



Neutronic Evaluation of MSBR System Using MCNP Code

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ABSTRACT

The concept of Molten Salt Reactor use Th to breed fissile ^{233}U , where an initial source of fissile material needs to be provided. However, there is no available ^{233}U and so; the fissile fuel supply is one of the unresolved problems. Thus, it is necessary to use existing fissile materials such as ^{235}U or Pu to produce ^{233}U . Current studies analyze the fuel transition from $^{235}\text{U}/\text{Th}$ or Pu/Th to $^{233}\text{U}/\text{Th}$ and, in this context, the present work evaluates the criticality and the neutron flux of MSBR (Molten Salt Breeder Reactor) considering the fuel: (i) mix of Th and enriched U; (ii) the combination of Th and reprocessed Pu; and (iii) matrix of reprocessed Pu/minor actinides (MAs) and Th. The goal is to verify which of these fuels can be used as initial fissile supply. The MSBR core was simulated by MCNPX 2.6.0 code and the criticality model presents similar behavior of previous studies. The results show that reprocessed fuels could have a potential to be used as initial fissile supply, but these fuels present a neutron flux profile less flattens than traditional $^{233}\text{U}/\text{Th}$. It is possible that a new distribution of fuel elements may improve this profile and future simulations will be performed to evaluate this behavior. The uranium, must has high enrichment value to be used as initial seed. Other studies need be performed to evaluates the uranium enrichment and the U/Th ratio that produces similar core criticality to traditional fuel.

Keywords: MSBR, Nuclear Fuel Cycle, Neutronic Simulation, MCNPX.

1. INTRODUCTION

The energy demands are expected to grow at different rates around the world. Several studies have shown that nuclear power plants may represent very attractive options to the energy generation especially due to greenhouse gas emissions reductions. However, the economics, proliferation, safety, and waste production are major barriers to the expansion of nuclear power. In this context, the new generation of nuclear systems has the goal minimize such barriers. The MSR (Molten Salt Reactor) is one of the promising future nuclear reactor concepts included in the Generation IV roadmap. Such reactor concept has already been tested in the past through ORNL (Oak Ridge National Laboratory) investigations, with experimental studies (Aircraft Reactor Experiment and Molten Salt Reactor Experiment) and with the very complete MSBR (Molten Salt Breeder Reactor) design, though it was never experienced in practice [1]. In a MSR system, the fuel is a mixture of salts, generally molten fluoride containing fissile materials, which circulate between the reactor core and the heat exchanger. Thorium fueled MSR concept is one of the most promising nuclear reactor designs currently being studied, because to the high availability of naturally occurring this fuel. Several works have been studied this nuclear system (e.g. [2-10]) where the main interest is to using ^{232}Th to breed ^{233}U . However, considering that is no available ^{233}U in the nature, it is crucial to analyze which initial fissile material that could give a flexible transition in fuel cycle to MSR system. The traditional concept proposes to use ^{235}U as initial fissile fuel supply, but Pu and minor actinides (MAs) could be used instead of U for initial fissile loading. The present work studies the neutronic behavior of MSBR system using the following initial fissile source: (a) uranium, (b) reprocessed Pu and (c) a matrix of reprocessed Pu/MAs. The goal is evaluates the use these nuclides as initial seed of the system in the fission/transmutation process. The MCNPX 2.6.0 code was used to calculate the criticality and the neutron flux profile at steady state of MSBR [11].

2. METHODOLOGY

2.1. Evaluated Fuels and Material Compositions

The standard fuel salt of MSBR system is a mixture of $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$. In the ORNL report there are two mole percentage of UF_4 , 0.3 and 0.232% which represent a conceptual and an optimized MSBR design respectively [12]. These values correspond to mass percentage (*mp*) of fissile material (^{233}U) of 2.26% and 1.76%. In order to evaluate other nuclides as initial fissile loading in MSBR system, the current study simulates four fuels types considering the two values of fissile content reported by ORNL. The following fuel types were simulated:

- (a) Recovery Uranium (RU): Conventional ^{233}U from the U-Th cycle;
- (b) Enriched Uranium (EU): ^{235}U manufactured by enrichment plant;
- (c) Reprocessed Plutonium (RP): Nuclides of Pu from spent PWR fuel; and
- (d) Reprocessed Actinides (RA): Matrix of Pu/MAs from spent PWR fuel.

