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Analysis of Thorium Performance in Lead cooled Fast Reactor

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ABSTRACT

The exploitation of thorium fuel in present and future reactors is essential for sustainability of nuclear energy. In this work the performance of thorium fuel in fast reactors as alternative fuel for the standard natural uranium fuel is analyzed. The European Lead cooled Fast Reactor (ELFR) is an advanced system that has been chosen for research and development by generation-IV (GENIV) initiative. A representative fuel assembly of this fast spectrum concept was modeled using MCNPX transport code. The performance of thorium-based fuel is investigated and analyzed in the ELFR concept using the modeled assembly. Also, for the sake of comparison with the studied case, the reference (uranium) case for the modeled ELFR assembly has been also simulated. The analysis and comparison included a number of important neutronic and safety parameters such as the infinite multiplication factor (k- ∞), neutron yield, energy of neutrons causing fission, effective delayed neutron fraction (β -eff), and Doppler effect. The performance analysis has showed that using thorium has a positive impact on evolution of the multiplication factor and can help in degrading the plutonium vector.

Graphical abstract



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1. Introduction

It is anticipated that low-carbon technology will see a significant rise in global energy demand in the future. As a result, nuclear energy emerges as an important option for meeting this demand with the least amount of impact on the environment (1). The so-called generation IV (GENIV) reactors are currently being developed by a number of nations. Six cutting-edge reactors were chosen by these nations in 2002, and it is anticipated that they will be commercially available by 2030 (2). The GENIV concepts promise to be more costeffective, secure, long-lasting, and resistant to proliferation than previous generations. In order to achieve the objective of sustainability, there must be fuel alternatives that enable a better and more effective use of the resources at hand in order to extend the useful life of nuclear energy. The majority of nuclear reactors in use today use MOX and UO2 as fuel. On the other hand, thorium (232Th) ore is regarded as an excellent candidate for use as nuclear fuel due to its abundance in the Earth's crust, which is three to four times greater than that of uranium (3). However, since it lacks any isotope that can undergo fission, it cannot be used by itself to initiate a fission chain reaction. Nonetheless, it can be transformed into the fissile isotope ²³³U, and fast reactors are a good option for carrying out this process because the high neutron flux increases the chance of this transmutation.

Although, there are several economic and technical obstacles making the deployment of thorium challenging. There have been a number of thorium research facilities built in the past, including Elk River BWR, Peach Bottom HTR, Edison Indian Point-1 PWR, Shippingport PWR, etc. In addition to these efforts recent neutronic studies on thorium have showed it can be employed in advanced reactors. The suitability of the use of thorium in a number of GENIV reactors: the Very-High-Temperature Reactor (VHTR), the gas cooled fast reactor (GFR), the SuperCritical-Watercooled Reactor (SCWR), the Sodium cooled Fast Reactor (SFR) and the Lead cooled Fast Reactor (LFR) had been analyzed using infinite lattice models of fuel assembly of each concept in (4). Also, in relation to Sodium cooled Fast Reactor (SFR), a comparison of thorium and uranium fuel was conducted in (5). In addition, a neutronic model of a fast reactor core (an ASTRID-like) with thorium-based fuel was proposed in (6). In relation to LFRs, a comparative neutronic analysis of the design of thorium-based fuel cores with both homogeneous and heterogeneous configurations was carried out in (7). Under both open and closed fuel cycles, the viability of thorium as a fuel in the Gascooled Fast Reactor (GFR) 2400 was investigated in (8). Additionally, in a heterogeneous model with a 2400 MWth GFR and a single batch irradiation scheme, thorium utilization was investigated in (9).

Also, the behavior of molten salt iso-breeder reactor (MSiBR) system fueled with a thorium was investigated

in (10). Moreover, thorium fuel was investigated as alternative fuel in thermal reactors such as Candu and PWR (11, 12).

