

Fuel depletion analysis for a single-fluid double-zone thorium molten salt reactor (SD-TMSR) using Serpent 2 code

Osama Ashraf^{a,b,*}, Andrei Rykhlevskii^c, Anton Smirnov^a, G. V. Tikhomirov^a, Kathryn D. Huff^c

^a*Laboratory of Engineering Computer Modeling in Nuclear Technologies, Institute of Nuclear Physics and Engineering, National Research Nuclear University MEPhI, Moscow, Russia, 115409*

^b*Physics Department, Faculty of Education, Ain Shams University, Cairo, Egypt, 11341*

^c*Dept. of Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801*

Abstract

SD-TMSR 2,250 MWth is a Single-fluid Double-zone Thorium-based Molten Salt Reactor. The active core of this reactor is divided into the inner zone (486 fuel tubes) and the outer zone (522 fuel tubes), in order to improve the Th-U breeding performance. The lack of computational codes which deal with MSR cores stands against developing such a concept. Here, Serpent 2 Monte Carlo code has been adopted to analyze the whole core model of the SD-TMSR with online reprocessing and refueling. In the steady state calculations, molten salt with composition: LiF-BeF₂-ThF₄-233UF₄ at 70-17.5-12.3-0.2 mole%, respectively is used, this leading to $K_{eff} = 1.00055 \pm 0.00089$. The breeding ratio (BR) calculated by the Serpent 2 code in the steady state is 1.11604 ± 0.00033 . The molten salt Temperature Coefficient of Reactivity (TCR) found to be negative and equal to -3.13 pcm/K. In addition, the variation of multiplication factor, BR and build-up of the important nuclides in the core as a function of burnup time have been investigated. Under online reprocessing and refueling, the multiplication factor is increased from 1.00055 ± 0.00089 to 1.10473 ± 0.00082 over 60 years. When the chemical reprocessing rate is 5 m³/d, the reactor can

*Corresponding Author

Email address: osama.ashraf@edu.asu.edu.eg (Kathryn D. Huff)

operate as an iso-breeding. In conclusion, the full core of SD-TMSR has been modeled and the net production of ^{233}U is found to be positive and linearly proportional to the burnup time. The net production of ^{233}U reached 4.25 tons at the end of the period.

Keywords: molten salt reactor, single-fluid double-zone thorium molten salt reactor, sd-tmsr, depletion, online reprocessing, nuclear fuel cycle, salt treatment

1. Introduction

2. Methods

2.1. SD-TMSR design and model description

The Molten Salt Breeder Reactor (MSBR) vessel has a diameter of 680 cm and a height of 610 cm. It contains a molten fluoride fuel-salt mixture that generates heat in the active core region and transports that heat to the primary heat exchanger by way of the primary salt pump. In the active core region, the fuel salt flows through channels in moderating and reflecting graphite blocks. Fuel salt at 565°C enters the central manifold at the bottom via four 40.64-cm-diameter nozzles and flows upward through channels in the lower plenum graphite. The fuel salt exits at the top at about 704°C through four equally spaced nozzles which connect to the salt-suction pipes leading to primary circulation pumps. The fuel salt drain lines connect to the bottom of the reactor vessel inlet manifold.

Figure 1 shows the configuration of the MSBR vessel, including the “fission” (zone I) and “breeding” (zone II) regions inside the vessel. The core has two radial zones bounded by a solid cylindrical graphite reflector and the vessel wall. The central zone, zone I, in which 13% of the volume is fuel salt and 87% graphite, is composed of 1,320 graphite cells, 2 graphite control rods, and 2 safety¹ rods. The under-moderated zone, zone II, with 37% fuel salt, and radial reflector, surrounds the zone I core region and serves to diminish neutron leakage.

¹ These rods needed for emergency shutdown only.

Zones I and II are surrounded radially and axially by fuel salt (figure 2). This space for fuel is necessary for injection and flow of molten salt.