Table 1 present the salt composition of the evaluated fuels. The isotopic calculation was based on mole percentage and on *mp* of fissile material. In the heavy metal mass, the highest *mp* of fissile isotopes implies in the lowest thorium concentration. Thorium, uranium, plutonium and minor actinides all form suitable fluoride salts that readily dissolve in the $\text{LiF-BeF}_2\text{-ThF}_4$ mixture. The fissile and fertile isotopes can be easily separated from one another in fluoride form. The isotopic composition of the Pu and Pu/MAs was calculated by previous studies using ORIGEN code [13]. These studies simulate a typical burnup of PWR fuel with 4.5% of initial enrichment at a cycle of 33 GWd/MTU, where the spent fuel remained in the cooling pool for five years. Table 2 present the main characteristics of the evaluated fuels as the heavy metal concentration and the fissile content in U, Pu or Pu/MAs matrix.

In the MSBR, the graphite is the principal material other than salt. The core contains graphite for neutron moderation and reflection. In the simulations the moderator elements and the reflector blocks are made of natural graphite. Also, the reactor vessel is composed of Hastelloy N that is an alloy developed specially for use in molten fluoride systems. Among the major constituents, chromium is the least resistant to attack by fluorides. The chromium content of Hastelloy N is low

enough for the alloy to have excellent corrosion resistance toward the salts. Table 3 present the composition in mass percentage of moderator, reflector and reactor vessel used in the simulations.

Table 1: Isotopic composition (*mp*) of the evaluated fuels.

Salt	Nuclide	Fuel Type							
		RU		EU		RP		RA	
LiF	⁶ Li	0.0004	0.0004	0.0004	0.0004	0.0004	0.0004	0.0004	0.0004
	⁷ Li	7.8500	7.8800	7.8500	7.8800	7.8100	7.8500	7.8100	7.8500
	¹⁹ F	21.3000	21.3000	21.3000	21.3000	21.2000	21.3000	21.2000	21.3000
BeF ₂	⁹ Be	2.2500	2.2600	2.2500	2.2600	2.2400	2.2500	2.2400	2.2500
	¹⁹ F	9.4900	9.5200	9.4900	9.5200	9.4300	9.4800	9.4300	9.4800
ThF ₄	²³² Th	43.5000	43.6000	43.5000	43.6000	43.2000	43.4000	43.2000	43.4000
	¹⁹ F	14.2000	14.3000	14.2000	14.3000	14.1000	14.2000	14.1000	14.2000
UF ₄	²³² U	0.0002	0.0002	—	—	—	—	—	—
	²³³ U	0.9980	0.7740	—	—	—	—	—	—
	²³⁴ U	0.0833	0.0646	—	—	—	—	—	—
	²³⁵ U	0.0077	0.0060	1.0100	0.7810	—	—	—	—
	²³⁶ U	0.0006	0.0004	—	—	—	—	—	—
	²³⁸ U	0.0016	0.0012	0.0954	0.0740	—	—	—	—
	¹⁹ F	0.3560	0.0276	0.3560	0.2760	—	—	—	—
PuF ₃ or (Pu/MAs)F ₃	²³⁸ Pu	—	—	—	—	0.0652	0.0505	0.0655	0.0506
	²³⁹ Pu	—	—	—	—	0.8100	0.6280	0.8140	0.6300
	²⁴⁰ Pu	—	—	—	—	0.4070	0.3150	0.4090	0.3160
	²⁴¹ Pu	—	—	—	—	0.2020	0.1560	0.2030	0.1570
	²⁴² Pu	—	—	—	—	0.1450	0.1120	0.1450	0.1130
	²⁴¹ Am	—	—	—	—	—	—	0.0647	0.0500
	^{242m} Am	—	—	—	—	—	—	0.0002	0.0002
	²⁴³ Am	—	—	—	—	—	—	0.0408	0.0316
	²⁴³ Cm	—	—	—	—	—	—	0.0002	0.000
	²⁴⁴ Cm	—	—	—	—	—	—	0.0167	0.0129
	²⁴⁵ Cm	—	—	—	—	—	—	0.0011	0.0008
	²⁴⁶ Cm	—	—	—	—	—	—	0.0002	0.0002
	²³⁷ Np	—	—	—	—	—	—	0.1390	0.1080
¹⁹ F	—	—	—	—	0.3870	0.3000	0.1120	0.0870	
Fissile isotopes		2.26	1.76	2.26	1.76	2.26	1.76	2.26	1.76

Table 2: Main characteristics of the evaluated fuels.