The primary goal of this work is to examine the neutronic performance of thorium fuel in the European Lead-cooled Fast Reactor (ELFR) rather than the reference uranium fuel. MCNPX was used to simulate a representative ELFR fuel assembly. The ELFR uses a mixed oxide (MOX) fuel with a reference fuel vector made up of an equilibrium mixture of uranium, plutonium, and minor actinides (U/Pu/MAs). To investigate the behavior and performance of the fertile material ²³²Th as a fuel in the ELFR concept, the reference fuel vector was modified as follows: first, all uranium isotopes were replaced by ²³²Th while leaving the Minor Actinides (MAs) vector unchanged; second, the Pu content was increased to achieve the same initial value of infinite multiplication factor $(k-\infty)$ for reference model.

2. ELFR

With a thermal power of 1500 MW, ELFR is an upgraded design of its predecessor European Leadcooled System (ELSY). It is pool-type and uses pure lead as a coolant (13). The density of the exit lead above the active core is 10.48 g.cm⁻³ at 480 °C, whereas the density of the inlet lead below it is 10.58 g.cm⁻³ at 400 °C. The reactor core has 427 fuel assemblies (FA), each of which is made up of 169 fuel pins with a 140 cm active length. The reference fuel is an equilibrium blend of MOX/MAs fuel, and the clad is constructed of a special Stainless Steel called T91 (14). To flatten the power radially through the core while keeping the fuel enrichment (in terms of Pu) constant, the active core is partitioned into two kinds of fuel assemblies with different volumetric fractions of fuel: 157 FAs with a central pellet hole radius of 2 mm and 270 FAs with a central pellet hole radius of 1 mm. The pellet hole is also used for gas clearance. On the other hand, the use of lead as a coolant in ELFR has various advantages. Lead, unlike other metals, does not interact with air or water, has a substantially high boiling point (1745 °C), and has a low absorption cross-section for fast neutrons.

Figure 1 depicts the geometrical arrangement of the fuel cells assembly and fuel pin (at fuel level). The MCNP designed model of the ELFR's fuel assembly is based on the reference design parameters (13). However, for the modelled fuel assembly to be reflective of the whole core it was modelled with an average fuel volume (13199 cm³). The inner and outer FAs' fuel volumes were estimated and averaged. The obtained mean volume (13199 cm³) was utilized to calculate the radius of the center pellet hole (0.1581 cm) employed in the model, while the outside radius of the fuel pellets remained unaltered. The reference enrichment of Pu in MOX fuel pellets is 18.15 w%, which corresponds to the core-average value. The isotopic fractions of the reference fuel at the beginning of life (BOL) are given in Table 1.



Fig. 1 Geometrical configuration of the ELFR fuel assemblies at left and pin lattice configurations at right.

Table 1 Reference fuel compositions at BOL (13).

Isotope	wt.%	Isotope	wt.%
²³⁴ U	0.002	²⁴¹ Am	1.016
²³⁵ U	0.325	^{242m} Am	0.003
²³⁶ U	0.008	²⁴³ Am	0.218
²³⁸ U	80.17	²⁴³ Cm	0.0009
²³⁸ Pu	0.423	²⁴⁴ Cm	0.041
²³⁹ Pu	10.32	²⁴⁵ Cm	0.016
²⁴⁰ Pu	4.90	²⁴⁶ Cm	0.0012
²⁴¹ Pu	1.108	²⁴⁷ Cm	0.000023
²⁴² Pu	1.396	²⁴⁸ Cm	0.000002
²³⁷ Np	0.05		

3. Neutronic Calculations

Calculations for the GENIV ELFR were done using MCNPX code (15) and the ENDF-VII cross section library. This study involved modelling and simulation of a single representative assembly of the reactor core. The fuel assembly was modelled using an infinite lattice with just radial reflective boundary requirements. Where the primary fertile component in the fuel matrix is changed from the natural uranium vector to thorium. ²³²Th was employed to substitute uranium isotopes in the UO₂ matrix, mixing thorium $(^{232}$ Th) similarly to uranium isotopes. Nevertheless, plutonium enrichment was raised from 18.15 w% for reference fuel to 21.90 w% for the thorium-reactor grade plutonium (Th-rgPu) fuel due to ²³²Th's lower density and poorer reactivity when compared to U (3). This enrichment achieves the same initial $k-\infty$ value (1.0778) of the reference uranium model.