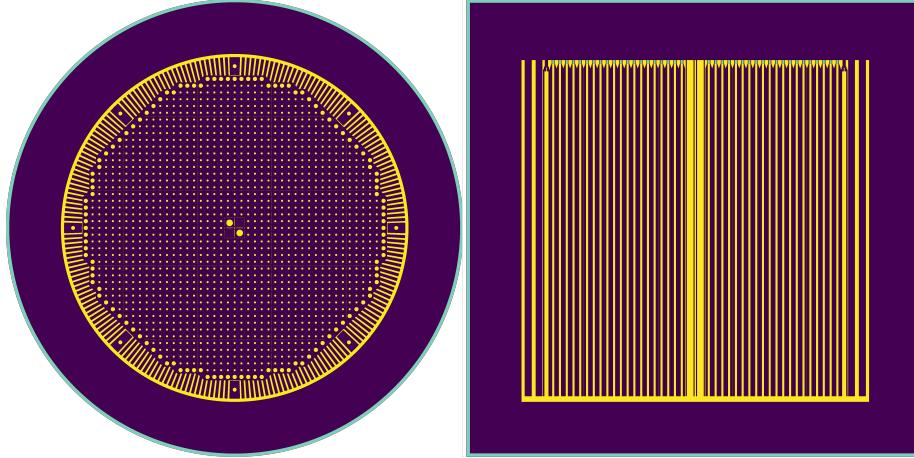


Figure 1: Plan and elevation views of SERPENT 2 MSBR model developed in this work.

Since reactor graphite experiences significant dimensional changes due to
25 neutron irradiation, the reactor core was designed for periodic replacement. Based
on the experimental irradiation data from the Molten Salt Reactor Experiment
(MSRE), the core graphite lifetime is about 4 years and the reflector graphite
lifetime is 30 years [1].

There are eight symmetric graphite slabs with a width of 15.24 cm in zone II,
30 one of which is illustrated in Figure 2. The holes in the centers are for the core
lifting rods used during the core replacement operations. These holes also allow
a portion of the fuel salt to flow to the top of the vessel for cooling the top head
and axial reflector. Figure 2 also shows the 5.08-cm-wide annular space between
35 the removable core graphite in zone II-B and the permanently mounted reflector
graphite. This annulus consists entirely of fuel salt, provides space for moving
the core assembly, helps compensate for the elliptical dimensions of the reactor
vessel, and serves to reduce the damaging flux at the surface of the graphite
reflector blocks.

^{135}Xe is a strong neutron poison, and some fraction of this gas is absorbed

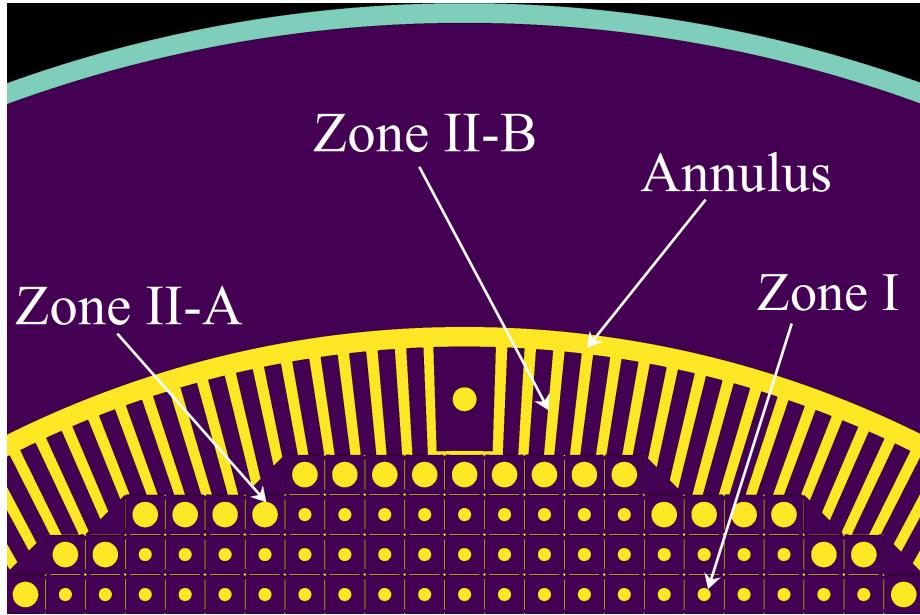


Figure 2: Detailed view of MSBR two zone model. Yellow represents fuel salt, purple represents graphite, and aqua represents the reactor vessel.

40 by graphite during MSBR operation. ORNL calculations show that for unsealed commercial graphite with helium permeability 10^{-5} cm²/s the calculated poison fraction is less than 2% [1]. This parameter can be improved by using experimental graphites or by applying sealing technology. The effect of the gradual poisoning of the core graphite with xenon is not treated here.

