



Thorium-based Fast Reactors: Potential Benefits and Challenges

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ABSTRACT

Use of thorium in Fast Reactors (FR) is gaining consideration in the scientific community thanks to its potential benefits in terms of waste management. The present paper investigates the performance of Th in three FRs: a TRU-burner sodium-cooled FR, an iso-breeder lead-cooled FR and a fast-spectrum Molten Salt Reactor (MSR). The study confirms the relatively low actinide radiotoxicity generated by Th fuel in a closed cycle that could result beneficial to waste management. In addition, notably improved safety parameters are generally observed, which in turn allows increasing the TRU-burning rate while complying with safety requirements. The MSR emerges as the most promising option from the safety features standpoint and thanks to the unique flexibility fostered by a liquid fuel.

1 INTRODUCTION

Fast Reactors (FRs) have been historically developed for the purpose of Pu breeding in view of the perceived low ²³⁵U natural resources, and expecting an intense growth in nuclear energy production. Starting from the eighties, construction of nuclear power plants experienced a worldwide stagnation and research activities gradually focused on the use of innovative nuclear reactors for waste minimization and transmutation, and for enhanced safety. These remain primary goals for FR development in western countries. From the viewpoint of waste minimization, thorium use is considered because of its low mass number that fosters a very limited TRU (TRansUranic isotope) build-up in a closed fuel cycle [1]. Adoption of thorium as fertile material is also known to improve some safety features [2]. Furthermore, use of ²³³U instead of ²³⁹Pu as main fissile material may discourage proliferation thanks to the intense and penetrating gamma field generated by the progeny of ²³²U that accompanies the in-bred ²³³U. Following these considerations, studies about thorium use in FRs have started gaining momentum. A new impetus to this option has been recently given by the cancellation of the Yucca Mountain nuclear waste repository project in the US, as well as by the Fukushima accident. The latter focused once again the attention of the public opinion on safety-related aspects of the nuclear energy production and on spent fuel accumulation at the reactor pools. For countries that have decided to phase out the nuclear

energy option, management of the TRU legacy from Light Water Reactors (LWRs) has become a priority and Th may be the carrier to expedite TRU burning.

Three reactor concepts are considered in this paper to assess the pros and cons of Th use: a TRU-burner Sodium Fast Reactor, an iso-breeder Lead Fast Reactor and a fast-spectrum liquid-fuelled Molten Salt Reactor (MSR). Comparison between U and Th performance in these reactor concepts is based on safety-related parameters, TRU-burning capabilities, radiotoxicity generation and decay heat. Aspects related to fuel fabrication and proliferation resistance are also briefly discussed. Calculations are based on state-of-the-art core physics codes and equilibrium cycle methodologies.

2 METHODOLOGY

The results presented in this paper have been obtained using the core physics code ERANOS 2.2-N. The lattice data for the core calculations have been generated using the ECCO cell code with 1968-group neutronic library based on the JEFF 3.1 evaluation. The core calculations are performed using 33-group energy collapsed lattice data from ECCO. The nodal transport VARIANT calculation scheme is employed for core flux calculations related to the solid-fuelled FRs while the discrete-ordinate code BISTRO is used to investigate the MSR behaviour.

The EQL3D procedure [3,4] developed at the Paul Scherrer Institut (Switzerland) for the fuel cycle analysis of FRs has been extensively employed. EQL3D allows to simulate a FR behaviour over multiple cycles of operation starting from a given core configuration with an initial fuel composition. Under the main assumptions of constant core power, actinide mass and fuel management scheme (reloading, cooling, reprocessing, feed type), the simulated reactor eventually reaches an equilibrium state which is determined by the feed composition, independently of the initial fuel composition. Such equilibrium state will be considered as the reference core state in the present paper.

Further details about the use of EQL3D and ERANOS for the calculations here presented can be found in Refs. [5-9]. These references also describe the main modifications which have been introduced in the original EQL3D procedure to investigate the Th fuel cycle, to compute radiotoxicity and decay heat, and to simulate the on-line reprocessing typical of liquid-fuelled MSRs.