For the modelled assembly, the burnup period was equal to the reactor cycle duration, i.e. 1800 Effective Full Power Days (EFPDs). The first time step was after three EFPDs to account for the buildup of Xenon (Xe) in fuel and its poisonous influence on reactivity. The burnup was estimated for a fuel assembly with an average power of 3.51 MW. There were 10,000 starting source neutrons every cycle, 150 total cycles per time step, and 25 cycles that were skipped.

4. Results and Discussion

4.1. Neutronic and Safety Characteristics

The neutronic and safety of the reactor is characterized by main parameters such as neutron yield, energy of neutrons causing fission, the Doppler effect, and the effective delayed neutron fraction (β -eff). The results of these parameters are summarized and compared in Table 2.

The results show that for both thorium and reference fuel, the percentages of fission caused by thermal, intermediate, and fast neutrons are comparable, and that nearly all fission events are caused by both intermediate and fast neutrons, which is advantageous for fast spectrum concepts like ELFR. Also, for both fuel models, the average energy of the neutrons that cause fission and the number of neutrons emitted per fission are both reasonably high and comparable, as would be expected for fuel containing high percentage of fissile Pu.

The safety- related characteristics for both the proposed Th-rgPu fuel and the reference U-rgPu fuel, Doppler effect and the effective delayed neutron fraction (β -eff)

are estimated at BOL conditions (8). The calculated Doppler Constant is somewhat more negative (-1003.7 pcm) than the value for uranium fuel (-979.6 pcm). This improvement is due to ²³²Th's greater energy threshold and smaller fission cross-section when compared to 238 U (16). The obtained value of β -eff (249 pcm) is equivalent to that of previous fast reactors powered by thorium, but it is lower than that of the uranium-based fuel (314 pcm), which is seen as a significant safety flaw. Similar outcomes were attained by (8) for the thorium-fueled gas-cooled fast reactor (GFR2400). This degradation is brought on by the fact that thorium fuel has a smaller fertile proportion than uranium fuel. Moreover, ²³⁸U contributes significantly to fission and has a very large delayed neutron output, both of which assist to increase the uranium fuel's β -eff. Moreover, thorium-based fuel has a greater Pu proportion than uranium-based fuel, which results in a sizable fission share from Pu isotopes and their low delayed neutron yield.

The variation of $k-\infty$ for the proposed Th-rgPu fuel and the reference U-rgPu fuel is shown in Fig. 2. The following can be observed for Th-rgPu fuel the k-∞ value dropped steadily throughout the first 150 days, dropping by around 337 pcm to its lowest value (1.0743 ± 23) below the initial starting point (1.0778 ± 21) . This was caused by the buildup of Protactinium (²³³Pa), which has a high capture cross section and acts as a neutron poison in comparison to the fertile isotope ²³²Th. According to Fig. 2, the ²³³Pa requires 150 effective full power days (EFPDs) to achieve its maximum concentration in the fuel. After 150 effective full power days EFPDs, the $k-\infty$ value begins to increase again powered by the buildup of ²³³U

in the fuel. After about 600 EFPDs, the k- ∞ value reaches its initial value and then increases notably until it reaches its maximum value of 1.0832±27 (at around 1250 EFPDs). There after $k-\infty$ value start to decrease until it reaches its initial value again at the end of the fuel cycle, due to fuel depletion and fission products accumulation with a final average burnup of 58 MWd/kgU. A similar pattern of behavior was noted in another investigation (4). It is evident that the evolution of $k-\infty$ for the Th-rgPu fuel follows a pattern that differs from that of the reference fuel, with a reactivity difference of 3138 pcm at the end of the reference operating cycle (1800 EFPDs). The creation of ²³³U by breeding from ²³²Th is significant for the Th-rgPu fuel, as seen in Fig. 3. This is accomplished through the fission of ²³⁹Pu (see table 3). The generation of ²³³U maintains high reactivity and allows for a longer fuel cycle as compared to the reference fuel cycle.



