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EVALUATION OF FUEL OPTIONS FOR THE FUJI-U3-(0) REACTOR

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ABSTRACT

In this study, the core physics of the small modular molten salt reactor type FUJI-U3-(0) was simulated using the SERPENT code. The thermal reactor cylindrical core was divided into three concentric regions, each with different radii for the fuel channels located at the center of graphite moderator hexagonal rods. Four different molten salt compositions were simulated by varying the fissile and fertile materials and their respective quantities. After achieving reactor criticality, the conversion ratio and waste generated by each salt composition were analyzed. The results include a breeder reactor using U-233 as fuel and a reactor with the potential to become a breeder, as it has a higher conversion ratio than usual. This second reactor uses plutonium recovered from PWR spent fuel, such as that from ANGRA-I, creating a burner reactor that recycles nuclear waste. Both reactors generate less waste compared to when enriched uranium is used as fuel.

1. INTRODUCTION

The increase in global population [1] and improving living conditions are driving a substantial rise in energy demand [2]. Current energy production methods exacerbate environmental issues such as pollution and global warming [3], leading to more frequent environmental disasters and food cultivation challenges [4]. Clean energy sources have been developed and nuclear energy remains crucial. However, the finite supply of uranium and price variability highlight the need for alternative fuels to extend the lifespan of nuclear fission energy. Thorium, a potential alternative, has been studied extensively for use in molten salt reactors.

The 2002 Generation IV roadmap identifies the Molten Salt Reactor (MSR) as a promising technology for future research [5]. The feasibility of MSRs was demonstrated in the 1950s with the Aircraft Reactor Experiment and further validated in the 1960s with the Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) [6]. Since the 1980s, the MSR-FUJI, developed by the International Thorium Molten-Salt Forum (ITMSF), has built on ORNL's findings. This reactor can use thorium as fertile material and uranium or plutonium as fissile material, potentially functioning as a breeder [7].

Brazil may have thorium as it’s largest energy reserve [8][9], and holds the second-largest thorium reserves globally [10]. The Thorium Group, first group dedicated to the study and development of nuclear energy in Brazil, has collaborated with France and Germany in advancing thorium-based nuclear energy technology [11].

Molten salt reactors use a liquid fuel mixture combined with the coolant, which circulates through the reactor core. The unique properties of molten salts allow the reactor to operate at high temperatures and low pressures safely. This mixture remains in liquid form at low pressures, preventing boiling under normal conditions [12]. Advantages of molten salt reactors include higher efficiency due to higher operational temperatures, reduced accident risks from lower pressure operations, versatility in fuel cycles, and the ability to act as nuclear waste "burners" or breeders [13].

The FUJI-U3-(0) is a thermal reactor using graphite as a moderator [7] and is also a Small Modular Reactor (SMR). SMRs are compact and produce lower power compared to traditional reactors, with benefits including suitability for sites unsuitable for larger plants, reduced construction costs, and modular design allowing incremental deployment to meet energy demands [14].

2. METHODOLOGY

The reactor FUJI-U3-(0) was simulated using the SERPENT code, and key parameters such as criticality, conversion ratio, and nuclear waste generation were evaluated for different fissile materials in the fuel. In the simulation, the salt composition remained constant at 68.9% LiF and 14.99% BeF_2, while the remaining percentage was adjusted between ThF_4 and UF_4/PuF_4, depending on the specific fuel being analyzed.

The reactor vessel of FUJI-U3-(0) has a cylindrical shape (Fig. 1.), and the core structure consists of hexagonal graphite moderator blocks, with pitch p = 9.5 cm (Fig. 2.) and height h = 466 cm. These blocks contain cylindrical concentrical fuel channels, with variable radii r, as shown in Fig. 1., through which the molten fuel salt flows, driven upward by the primary pump [7].