3 REACTOR CONCEPTS

This work investigates some of the implications related to the use of Th in three FR concepts. Table 1 summarizes their main features.

The sodium-cooled Toshiba-Westinghouse Advanced Recycling Reactor (ARR) [10,11] is selected as representative of a burner design. The configuration adopting U-TRU oxides (U-ARR in Table 1) features a Conversion Ratio (CR) of 0.45. The Th-based version of this core (Th-ARR) obtained by substituting U with Th as support fertile material decreases the core CR to 0.39. This decrement in CR is the consequence of a ~10% reduction in fuel density from UO₂ to ThO₂ and of the lower neutron economy of Th vs. U in the fast energy range. The lower TRU CR and the reduced contribution to fissions from the fertile isotopes (2.1% of total fissions from Th-232 compared to 9.4% from U-238) lead to a 40 kg/GWth-yr increase in TRU consumption in Th compared to U. The related impact on core safety features will be discussed in the following.

Table 1: Main characteristics of the investigated reactor concepts [10-13]

	Burner configurations			Breakeven configurations		
	U-ARR	Th-ARR	MSFR	U-ELSY	Th-ELSY	MSFR
Reactor thermal / electrical power [MW]	1000/450	1000/450	3000/1500	1500/600	1500/600	3000/1500
Coolant	Na	Na	22.5mol%AcF ₄	Pb	Pb	22.5mol%AcF ₄
Fuel type*	AcO ₂	AcO ₂	77.5mol%LiF	AcO ₂	AcO ₂	77.5mol%LiF
Fuel fraction [vol%]	41.1	41.1	100	30.8	30.8	100
Coolant fraction [vol%]	32.7	32.7		53.6	53.6	
Steel fraction [vol%]	26.2	26.2	0	15.7	15.7	0
Core actinide inventory [t]	10.1	9.5	26.4	51.1	72.1	33.8
Actinide inventory in the blanket [t]	0	0	17.5	0	0	17.5
Feed**	U-TRU	Th-TRU	Th-TRU	U	Th	Th
CR*** [-]	0.45	0.39	0.77	1.0	1.0	1.0
TRU burning rate [kg/GW _{th} -yr]	186	226	153	0	0	0
Average burn-up [GWD/t _{HM***}]	110	115	622	64	64	246
Reprocessing strategy	1/3 of the core each year	1/3 of the core each year	3.3 liters per day	1/3 of the core each 2 years	1/3 of the core each 2.83 years	6.5 liters per day

*Ac indicates actinides

**TRU from once through LWR after 5 years of cooling [12]

*** Evaluated considering ²³²Th and ²³⁸U as fertile isotopes and all the other isotopes as fissile. It corresponds to the “TRU regeneration rate” defined in Ref. [14]

****HM indicates Heavy Metals

The European Lead SYstem (ELSY) [12] is selected as representative of a breakeven FR operating in a closed cycle. It is an iso-breeder (CR=1) lead-cooled FR operating on U-PuO₂ fuel (U-ELSY in Table 1). A ThO₂-based iso-breeder version of ELSY (Th-ELSY) has also been developed (see Refs. [5,6] for details) featuring same pin and assembly designs as the U-based counterpart [12] but a ~50% taller active fuel. The increase in the fuel height was adopted to overcome the lower breeding of Th compared to U while preserving the cross-sectional flow area. Velocities and axial temperature rise in the core (two major constraints in the ELSY design [12]) are thus unchanged if the flow rate is unchanged. A 50% increase of the core inventory negatively impacts economics but, by preserving the fuel burn-up, does not affect the required reprocessing capacity (in t_{HM}/yr).