Description		Fuel Type							
		RU		EU		RP		RA	
Fissile isotopes		2.26	1.76	2.26	1.76	2.26	1.76	2.26	1.76
Heavy Metal	Th	97.40	98.20	97.50	98.10	96.40	97.20	95.80	96.70
	U	2.26	1.76	2.47	1.92	—	—	—	—
	Pu	—	—	—	—	3.63	2.83	—	—
	Pu/AMs	—	—	—	—	—	—	4.21	3.67
Fissile content in U, Pu or Pu/MAs matrix		92.20		91.30		62.10		53.60	

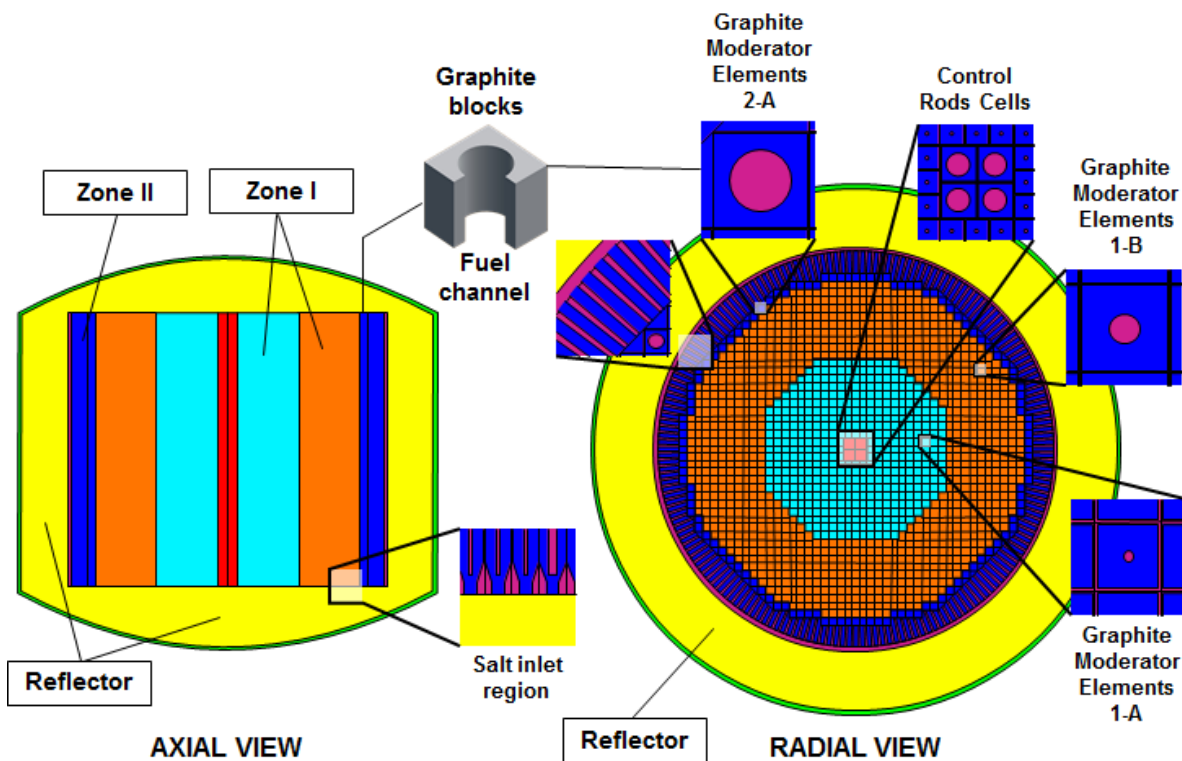
Table 3: Material composition (*mp*) of the principal MSBR components.

Moderator and Reflector		Reactor Vessel – <i>Hastelloy N</i>			
^N C	100	^N Mo	12.0000	Si	0.1000
		Ni	70.1000	^N Cu	0.1000
		^N Cr	7.0000	^N C	0.0600
		^N Fe	5.0000	P	0.0150
		^N Ti	2.0000	^N S	0.0150
		Nb	2.0000	^N W	0.0100
		^N Hf	1.0000	^N Al	0.0100
		Mn	0.2000	¹¹ B	0.0008
		Co	0.2000	¹⁰ B	0.0002

N. indicates the natural isotopic concentration.

2.2. Computational Model of MSBR

The MSBR configurations use the data from a conceptual design developed by ORNL [12]. Figure 1 illustrates the geometry of the simulated system and Table 4 present, the main dimensions of the simulated system.