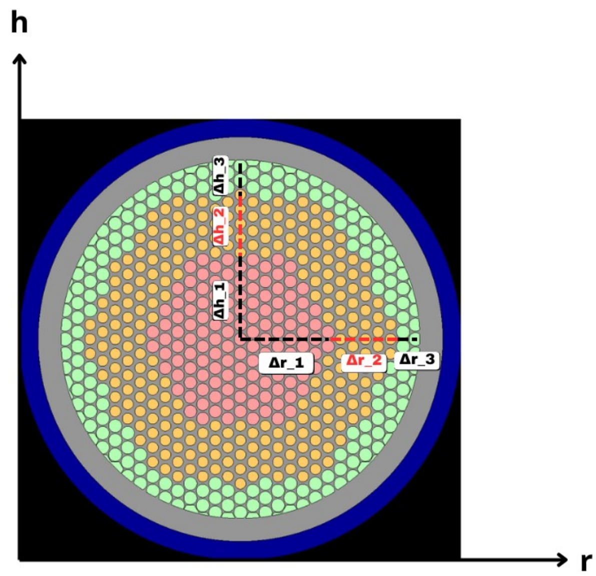

Fig. 1. Core regions.

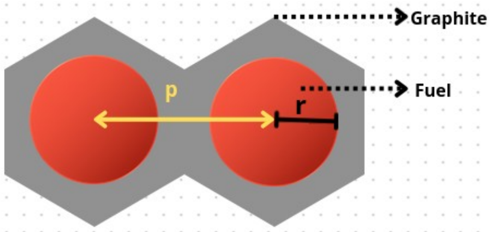

Fig. 2. FUJI-U3-(0) graphite rod.

The reactor core was divided into three regions, as can be seen in Fig. 1. with the dimentions of Tab. 1. Each region has different fuel channel radii, r_f, to optimize neutron distribution and flux uniformity, following the approach outlined in [15].

Tab. 1. Fuel regions dimensions

	Central region (red)	Intermediate region (orange)	Outer region (green)
Δr	1.16 m	0.8 m	0.4 m
Δh	1.23 m	0.7 m	0.4 m
r_f	8.28 cm	7.18 cm	8.70 cm

The reactor performance was evaluated using the following fuel compositions:

1. **ThF_4 14.45% + BeF_2 14.99% + LiF 69.98% + (U-233) UF_4 0.58%**: Thorium as the fertile material and U-233 as the fissile material.
2. **ThF_4 4.5% + BeF_2 14.99% + LiF 69.98% + (80% U-238 + 20% U-235) UF_4 10.53%**: Thorium as the fertile material and uranium enriched to 20% U-235 as the fissile material.
3. **BeF_2 14.99% + LiF 69.98% + (90% U-238 + 10% U-235) UF_4 15.3%**: Uranium enriched with no thorium.
4. **ThF_4 12.25% + BeF_2 14.99% + LiF 69.98% + (2.1% Pu-238 + 52.6% Pu-239 + 24.2% Pu-240 + 14.8% Pu-241 + 6.4% Pu-242) PuF_4 2.78%**: Thorium as the fertile material with a mixture of plutonium isotopes typical of spent fuel from a Pressurized Water Reactor (PWR).

U-233 was selected as the fissile material of the first composition based on the reactor's technical specifications [7] and the prevalence of its use in the majority of relevant studies, including [15]. Although the use of U-233 is not widespread, it has been produced and utilized as fissile fuel since the Molten Salt Reactor Experiment (MSRE) in the 1960s, demonstrating its proven feasibility.

3. RESULTS

The simulation used a population of 50,000 neutrons with 500 active cycles in Serpent Version 2.1.30 (February 14, 2018) that uses ENDF/B-VII cross-section library. Although the population is not very large, the uncertainty of the eigenvalues is, on avarage, 40 pcm, and the results are consistent with the reference [15]. Since we are evaluating only integral parameters and not differential ones, the statistical error is smaller, allowing a reduced number of histories to yield accurate results. The burnup period was set to 140 days at full power, with reflective boundary conditions applied. The choice of 140 days was based on the fact that these reactors have online refueling to maintain criticality and fissile material production. Since neutron absorption without fission is significant for fuel production, the multiplication factor decreases rapidly as the chain reaction cannot remain stable for long periods. Consequently, the multiplication factor is higher at the beginning, accounting for the high absorption rate and the need for refueling to maintain reactor criticality. Extending the refueling interval is crucial due to the amount of fuel consumed.