As an alternative route toward a safe and sustainable nuclear energy production, the performances of the Molten Salt Fast Reactor (MSFR) [13], which represents the circulating-fuel reference configuration in the framework of the GIF-IV, are finally investigated. The MSFR has been developed to use thorium as fertile material. Both an iso-breeder and a TRU-burner reactor are here considered. Use of the MSFR with a variable CR is made possible by its on-line reprocessing system, which guarantees the possibility to affect the neutron economy through the reprocessing rate. The CR equal to 0.77 is a lower limit dictated by the solubility limit of Fission Products (FP) in the molten salt. Further details about the MSFR modelling and design can be found in Refs. [7-9,13].

4 WASTE MANAGEMENT, FUEL HANDLING AND PROLIFERATION RESISTANCE

One claimed advantage of thorium is the low build-up of TRUs fostered by its low mass number, which is expected to limit radiotoxicity and decay heat generation in a closed cycle, such as that pursued by the proposed iso-breeder concepts. Figs. 1a and 1b show the specific

radiotoxicity and the radiotoxicity generation for the iso-breeders Th-ELSY, U-ELSY and MSFR. Results have been obtained using equilibrium compositions [5-9], thus representing asymptotic upper limits [5,6]. Only the actinide contribution is included since FPs have a minor impact in the long term [9,15].

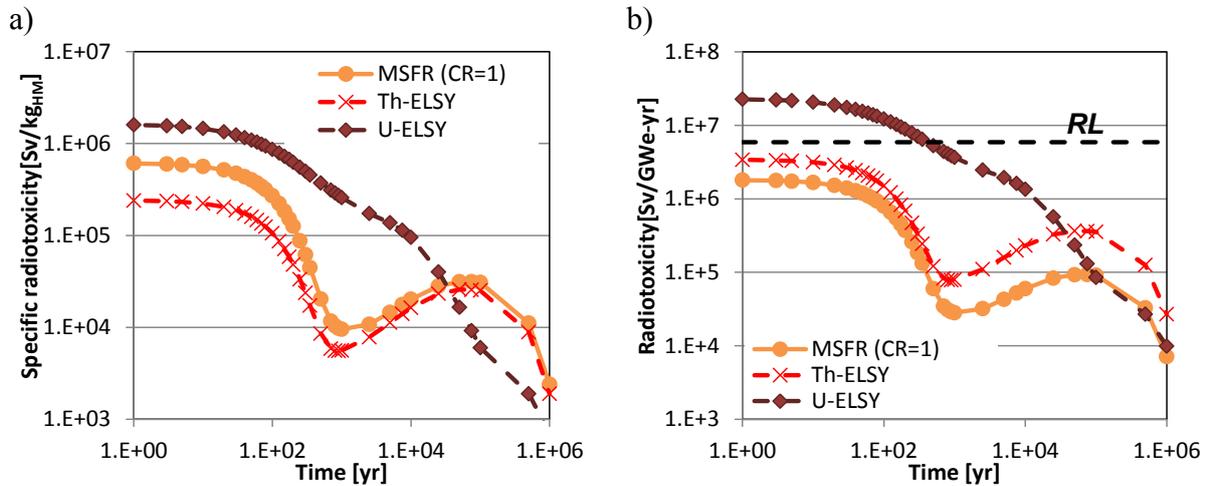


Fig. 1: a) Specific radiotoxicity and b) radiotoxicity generation for the iso-breeder concepts of Table 1, considering only the actinide contribution [8,9]

Fig. 1a shows that the Th cycle features noticeably lower specific (per kg of HM) fuel radiotoxicity for the first 25000 years, which is a consequence of the reduced TRU build-up [5,6]. On the other hand, ^{233}U and ^{234}U are characterized by highly radiotoxic progenies, whose (slow) build-up causes the radiotoxicity of the Th option to surpass the U counterpart in the long term. The slightly higher radiotoxicity for the MSFR compared to the Th-ELSY is a consequence of the softer neutron spectrum and ensuing higher build-up of TRUs (especially the ~ 90 -yr half-life ^{238}Pu) [7-9,13].