Fig. 2: Evolution of k-∞ factor for Th-rgPu and reference UrgPu fuel.

Table 2: Safety parameters of Th-rgPu as well as reference U-rgPu fuel at BOL.

Neutronic and safety parameter	Th-rgPu	U-rgPu	
Percentages of fission caused by:	0.01/43.56/56.43	0.01/44.56/55.43	
thermal/intermediate/fast neutrons [%]			
Average neutron energy causing fission [MeV]	0.50	0.62	
Average number of neutrons released per fission	2.93	2.92	
Doppler constant [pcm] ^a	-1003.7±33	-979.6±31	
β-eff [pcm]	314±26	249±23	

a: due to an assumed variation of the fuel temperature from a perturbed value of T_{per} = 300 K to a reference value of T_{ref} =1200 K.

4.2. Actinides inventory

Figure 3 depicts the mass variations of the ²³³Pa, ²³²Th and ²³³U. Because of its relevance in nonproliferation problems, ²³³Pa was considered as it is the parent of ²³³U. Both ²³³U and ²³³Pa are generated from ²³²Th through radiative capture:

232
Th $(n,\gamma) \rightarrow ^{233}$ Th $(\beta^{-}) \rightarrow ^{233}$ Pa $(\beta^{-}) \rightarrow ^{233}$ U

As a result, the mass of ²³³U and ²³³Pa increases at the expense of the loss of ²³²Th content. In contrast, the capture cross section values for ²³³Pa are greater than those for ²³²Th over fast spectrum, explaining the decline in reactivity during the initial time period of fuel burnup (see Fig. 2).



Fig. 3 The evolution of ²³³Pa, ²³²Th, and ²³³U masses.

The variations in Pu isotope composition with fuel burnup time for the Th-rgPu fuel is shown in Fig. 4. Because ²³⁹Pu is the predominant fissile isotope present at BOC for the Th-rgPu fuel, its mass decreases considerably throughout fuel burnup, as demonstrated. Moreover, by neutron capture in ²³⁹Pu, the other Pu isotopes, including the initially loaded ²⁴⁰Pu, steadily accumulate. The Pu vector's composition at the end of the cycle is 3.10 % ²³⁸Pu, 47.20 % ²³⁹Pu, 34.40 % ²⁴⁰Pu, 5.80 % 241 Pu, and 9.50 % 242 Pu. As the concentration of ²³⁹Pu declined by 9.60% while the concentration of ²⁴⁰Pu grew up by 7.40%, which shows that the Pu vector is considerably degraded. At the end of the radiation session, 35.2% of the ²³⁹Pu that was initially loaded had been consumed. Similar results were obtained for thorium-fueled gas cooled fast reactor (4).



Fig. 2: The evolution of Pu isotopes with burnup time.

In Table 3, fuel inventories for the reference and ThrgPu fuels are shown for both beginning of cycle (BOC) and end of cycle (EOC). These results show that ²³⁸U and the Pu isotopes (²³⁸Pu to ²⁴²Pu) are the primary isotopes found at EOC for the U-rgPu. On the other hand, ²³⁸U is reduced while the total Pu content nearly stays same. However, compared to the usage of ²³⁸U, the use of thorium as a fertile material seems to yield significantly fewer Pu isotopes. This may be observed in the difference in Pu relative inventory between BOC and EOC, for example, in the Th-rgPu case where it decreased by 4.76% whereas it nearly stayed constant for the U-rgPu case. This result is attributed to the different breeding pathway in U and Th, i.e. $^{238}U \rightarrow ^{239}Pu \text{ vs. }^{232}Th \rightarrow ^{233}U$, respectively.