45 *2.1.1. Core zone I*

The central region of the core, called zone I, is made up of graphite elements, each 10.16cm×10.16cm×396.24cm. Zone I has 4 channels for control rods: two for graphite rods which both regulate and shim during normal operation, and two for backup safety rods consisting of boron carbide clad to assure sufficient negative reactivity for emergency situations.

50 These graphite elements have a mostly rectangular shape with lengthwise ridges at each corner that leave space for salt flow elements. Various element sizes reduce the peak damage flux and power density in the center of the core to

prevent local graphite damage. Figure 3 shows the elevation and plan views of
 55 graphite elements of zone I [1] and their SERPENT model [?].

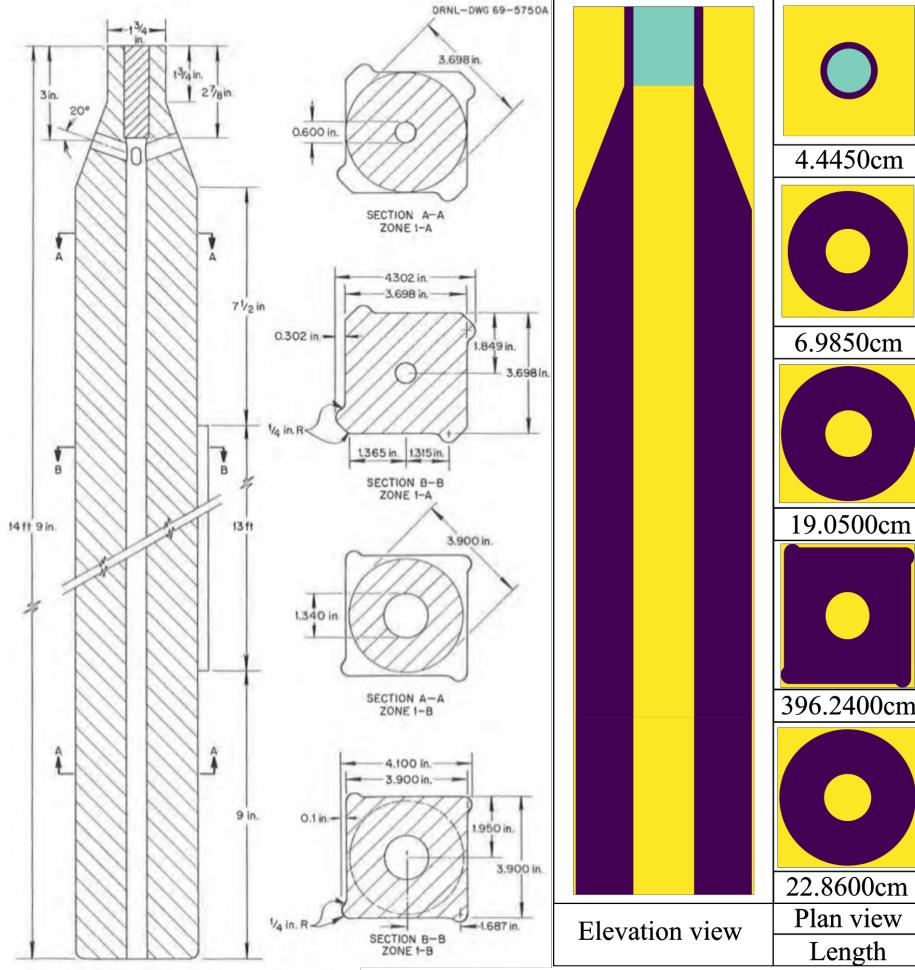


Figure 3: Graphite moderator elements for zone I [1?]. Yellow represents fuel salt, purple represents graphite, and aqua represents the reactor vessel.

2.1.2. Core zone II

Zone II, which is undermoderated, surrounds zone I. Combined with the bounding radial reflector, zone II serves to diminish neutron leakage. Two kinds of elements form this zone: large-diameter fuel channels (zone II-A) and radial

60 graphite slats (zone II-B).

Zone II has 37% fuel salt by volume and each element has a fuel channel diameter of 6.604cm. The graphite elements for zone II-A are prismatic with elliptical dowels running axially between the prisms. These dowels isolate the fuel salt flow in zone I from that in zone II. Figure 4 shows the shapes and dimensions of these graphite elements and their SERPENT model. Zone II-B elements are rectangular slats spaced far enough apart to provide the 0.37 fuel salt volume fraction. The reactor zone II-B graphite 5.08cm-thick slats vary in the radial dimension (average width is 26.67cm) as shown in figure 2. Zone II serves as a blanket to achieve the best performance: a high breeding ratio and a low fissile inventory. The harder neutron energy spectrum in zone II enhances the rate of thorium resonance capture relative to the fission rate, thus limiting the neutron flux in the outer core zone and reducing the neutron leakage [1].