All compositions were calculated to ensure that the reactor had enough fissile material to remain critical during the burnup period of 140 days, with k_{eff} values close to 1 as can be seen in Fig. 3., allowing for an analysis of conversion ratios and nuclear waste generation. This was achieved by keeping the percentages of LiF and BeF_2 in the salt fixed while varying the amount and enrichment of ThF_4 and UF_4, until the fuel maintained a multiplication factor greater than one throughout the 140-day burnup period.

The graph illustrating the conversion ratio for all fuel configurations over the burn time has been plotted together and is presented in Fig. 4. As anticipated based on the conclusions from [15], the first composition results in a breeder reactor. The conversion ratio varied from 1.0075(±0.003) to 1.0206(±0.003) during the burning period.

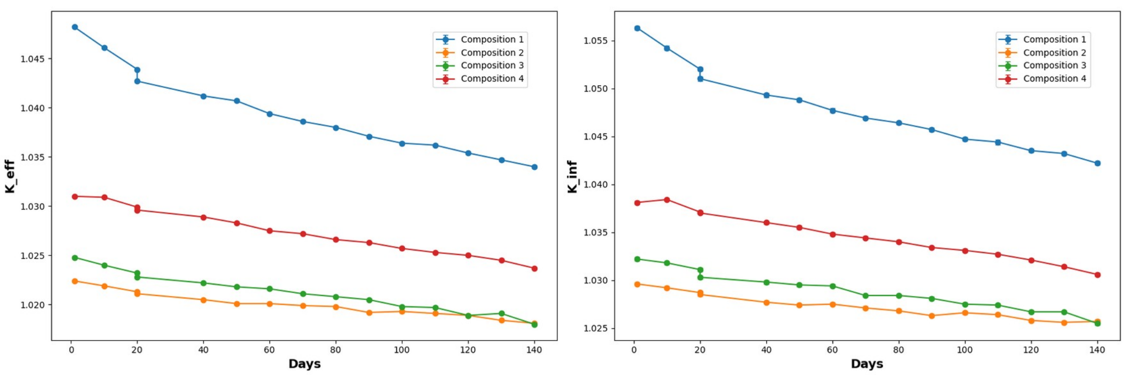

Fig. 3. k_{eff} and k_{inf} during burning period for different salt compositions.

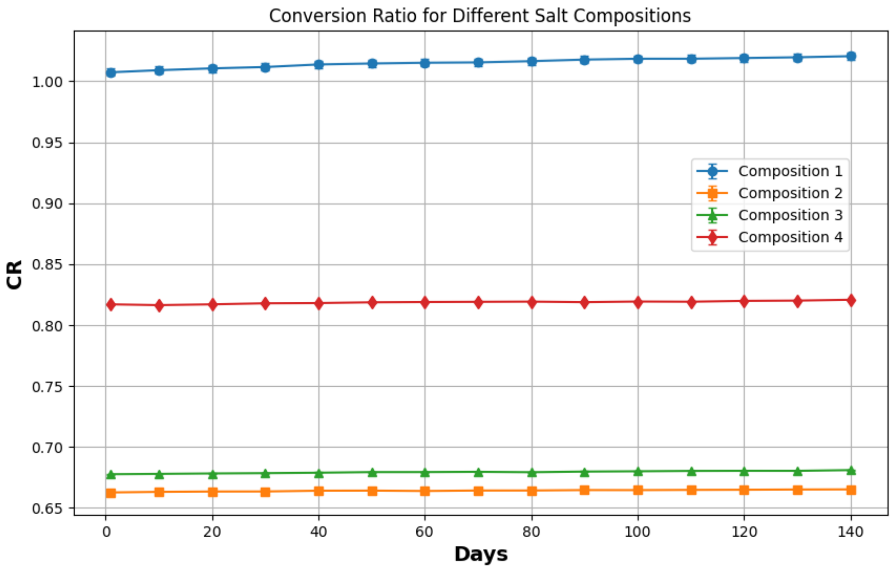

Fig. 4. Conversion Ratio for the different salt compositions during burning period.