Figure 1: Model of the MSBR simulated in MCNPX 2.6.0.

The reactor core has a square lattice that contains graphite blocks with a square cross section and a circular fuel channel. In the core center there are four cells for insertion of the control rods that move to displace the salt and regulate the nuclear power and average. Two holes are for safety rods primarily for providing adequate negative reactivity at emergency situations, and two holes are graphite control rods for fine reactivity control. The withdrawal of the graphite control rods will insert negative reactivity to the core due to the decrease of neutron moderation. However, the configured model does not encompass the control rods because the present study does not simulate control rods displacements. The MSBR core presents three regions: central (or Zone I), outer (or Zone II), and reflector. These regions have different moderation-to-fuel ratio (V_M/V_F), about 6.69 and 1.70 for Zone I and Zone II respectively. Between Zone II and reflector there is a gap that has 100% of salt to provide clearance when removing and inserting a core assembly. Zone I contains fuel elements 1-A and 1-B where 1-A has a smaller diameter of the fuel channel diameter than 1-B. This characteristic provides different flow rates of molten salt in the elements 1-A and 1-B and contributes to flatten the power distribution. Zone II is made up of two kinds of elements 2-A and 2-B. The type

2-A is similar to 1-A and 1-B but 2-A has bigger diameter of fuel channel than last ones. The elements 2-B are graphite plates arranged radially around the active core. Between these graphite blocks there is a clearance of 3 cm from each other to allow the molten salt to flow amidst them.

The simulations in MCNPX 2.6.0, consider 200 active cycles with 15000 neutrons per cycle. Eight initial neutron sources were uniformly distributed in active core of the reactor. The estimated standard deviation is around 3×10^{-4} .

The nuclear data were downloaded from ENDF/BVII-1 (Evaluated Nuclear Data File) website [14] and processed with NJOY99 (Nuclear Data Processing System) [15] at the operational temperature of MSBR, about 900 K. Thus, these data was added to the MCNPX 2.6.0 library.

Table 4: Main dimensions MSBR core.

Description		Value (cm)	
Vessel internal diameter		677.00	
Vessel height at center		610.00	
Vessel wall thickness		5.08	
Vessel head thickness		7.62	
Active core height		396.00	
Radial thickness of reflector		76.20	
Axial thickness of reflector		56.20	
Pitch distance of fuel blocks		10.20	
Gap between the fuel blocks	1-A	0.770	
	1-B	0.250	
	2-A	0.250	
	2-B	3.000	
Graphite Fuel Block	Width × Length × Active Height	1-A	9.40 × 9.40 × 396.00
		1-B	9.91 × 9.91 × 396.00
		2-A	9.91 × 9.91 × 396.00
		2-B	5.10 × 26.7 × 396.00
	Fuel hole diameter	1-A	1.50
		1-B	3.40
		2-A	6.60
		2-B	—

2.3. Evaluated Parameters

The MCNPX estimates the effective multiplication factor (k_{eff}) with the respective standard deviation (σ_{ST}) of the simulated model. This work evaluates the criticality and the neutron flux of the MSBR at steady state condition to the EU, RU, Pu and Pu/AMs fuels.

To evaluate the neutron flux distribution in the core, the average neutron flux was calculated by MCNPX to each reactor cell. The TMESH card was used which allows the user to tally particles on a mesh independent of the problem geometry [11]. The used code estimates the flux using the source specified by the user. The MSBR model simulate an axial mesh 55×55 with same dimensions of reactor cells. The code calculates the average of neutron flux to each square cell of this mesh. The flux estimation does not match the actual neutron source of the reactor. Thus, it is necessary to normalize the flux values initially calculated by MCNPX. In the simulations, this normalization was performed using the following equation [11]:

$$\phi_N = \phi_{MCNPX} \cdot \left[\frac{P \cdot \nu}{Q \cdot k_{eff}} \right] \quad (1)$$

where ϕ_N is the normalized flux; ϕ_{MCNPX} is the flux estimated by MCNPX; P is the reactor power level; ν is the average number of fission neutrons and Q is the recoverable energy per fission event. The values of ν , Q and k_{eff} are calculated by the MCNPX and the power level is designed by MSBR project at $P = 2250$ MWt [11].