The sophisticated, irregular shapes of the fuel elements challenge an accurate representation of zone II-B. The suggested design [1] of zone II-B has 8 irregularly-shaped graphite elements as well as dozens of salt channels. These graphite elements were simplified into right-circular cylindrical shapes with central channels. Figure 2 illustrates this core region in the SERPENT model. The volume of fuel salt in zone II was kept exactly at 37%, so that this simplification did not considerably change the core neutronics. Simplifying the eight edge channels was the only simplification made to the MSBR geometry in this work.

2.1.3. Material composition and normalization parameters

The fuel salt, reactor graphite, and modified Hastelloy-N are all materials created at Oak Ridge National Laboratory (ORNL) specifically for the MSBR. The initial fuel salt used the same density (3.35 g/cm^3) and composition $\text{LiF-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4$ (71.75-16-12-0.25 mole %) as the MSBR design [1]. The lithium in the molten salt fuel is fully enriched to 100% ${}^7\text{Li}$ because ${}^6\text{Li}$ is a very strong neutron poison and becomes tritium upon neutron capture.

The JEFF-3.1.2 neutron library provided cross section generation [?]. The specific temperature was fixed for each material and did not change during the

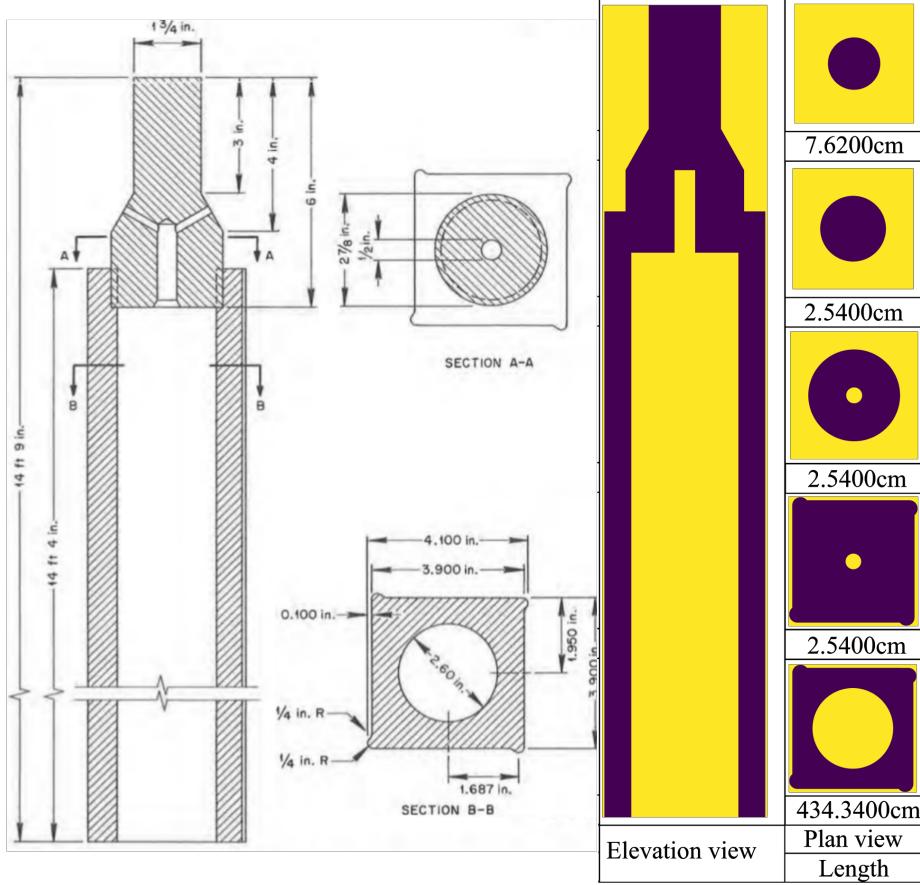


Figure 4: Graphite moderator elements for zone II-A [1?]. Yellow represents fuel salt and purple represents graphite.

reactor operation. The isotopic composition of each material at the initial state was described in detail in the MSBR conceptual design study [1] and has been applied to the SERPENT model without any modification. Table 1 is a summary of the major MSBR parameters used by this model [1].