Despite both compositions containing U-238, a fertile nuclide, the second and third compositions exhibited the lowest conversion ratios. The second composition had conversion ratios ranging from 0.66274 (±0.003) to 0.66524 (±0.0029), while the third composition ranged from 0.6777 (±0.0029) to 0.68106 (±0.0031).

The fourth composition, which allows a higher percentage of ThF_4 to achieve criticality, exhibited a favorable conversion ratio, ranging from 0.81639 (±0.003) to 0.82078 (±0.0031). This result suggests that using plutonium derived from the spent fuel of a PWR as the fissile material in the FUJI-U3-(0) reactor is more promising for achieving breeder reactor capabilities compared to any combination of enriched U-238 and U-235.

Another important aspect to analyze is the total mass of certain nuclides produced at the end of the 140-day burnup period. For each different composition, we will evaluate the total mass of fissile nuclides and waste products, as depicted in Fig. 5.

Let's begin by analyzing the waste products excluding uranium and plutonium, specifically Tc-99, Sn-126, Se-79, Cs-135, Zr-93, Pd-107, I-129, Np-237, and Am-241, which are alpha and beta emitters. We can observe that the total mass of these isotopes does not vary significantly among compositions 1, 2, and 3, although it is slightly higher in compositions 2 and 3. However, composition 4 shows a substantially higher total mass, primarily due to the amounts of Am-241 and Pd-107 formed in the core.

Am-241 is higher in composition 4 due to Pu-239 decay, while Pd-107 also contributes to its increased mass. Composition 1, with less plutonium, produces the least Am-241. Neptunium is mostly found in compositions 2 and 3, with much lower levels in compositions 1 and 4. I-129 is produced more in compositions 1 and 4, while Zr-93 production is similar in compositions 1-3 but lower in composition 4. Cs-135 production is consistent across all compositions, slightly lower in composition 1. Se-79 and Sn-126 vary more, with Se-79 three times higher in composition 1, and Sn-126 higher in compositions 1 and 4. Tc-99 is produced consistently across all compositions.

Nuclide	composition 1	composition 2	composition 3	composition 4
	mass (g) EOF	mass (g) EOF	mass (g) EOF	mass (g) EOF
U-232	**1.9054**	**0.12368**	**2.44E-05**	**0.37824**
U-233	**3.48E+06**	**17239**	**0.0047797**	**40623**
U-234	**10415**	**58.441**	**6.7528**	**1182.8**
U-235	**73.153**	**1.27E+07**	**9.05E+06**	**100.85**
U-236	**0.11321**	**26045**	**25346**	**165.58**
U-238	**1.27E-07**	**5.13E+07**	**8.21E+07**	**0.75402**
Pu-238	**4.93E-07**	**0.25465**	**0.54509**	**3.58E+05**
Pu-239	**7.39E-10**	**35266**	**59130**	**8.98E+06**
Pu-240	**2.00E-12**	**99.923**	**266.99**	**4.17E+06**
Pu-241	**2.27E-14**	**1.4528**	**6.3006**	**2.49E+06**
Pu-242	**2.67E-17**	**0.0011864**	**0.0080281**	**1.08E+06**
Tc-99	**1317.6**	**1623.1**	**1625**	**1566.5**
Sn-126	**80.758**	**23.71**	**24.085**	**59.805**
Se-79	**31.761**	**10.445**	**10.226**	**7.8791**
Cs-135	**2218.1**	**2417.4**	**2395.1**	**2681.6**
Zr-93	**1819.8**	**1622.4**	**1614**	**888.13**
Pd-107	**35.511**	**57.394**	**65.382**	**1095.2**
I-129	**572.67**	**208.87**	**210.69**	**416.21**
Np-237	**0.00016881**	**78.009**	**115.83**	**15.494**
Am-241	**4.60E-17**	**0.0068103**	**0.029414**	**46556**
Sum of the wastes excluding Uranium and Plutonium				
	composition 1	**composition 2**	**composition 3**	**composition 4**
	6076.2001688	**6041.3348103**	**6060.342414**	**53286.8181**
Sum of uranium: U-232, U-234 and U-236				
	composition 1	**composition 2**	**composition 3**	**composition 4**
	10417.01861	**26103.56468**	**25352.752824**	**1348.75824**
Sum of plutonium excluding composition 4				
	composition 1	**composition 2**	**composition 3**	
	4.933113E-07	**35367.631636**	**59403.843718**	
Total sum of the nuclides (excluding plutonium from comp. 4).				
	composition 1	**composition 2**	**composition 3**	**composition 4**
	16493.218779	**67512.531127**	**90816.938956**	**54635.57634**