3. RESULTS AND DISCUSSION

Table 5 present the effective multiplication factor (k_{eff}) of MSBR system among several works. The k_{eff} was calculated at beginning of cycle (BOC) to traditional RU considering 1.76% of fissile content. The k_{eff} calculated by de MCNPX 2.6.0, in the present study, agree with others works. The biggest difference is related to SERPENT code. According to reference [8], this discrepancy may be due the simplifications in Zone II performed in SERPENT model.

Table 5: MSBR criticality at BOC to RU fuel.

Evaluated Case	Present study	Reference [8]	Reference [8]	Reference [10]
Used code	MCNPX 2.6.0	MCNP6	SERPENT 2	MCNP6
k_{eff}	1.01010	1.00736	1.00389	1.01277
Diff. (pcm)	—	274	621	267

Table 6 presents the k_{eff} of the MSBR core for the evaluated fuels. The RU presents the highest k_{eff} values and the RA has the smallest one. The same mass percentage (mp) of fissile isotopes does not produce similar k_{eff} values, because the RP and RA fuels have nuclides which present high neutrons cross section for radiative capture. Furthermore, the atomic masses of the fissile isotopes are different. For the same mass and the same mp , the fuel that contains fissile isotopes with lightest atomic mass has more fissile atoms. In this way, among the evaluated fuels, the fissions number is the highest for the RU, which produces the highest k_{eff} value. Because these factors, the concentration of fissile isotopes in EU, RP and RA, must be higher than 2.26% in fuel mass. However, in the uranium mass, the EU fuel has 91% of ^{235}U (Table 1). This fact is negative for the non-proliferation issue because this high enrichment value is forbidden by international treaties. Thus, the present study forsakes the use ^{235}U and focuses on the analysis of RP and RA as initial fissile seed.

Table 6: Effective multiplication factor (k_{eff}) for the evaluated fuels.

Fissile isotopes in fuel matrix (mp)	Fuel Type (k_{eff})			
	RU	EU	RP	RA
1.76	1.01010	0.89823	0.88793	0.82751
2.26	1.12413	0.99576	0.90546	0.83596

The respective fissile content of Pu and Pu/AMS is 62 and 54% (Table 2). Although it is a high fissile content value, the radiotoxicity of the spent fuel hinders its proliferation. Moreover, the Pu and Pu/AMs are diluted in thorium mass to form the RP and RA fuels, where the percentage of fissile isotopes in fuel matrix is less than 54%. Table 7 present the k_{eff} to RP and RA fuels as

function of fissile content in the fuel matrix and as function of mass percentage of heavy metal. As expected, the gradual increase of fissile isotopes concentration produces higher k_{eff} values (Table 7). When RP and RA have 16% and 25% of fissile isotopes the k_{eff} is similar to RU fuel with 1.76% (see Table 6). Note that the fissile content $^{233}\text{U}/\text{U}$ in RU is 92% (Table 1) while the fissile content $(^{239}\text{Pu}+^{241}\text{Pu})/\text{Pu}$ and $(^{239}\text{Pu}+^{241}\text{Pu})/(\text{Pu}+\text{AMs})$ in reprocessed fuels is 62 and 54%, respectively (Table 2). Regarding the heavy metal mass, the RU uses 92% of ^{232}Th while the RP and RA use about 74 and 53% respectively (Table 6).

Table 7: Effective multiplication factor (k_{eff}) as a function of fissile isotopes content (mp) and heavy metal concentration (mp).