2.2. Online reprocessing method

Removing specific chemical elements from a molten salt requires intelligent design (e.g., chemical separations equipment design, fuel salt flows to equipment) and has a considerable economic cost. All liquid-fueled Molten Salt Reactor

Table 1: Summary of principal data for MSBR [1].

Thermal capacity of reactor	2250 MW(t)
Net electrical output	1000 MW(e)
Net thermal efficiency	44.4%
Salt volume fraction in central zone I	0.13
Salt volume fraction in outer zone II	0.37
Fuel salt inventory (Zone I)	8.2 m ³
Fuel salt inventory (Zone II)	10.8 m ³
Fuel salt inventory (annulus)	3.8 m ³
Total fuel salt inventory	48.7 m ³
Fissile mass in fuel salt	1303.7 kg
Fuel salt components	LiF-BeF ₂ -ThF ₄ - ²³³ UF ₄
Fuel salt composition	71.75-16-12-0.25 mole%
Fuel salt density	3.35 g/cm ³

(MSR) designs involve varying levels of online fuel processing. Minimally, volatile gaseous fission products (e.g. Kr, Xe) escape from the fuel salt during routine reactor operation and must be captured. Additional systems might be used to enhance removal of those elements. Most designs also call for the removal of noble and rare earth metals from the core since these metals act as neutron poisons. Some designs suggest a more complex list of elements to process (figure 5), including the temporary removal of protactinium or other regulation of the actinide inventory [?].

2.2.1. Fuel material flows

The ²³²Th in the fuel absorbs thermal neutrons and produces ²³³Pa which then decays into the fissile ²³³U. Furthermore, the MSBR design requires online reprocessing to remove all poisons (e.g. ¹³⁵Xe), noble metals, and gases (e.g. ⁷⁵Se, ⁸⁵Kr) every 20 seconds. Protactinium presents a challenge, since it has a large absorption cross section in the thermal energy spectrum. Moreover, ²³³Pa left in the core would produce ²³⁴Pa and ²³⁴U, neither of which are useful as fuel.

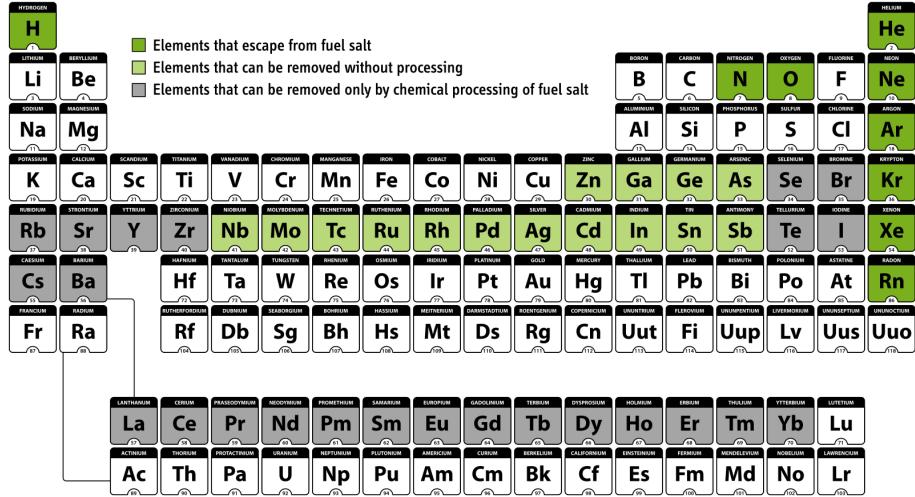


Figure 5: Processing options for MSR fuels. Reproduced from [?] where it was adapted from a chart courtesy of Nicolas Raymond, www.freestock.ca.

Accordingly, ^{233}Pa is continuously removed from the fuel salt into a protactinium decay tank to allow ^{233}Pa to decay to ^{233}U without the corresponding negative neutron impact. The reactor reprocessing system must separate ^{233}Pa from the molten-salt fuel over 3 days, hold it while ^{233}Pa decays into ^{233}U , and return it back to the primary loop. This feature allows the reactor to avoid neutron losses to protactinium, lowers in-core fission product inventory, and increases the efficiency of ^{233}U breeding. Table 2 summarizes full list of nuclides and the “cycle times”² used for modeling salt treatment and separations [1].