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Fig. 5. Total mass of notable nuclides at the end of life (EOL)

Now, let's analyze the production of U-232, U-234, U-236, and U-238. U-232, an alpha emitter with a short half-life, is predominantly produced in composition 1, with minimal production in composition 3. U-234, also an alpha emitter but with a much longer half-life, is significant in composition 1 due to neutron capture by U-233, and slightly less in composition 4, where U-233 is bred from thorium. U-236, formed by neutron absorption in U-235, is produced in low quantities in composition 1, more in composition 4, and much higher in compositions 2 and 3. U-238 is produced in very small amounts in compositions 1 and 4, but much more in compositions 2 and 3, which use enriched uranium.

To analyze plutonium isotopes, composition 4 is excluded since it uses plutonium as fuel, and after 140 days of burnup, there is still a significant amount of the initial plutonium remaining. In compositions 2 and 3, plutonium production is considerably higher than in composition 1. Since plutonium is a problematic waste product, reactors producing less of it or using it as fuel are advantageous.

Finally, when comparing the total waste products analyzed, compositions 2 and 3 produce more than compositions 1 and 4. The production of U-233 is highest in composition 4 due to its higher conversion ratio, followed by composition 1, which also breeds U-233, making it a breeder reactor. U-235 is produced in small quantities in compositions 1 and 4, but remains abundant in compositions 2 and 3, where it is the primary fissile fuel after 140 days of burnup. At thermal neutron energies, thorium fuel presents a good conversion/breeding ratio. Because Th-232 has a larger absorption cross section than U-238, high conversion from Th-232 to U-233 can be reached at thermal neutron energies.

CONCLUSION

Nuclear waste presents a major challenge to the viability of nuclear energy, highlighting the need to reduce its hazard and explore its potential for generating new fissile material. The FUJI-U3-(0) reactor tackles these issues by minimizing waste production and utilizing it to create additional fissile material. Breeder reactors are vital as they generate more fuel while producing energy, which is crucial for sustainable energy generation amidst global warming, climate change and extension of lifespan of nuclear fission energy. The FUJI-U3-(0) reactor, using U-233 and thorium, exemplifies this importance.

Among the fuels compared, U-235 and U-238 are less effective, producing the least fissile material and the most analyzed waste. U-233 with thorium performs best and achieves breeder status. The use of U-233 is feasible when produced in another reactor, something already done in history. However, using plutonium, waste from another reactor, is even more interesting since it is inevitably produced and represents recycling. Although the plutonium-salt reactor did not achieve breeder status, it produces more fissile material than compositions 2 and 3, indicating that it is closer to becoming a breeder. A potential solution involves the seed-blanket concept, where a plutonium-based salt is used as the seed and a thorium-only salt as the blanket, potentially leading to a breeder reactor that utilizes waste as fuel. Thorium and plutonium use extends the lifespan of fission energy by broadening fuel options. Besides that, a SMR MSR offers enhanced safety, advanced technologies, and adaptability to remote locations.

Given Brazil's substantial thorium reserves, prioritizing thorium reactors could lead to technological advancement, domestic fuel use, and export opportunities, benefiting both the environment and the economy. In conclusion, researching and utilizing thorium-based reactors holds promise for addressing the increasing demands of nuclear energy.

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