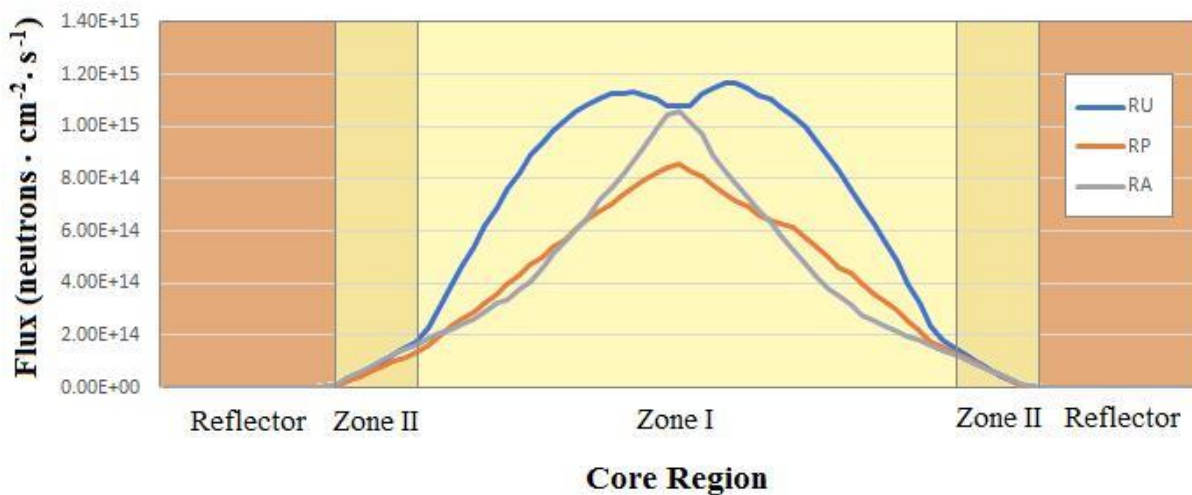
Fissile isotopes in fuel matrix (mp)	Heavy Metal (mp)			Fuel Type (k_{eff})	
	Th	Pu	Pu/AMs	RP	RA
10.0	83.9	16.1	—	0.94234	0.82546
15.0	75.9	24.1	—	0.99776	—
16.0	74.3	25.7	—	1.01143	—
17.0	72.6	27.4	—	1.02497	—
18.0	71.0	29.0	—	1.03864	—
19.0	69.4	30.6	—	1.05241	—
20.0	62.7	—	37.3	—	0.94310
21.0	60.8	—	39.2	—	0.95648
22.0	59.0	—	41.0	—	0.97081
23.0	57.1	—	42.9	—	0.98452
24.0	55.2	—	44.8	—	0.99907
25.0	53.4	—	46.6	—	1.01359
26.0	51.5	—	48.5	—	1.02701
27.0	49.6	—	50.4	—	1.04278
28.0	47.8	—	52.2	—	1.05600

Figure 2 illustrates the radial neutron flux profile in reactor core, where the highest relative error estimated by the code is about 10%. For evaluated fuels, Zone I presents the highest flux and the

reflector region the smallest one. In fact, the neutron flux in reflector is almost zero. Only in the inner annulus of this region there is a small neutron flux. This fact confirms that there is not neutron leakage from the reactor core. On the other hand, the central region in Zone I present the highest flux. The center core has a depletion of this flux which may have been produced by the absence of control rods. As related, the withdrawal of the graphite control rod inserts negative reactivity to the core due to the decrease of neutron moderation.

Comparing with the standard fuel (RU), the reprocessed fuels (RP and RA) present a reduction in the neutron flux. Although the RP and RA have concentration of fissile isotopes higher than RU, the reprocessed fuels have neutron absorbers that provoke a flux reduction. Between the reprocessed fuels RP present a neutron flux profile more flatten than RA. The presence of MAs in the RA may produce this behavior.

Figure 2: Total Neutron flux in MSBR core.



4. CONCLUSION

The criticality of the simulated MSBR, using RU fuel at BOC, is similar to previous studies. Among the evaluated fuels RP and RA seems to be promisor to initial fissile supply. Although the reprocessed Pu and Pu/MAs have about of 62 and 54% of fissile isotopes, the radiotoxicity of this spent fuel makes it difficult for proliferation. Diluting these isotopes in thorium mass, 16 and 25%

of the fissile isotope in fuel matrix of RP and RA, produce similar criticality of traditional RU. On the other hand, the presence of neutron absorbers in RP and RA reduce the radial neutron flux and produce a central peak in its profile. Studies to evaluate a new distribution of fuel elements are important to flatten the neutron flux.

Regarding to EU fuel, the used uranium needs to be highly enriched. The enrichment value estimated by this work is impractical due the limits of enrichment values established by the nonproliferation treaty. New studies need be performed to evaluate a uranium enrichment values that does not exceeds 20%. In these studies, it is necessary calculates the U/Th ratio that produces similar core criticality to traditional RU.

The use of plutonium and minor actinides as initial fissile source in MSBR, could contribute to reduction to waste production. However, it is essential evaluate the core behavior during the burnup, the spent fuel composition and the breeding ratio value among the evaluated fuels. Future works will simulate MSBR system to study these technical features where other nuclear codes can be used to compare the results.

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