The removal rates vary among nuclides in this reactor concept which dictate the necessary resolution of depletion calculations. If the depletion time intervals are very short, an enormous number of depletion steps are required to obtain the equilibrium composition. On the other hand, if the depletion calculation time interval is too long, the impact of short-lived fission products is not captured.

² The MSBR program defined a “cycle time” as the amount of time required to remove 100% of a target nuclide from a fuel salt [1].

Table 2: The effective cycle times for protactinium and fission products removal (reproduced from [1]).

Processing group	Nuclides	Cycle time (at full power)
Rare earths	Y, La, Ce, Pr, Nd, Pm, Sm, Gd	50 days
	Eu	500 days
Noble metals	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag,	20 sec
	Sb, Te	
Seminoble metals	Zr, Cd, In, Sn	200 days
Gases	Kr, Xe	20 sec
Volatile fluorides	Br, I	60 days
Discard	Rb, Sr, Cs, Ba	3435 days
Protactinium	^{233}Pa	3 days
Higher nuclides	^{237}Np , ^{242}Pu	16 years

To compromise, a 3 day time interval was selected for depletion calculations³ to correlate with the removal interval of ^{233}Pa and ^{232}Th was continuously added to maintain the initial mass fraction of ^{232}Th .

3. Results

¹³⁰ 3.1. Effective multiplication factor

Figures 6, 7 show the effective multiplication factors.

3.2. Fuel salt composition dynamics

The analysis of the fuel salt composition evolution provides more comprehensive information about the equilibrium state. Figure 8 shows the number densities of major nuclides which have a strong influence on the reactor core

³ Optimal depletion time step of 3 days for MSR batch-wise depletion simulation was first described and concluded by Powers *et al.* [?].

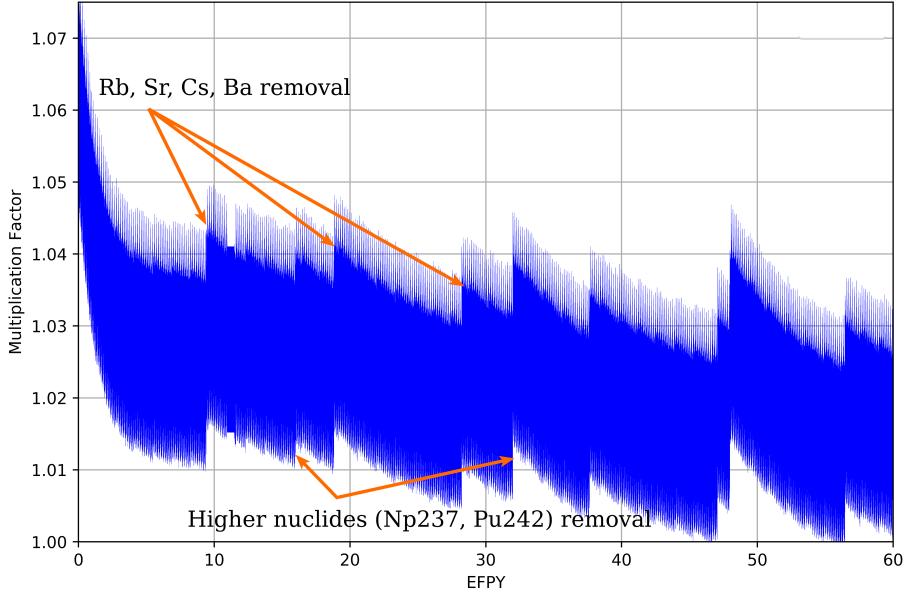


Figure 6: Effective multiplication factor dynamics for full-core MSBR model over a 60-year reactor operation lifetime.

physics. The concentration of ^{233}U , ^{232}Th , ^{233}Pa , and ^{232}Pa in the fuel salt change insignificantly after approximately 2500 days of operation. In particular, the ^{233}U number density fluctuates by less than 0.8% between 16 and 20 years of operation. Hence, a quasi-equilibrium state was achieved after 16 years of reactor operation.

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3.3. Neutron spectrum

Figure 9 shows the normalized neutron flux spectrum for the full-core MSBR model in the energy range from 10^{-8} to 10 MeV. The neutron energy spectrum at equilibrium is harder than at startup due to plutonium and other strong absorbers accumulating in the core during reactor operation.

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3.4. Neutron flux

Figure 11 shows the radial distribution of fast and thermal neutron flux for the both initial and equilibrium composition.