Fig. 1b shows the radiotoxicity generation per unit energy for the three options. It can be derived from the specific radiotoxicity dividing it by the fuel burn-up (as GW_e-yr/kg) and multiplying by the fabrication and reprocessing losses (here considered equal to 0.1%). Thanks to the extremely high average burn-up (see Table 1) that can be achieved with a liquid fuel, the MSFR features the lowest radiotoxicity generation despite the higher specific radiotoxicity compared to the Th-ELSY. Fig. 1b also plots the adopted Reference Level (RL) for radiotoxicity, corresponding to that of the natural uranium required to fuel a once-through LWR [5,6]. While 300-400 years are required for the U-ELSY radiotoxicity to cross the RL, the radiotoxicity generated by the Th-ELSY is already well below it at the beginning of the decay. On the other hand, the long term radiotoxicity of Th-ELSY is higher than that of U-ELSY.

The decay heat curves are similar to those reported in Fig. 1 for radiotoxicity [5,6,9]. The decay heat in the first few thousand years can be taken as a rough indicator of the number of geological repositories required for disposal of high level wastes. The long term behaviour is instead of limited concern, which suggests in this case a clear advantage for the Th option.

When FPs are taken into account, the advantages of Th from the lower actinide decay heat are partially offset by the 3-time higher ^{90}Sr fission yield for ^{233}U compared to ^{239}Pu . In fact, the 30-year half-life ^{90}Sr and ^{137}Cs are responsible for most of the heat load in the hundred years after fuel discharge [16]. An increased ^{90}Sr production will then result in incremental costs either for an interim storage or for the initial forced cooling in a geological repository. As concerns the impact of FPs on radiotoxicity, ^{126}Sn , one of the long-lived FPs,

features a slightly higher yield in the Th case, but the impact is negligible compared to that of the actinides [9].

Finally, a peculiar aspect of the Th cycle is related to the build-up, during irradiation, of ^{232}U (~1500 ppm in U for ELSY and MSFR and 4500 ppm for the ARR), whose decay progeny includes high energy gamma emitters, including 2.6 MeV gammas from Tl-208. While this may discourage proliferation [17], it greatly complicates fuel handling and manufacturing and historically represented a large obstacle to the implementation of the thorium cycle. From this perspective, the development of liquid-fuelled reactors like the MSFR appears particularly attractive as fuel handling would be greatly simplified, if not avoided. In addition, the high achievable burn-up enables low reprocessing requirements of few litres per day (Table 1).

5 SAFETY ASPECTS

Table 2 summarizes the safety-related parameters for the investigated FRs as computed for the equilibrium core configuration [5-9].

Table 2: Equilibrium safety parameters for the different reactor concepts [5-9]

	TRU burner concepts			Iso-breeder concepts		
	U-ARR	Th-ARR	MSFR	U-ELSY	Th-ELSY	MSFR
Doppler coefficient [pcm/K]	-0.38	-0.35	-1.95	-0.55	-0.79	-2.91
Coolant expansion coefficient [pcm/K]	-0.63	-0.89	-	0.53	0.26	-
Radial core expansion coefficient [pcm/K]	-0.90	-0.87	-	-0.61	-0.44	-
Fuel expansion coefficient [pcm/K]	-0.09	-0.07	-2.75	-0.18	-0.13	-2.67
Generation time [μs]	0.40	0.42	0.95	0.77	1.03	0.95
Effective delayed neutron fraction β_{eff} [pcm]	317	287	318*	320	326	335*
Reactivity swing in a cycle [pcm]	5050	5528	0	450	378	0
Reactivity insertion following voiding of the active core [pcm]	1267	764	-	5540	2415	-

* Evaluated for static fuel. Reported values are expected to be reduced by approximately 50% by fuel salt recirculation [18].

5.1 TRU-burner concepts

FRs are known to have unique TRU burning capabilities thanks to the improved neutron economy and higher chance of direct destruction by fission in fast-spectrum compared to thermal-spectrum reactors [15]. For a given reactor power, TRU burning rate increases with decreasing CR. One of the limits in CR reduction is represented by the degradation of some safety parameters like void reactivity, Doppler coefficient and β_{eff} [19]. Th can be advantageous to a burner design by allowing a reduction in the CR with respect to U-fuel while complying with safety requirements.

