TMSRs is reduced to 200 metric tons in 2150, instead of 6000 tons for the scenario based on SFRs. The Uranium inventory in the scenario with TMSRs, replacing the Pu inventory for SFRs, lies around 4000 tons. The amounts of others minor actinides are significantly reduced too thanks to TRU-started TMSRs, which really help close the current fuel cycle.

Finally, TRU-started TMSRs burn most of the TRU produced in the LWRs in the course of a transition towards a Th-U cycle operation, leading to an equilibrium salt composition whose radiotoxic content is significantly lower than that of the fuel in a SFR. Moreover, the uranium stockpile resulting from the fleet of TMSRs could be successfully burnt in identical TMSRs on an inert support, with a burning efficiency higher than 90% and a high deterministic safety level thanks to feedback coefficients ranging from -10 pcm/K to -5 pcm/K. As a result, waste management is simpler and easier to implement. Nuclear power deployment in this case is sustainable and efficient, the use of fissile matter and the production of wastes are optimized.

4. Conclusion

The Thorium Molten Salt reactor (TMSR) presented here with no moderator in the core appears as a very promising, simple and suitable concept of molten salt reactor. The non-moderated TMSR

configurations considered in this paper, based on a fast neutron spectrum, present particularly interesting characteristics. Their deterministic safety level is excellent. They can be started with a fuel made from the TRU wastes produced in current LWRs. Their rather large initial fissile inventory does not prevent fast deployment thanks to their good ²³³U breeding. The technology which in principle does not involve the transportation of radioactive materials outside the reactor site as well as the presence of ²³²U within the fuel can be considered as restricting proliferation risks.

The concept itself has some appealing aspects compared to earlier versions of MSRs. The reactor core is extremely simple. Simulation calculations do not point to major reprocessing constraints. In particular the fluxes considered should allow the batch mode reprocessing to be installed in the vicinity of the reactor. Initial studies of the scientific feasibility of the on-line control of the salt composition and of its chemical and physical properties have not unearthed a showstopper.

When it comes to Generation-4, it appears that the major nuclear energy powers have given a higher priority to the SFR concept. This mostly reflects a justified confidence in a technology which, although it has not yet reached all the performances expected for a GEN-4 reactor, has already been successfully tested in numerous projects. But all the properties detailed in this paper, especially its deterministic safety performances and its ability to reduce the radio-toxicity of wastes currently produced, put the TMSR in a very favourable position to fulfil the conditions defined by the GEN IV International Forum. Moreover this TMSR concept may be very appealing to countries which hold important thorium resources and have some remaining adjustment margins in the definition of their nuclear energy policy. The TMSR is thus an excellent candidate to produce the large amounts of nuclear energy that the world will need in the near future.

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6. References

- [1] A.Nuttin et al, "Potential of Thorium Molten Salt Reactors", *Prog. in Nucl. En.*, vol 46, p.77-99 (2005)
- [2] L.Mathieu et al, "Proposition for a Very Simple Thorium Molten Salt Reactor", *Global Conference*, Tsukuba, Japan (2005).
- [3] L. Mathieu, D. Heuer et al, "The Thorium Molten Salt Reactor: Moving on from the MSBR", Prog. in Nucl. En., vol 48, pp. 664-679 (2006)
- [4] L. Mathieu, "Cycle Thorium et Réacteurs à Sel Fondu : Exploration du champ des Paramètres et des Contraintes définissant le Thorium Molten Salt Reactor", PhD thesis, Institut National Polytechnique de Grenoble, France (2005) (in French)
- [5] E. Merle-Lucotte, D. Heuer C. Le Brun, L. Mathieu, et al, "Fast Thorium Molten Salt Reactors started with Plutonium", Proceedings of the *International Congress on Advances in Nuclear Power Plants (ICAPP)*, Reno, USA (2006)
- [6] J.F. Briesmeister, "MCNP4B-A General Monte Carlo N Particle Transport Code", Los Alamos Lab. report LA-12625-M (1997)
- [7] E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Brun, L. Mathieu, R. Brissot, E. Liatard, "Optimized Transition from the Reactors of Second and Third Generations to the Thorium Molten Salt Reactor", Proceedings of the *International Congress on Advances in Nuclear Power Plants (ICAPP)*, Paper 7186, Nice, France (2007)
- [8] V. Ignatiev, E. Walle et al, "Density of Molten Salt Reactor Fuel Salts", *Nureth Conference*, Avignon, France (2005)
- [9] C. de Saint Jean, M. Delpech, J. Tommasi, G. Youinou, P. Bourdot, "Scénarios CNE: réacteurs classiques, caractérisation à l'équilibre", CEA report DER/SPRC/LEDC/99-448 (2000) (in French)
- [10] E. Merle-Lucotte, D. Heuer, L. Mathieu, C. Le Brun, "Molten Salt Reactor: Deterministic Safety Evaluation", Proceedings of the *European Nuclear Conference*, Versailles, France (2005)