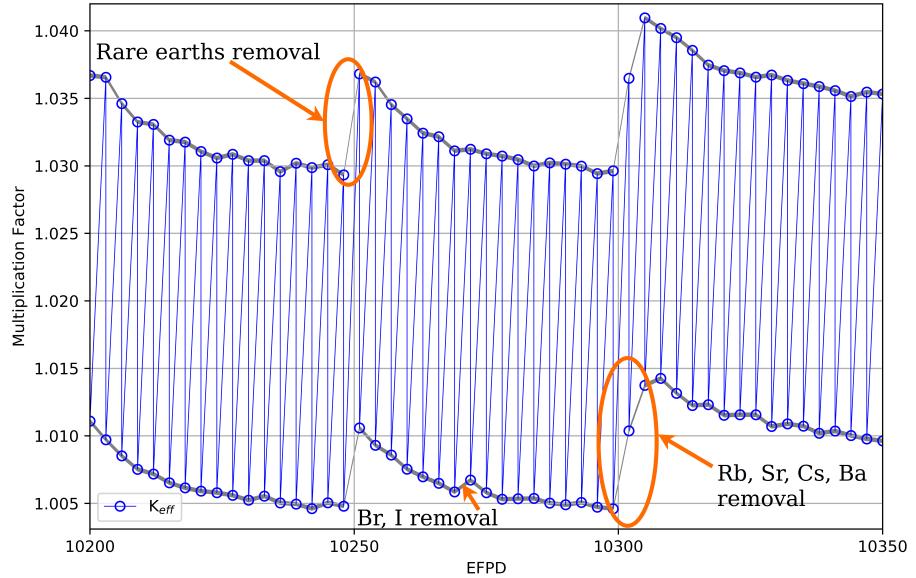


Figure 7: Zoomed effective multiplication factor for 150-EFPD time interval.

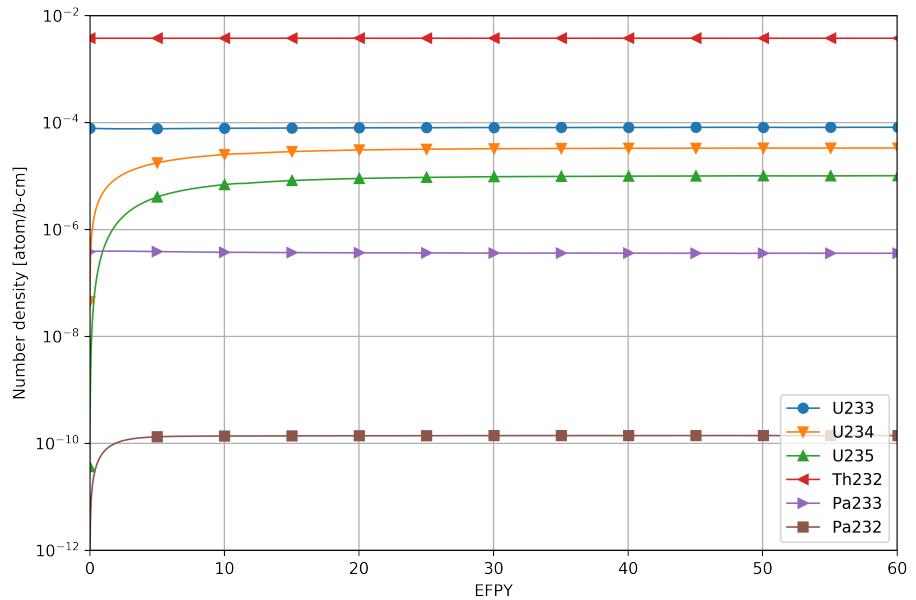


Figure 8: Number density of major nuclides during 60 years of reactor operation.