Results summarized in Table 2 actually show comparable reactor safety feedbacks for the Th-ARR with respect to the U-ARR in spite of the lower CR (Table 1). More specifically, Doppler, fuel expansion and radial core expansion coefficients are slightly reduced in the Th-ARR vs. U-ARR but the related safety deterioration is offset by a marked improvement in the coolant expansion coefficient.

Generation time is practically unaffected while β_{eff} and reactivity swing worsen. In fact, the typically low β_{eff} caused by a high TRU inventory is compensated in the U-ARR by a relatively large contribution to fissions from ^{238}U , featuring a high delayed neutron yield, while the much lower fission rate of ^{232}Th makes partly ineffective its potential beneficial contribution to the overall β_{eff} . A main consequence of reduced β_{eff} and increased reactivity

swing is related to the number of necessary control rods. In fact, it is common practice to limit a control rod worth to 0.8 \$ [20], which leads to an estimated number of control rods only to balance the reactivity swing equal to 24 for the Th case compared to the 20 of the U case. In addition, a reduced β_{eff} implies generally quicker transients and thus the necessity of a control system with improved performance.

On the other hand, reactivity insertion due to core voiding is drastically reduced in the Th core. In case of sodium-cooled FRs, core voiding may happen during accidental transients leading to sodium boiling (typically double-fault accidents like an unprotected transient overpower or an unprotected loss of flow) and may exacerbate the consequences of these accidents. The improvement achieved by using Th as support fertile material then emerges as a major advantage of this fuel cycle option. Such improvement becomes even more effective if one considers that sodium boiling may propagate from the active core to the upper plenum. In this case, the reactivity insertion would be drastically reduced by the increased axial leakages. For the U option, the reactivity insertion would be close to zero while, for the Th option, reactivity insertion would become strongly negative (~ -600 pcm), thus suggesting major improvements of the reactor safety during severe accidents. Void reactivity reduction in Th is mainly related to the higher energy threshold and lower value of the fission cross-section of ^{232}Th compared to ^{238}U , which helps limiting the increase in neutron production following a spectrum hardening [5-9]. The effects related to the presence of ^{233}U are of minor importance as a consequence of the low build-up in the driver fuel relatively to the TRU content (approximately 0.5 t of U-233 at equilibrium, to be compared with the 2.2 t of Pu and the 0.5 t of minor actinides (Am, Cm, Np)).

As regards the MSFR, the relatively high CR limits the TRU-burning rate compared to the ARR (Table 1). On the other hand, safety parameters are drastically improved. The softer spectrum compared to liquid metal FRs is disadvantageous in terms of TRU build-up but also implies a higher fraction of neutrons in the resonance region, which in turn improves the Doppler coefficient. In addition, the liquid fuel and high volumetric expansion coefficient ($\sim 1.8 \cdot 10^{-4} \text{ K}^{-1}$) determine a strong negative feedback. In fact, a fuel temperature increase causes part of the fuel to be pushed out of the core. Finally, there are no positive feedback coefficients in the MSFR (the molten salt plays the roles of both fuel and coolant) and generation time is longer compared to that of the ARR. The main drawback is related to the β_{eff} , whose value is expected to be reduced by more than half by the fuel recirculation and by the related out-of-core decay of the delayed neutron precursors [18]. However, the importance of β_{eff} is reduced for the MSFR since a reactivity swing equal to zero could ideally be achieved by means of a proper on-line reprocessing strategy. Nonetheless the MSFR dynamics is not adequately known and dedicated simulation tools would be required to better assess its potential safety improvements.