Also, the results show that the Th-rgPu model's discharged fuel is distinguished by a somewhat larger buildup of minor actinides (Np, Am, and Cm) in comparison to the U-rgPu model. As these elements are created by neutron capture in Pu isotopes, raising the Pu concentration of the thorium-based fuel increase its production. Nonetheless, the overall MA content is reduced by the EOC.

The fissile inventory ratios (FIRs) for both reference and thorium-based fuel are also included in Table 3. As shown switching from U to Th as a fertile material reduces the FIR by over 3%, from 96.47 to 93.57%. The Th-rgPu fuel exhibits a greater TRU burning rate as compared to its equivalent U-rgPu fuel due to the lower FIR. This is demonstrated by comparing the discharged actinide stocks for the two fuels in Table 3. The reduced Th-rgPu's FIR may be attractive for fuel cycle strategies intended for burning nuclear waste.

 Table 3 BOC and EOC relative fuel inventories for both fuel models.

	Reference fuel		Th-rgPu [m/m%]	
Element	[m/m%]			
	loaded	discharged	loaded	discharged
Th-232	-	-	76.75	71.6
Pa-233	-	-	-	0.11
Total U	80.50	75.20%	-	4.03
U-233	-	-	-	3.84
U-234	0.002	0.02	-	0.18
U-235	0.325	0.20	-	0.007
U-236	0.008	0.04	-	0.003
U-238	80.17	74.95	-	-
Total	18.15	18.16	21.9	17.14
Pu				
Pu-238	0.423	0.49	0.51	0.54
Pu-239	10.32	10.32	12.46	8.10
Pu-240	4.90	5.16	5.91	5.89
Pu-241	1.108	0.84	1.34	0.99
Pu-242	1.396	1.35	1.68	1.62
Total	1.347	1.272	1.347	1.335
MA				
Np-237	0.05	0.0544	0.05	0.0408
Am-241	1.016	0.836	1.02	0.872
Am-	0.003	0.017	0.003	0.0158
242m	0.005	0.017	0.003	0.0158
Am-243	0.218	0.266	0.218	0.268
Cm-242	-	0.0337	-	0.0306
Cm-243	0.0009	0.00429	0.0009	0.00353
Cm-244	0.041	0.0939	0.041	0.087
Cm-245	0.016	0.0161	0.016	0.0153
Cm-246	0.0012	0.00271	0.0012	0.00248
Cm-247	0.000023	0.00019	0.00002	0.00016
Cm-248	0.000002	0.0000014	0.000002	0.000011
FP	-	5.37	-	5.79
Total	100	100	100	100

The performance of thorium and its impact on safety and neutronic characteristics, including the infinite multiplication factor, Doppler effect, delayed neutron fraction, and fuel transmutation, were examined in relation to the European GENIV reactor ELFR. Calculations were performed on a core representative fuel assembly. The performance analysis's findings show that using thorium has a positive impact on how the multiplication factor evolves. Whereas, the low reactivity loss during cycle can be employed to achieve a longer fuel cycle or less initial reactivity (less initial fissile content) is needed to achieve a target fuel cycle length. To get the same starting multiplication factor, the fissionable isotope concentration must be raised by around 4%. Nonetheless, the amount of ²³³U that was created demonstrated that the ELFR is a promising concept in this aspect. By the end of cycle, ²³³U is still available (about 4% of fuel content) and can be reused in the thorium model. Despite of the variations in delayed neutron fraction when a full-core is studied, the delayed neutron fraction is low in the reference scenario and grows lower when thorium is used, which can be a serious safety concern that must be addressed.

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