5.2 Iso-breeder concepts

In the iso-breeder reactors, the only feed is represented by fertile isotopes (either U or Th), which then determine the prevailing fissile isotopes in the core (either ^{239}Pu or ^{233}U). Consistently, differences in safety parameters between U and Th options in the ELSY are amplified compared to what observed in the ARR, where most of the fissions derive from the same TRUs. In particular, void reactivity and coolant expansion coefficient in the Th-ELSY are less than twice those in the U-ELSY. In addition, in the Th-ELSY the Doppler coefficient and generation time are improved compared to the U counterpart, while the β_{eff} is not reduced contrarily to the Th-ARR vs. U-ARR.

The trend of ^{233}U fission cross-section with neutron energy explains most of the additional improvements observed in the transition from U to Th fuel in ELSY compared to the ARR. In fact, the ^{233}U fission cross-section strongly decreases with energy (whereas that of ^{239}Pu is nearly flat) which leads to a negative reactivity insertion as a result of spectrum hardening, thus reducing void reactivity and coolant expansion coefficient. Accordingly, fuel expansion and radial core expansion are less effective in the Th-ELSY (they affect reactivity mainly through a spectrum softening following the increased lead-to-fuel ratio [21]) but their contribution to safety is expected to be limited. The negative reactivity insertion due to ^{233}U in case of spectrum hardening explains also the improved Doppler coefficient. The Doppler coefficient is the result of increased captures in the fertile isotopes, which in turn leads to a local spectrum hardening and, due to the presence of ^{233}U in Th-ELSY compared to Pu in U-ELSY, to a further improvement in Doppler. Finally, a strongly decreasing fission cross-section with energy concentrates fissions at lower energies, thus increasing the generation time. A higher delayed neutron yield of ^{233}U compared to TRUs is instead the reason for an improved β_{eff} for the Th-ELSY compared to the Th-ARR.

It is interesting to observe that the reactivity swing is slightly improved for the Th ELSY compared to the U counterpart. In fact, at equilibrium all isotopes feature a constant mass during irradiation, except for the main fertile isotopes that, during each cycle, are partially consumed and substituted by FPs. The lower reactivity swing for the Th option is then explained by: 1) the lower one-group effective capture cross-section of ^{233}U FPs (0.17 b) compared to ^{239}Pu FPs (0.26 b) and 2) the larger one-group effective capture cross-section of ^{232}Th (0.34 b) compared ^{238}U (0.31 b).

Finally, the iso-breeder MSFR features improved safety parameters compared to the ELSY similarly to what observed for the TRU-burner MSFR compared to the ARR. Comparing the two MSFR designs, the increased ^{233}U (and lower TRU) content of the iso-breeder leads to improved safety parameters compared to the TRU-burner option. In particular, the Doppler coefficient is increased by $\sim 50\%$.

6 CONCLUSIONS

In this paper, thorium use in FRs is confirmed as beneficial in terms of actinide waste management, particularly in view of the low radiotoxicity and decay heat in a Th closed cycle. However, a higher ^{90}Sr fission yield partly offsets the decay heat advantages. TRU burning capabilities are improved thanks to the lower CR achievable while complying with safety constraints, with the caveats of a greater number of control rods required and a control system with a quicker response. Safety improvements, especially in terms of a drastically reduced void reactivity, emerge as a major advantage of the Th-option. In the Th-based ARR, void reactivity becomes strongly negative if one considers that sodium boiling during an accident may extend to the upper plenum. Nonetheless, thorium use also presents considerable challenges and requires R&D efforts well beyond those expected for the development of uranium-based FRs. Irradiation experience for thorium-based FR fuel is poor, and reprocessing techniques have never reached an industrial scale of application, and still needs to be demonstrated for TRU-bearing Th-based transmutation fuel. The gamma emission from ^{232}U 's progeny will inevitably result in additional costs, and challenges, for fuel reprocessing, handling and fabrication. The MSFR can alleviate this problem by avoiding the fuel fabrication step and by limiting fuel reprocessing requirements (but it will require developments in pyro-reprocessing, including its industrial scalability). The MSFR also shows potential benefits in terms of safety features and fuel cycle flexibility. The drawback is

the limited know-how available for fast-spectrum MSRs, whose deployment is hard to envisage in the near term despite the growing interest of the nuclear community.

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