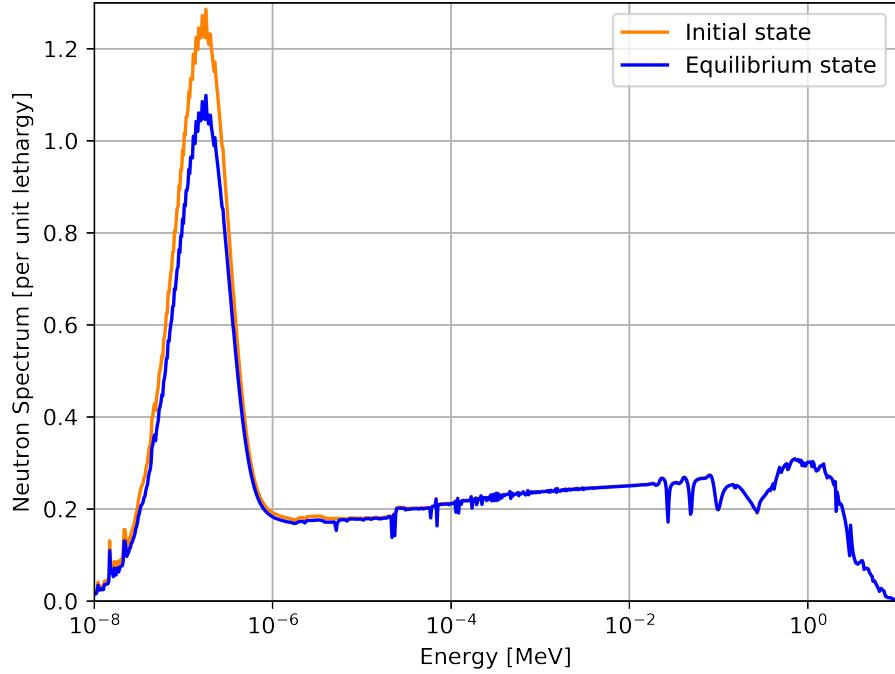


Figure 9: The neutron flux energy spectrum is normalized by unit lethargy and the area under the curve is normalized to 1 for initial and equilibrium fuel salt composition.

3.5. Power and breeding distribution

Table 3 shows the power fraction in each zone for initial and equilibrium fuel compositions. Figure 12 reflects the normalized power distribution of the MSBR quarter core for equilibrium fuel salt composition. For both the initial and equilibrium compositions, fission primarily occurs in the center of the core, namely zone I. The spectral shift during reactor operation results in slightly different power fractions at startup and equilibrium, but most of the power is still generated in zone I at equilibrium (table 3). Figure 13 shows the neutron capture reaction rate

3.6. Temperature coefficient of reactivity

Table 4 summarizes temperature effects on reactivity calculated in this work for both initial and equilibrium fuel compositions, compared with the original

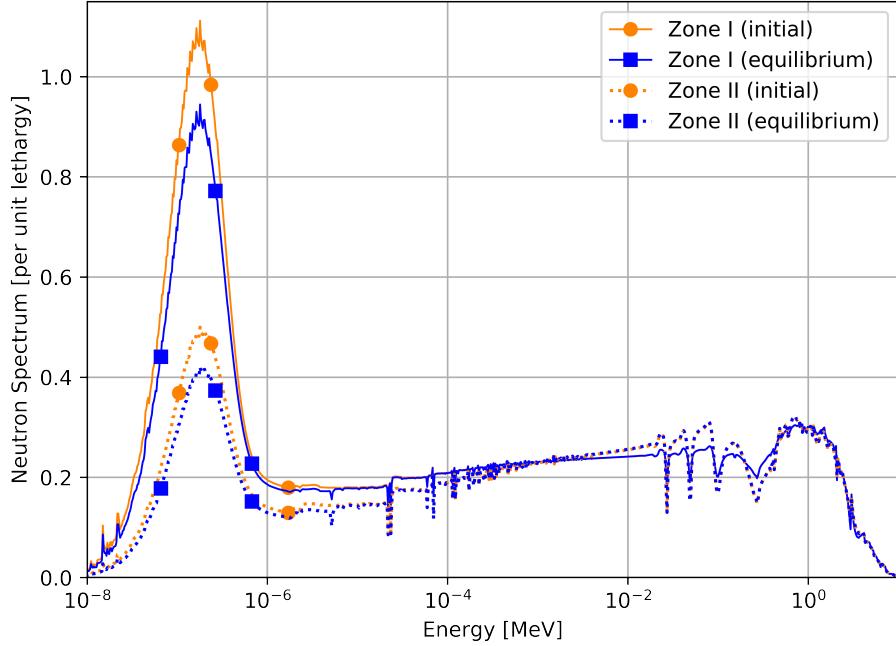


Figure 10: The neutron flux energy spectrum in different core regions is normalized by unit lethargy and the area under the curve is normalized to 1 for the initial and equilibrium fuel salt composition.

Table 3: Power generation fraction in each zone for initial and equilibrium state.

Core region	Initial	Equilibrium
Zone I	97.91%	98.12%
Zone II	2.09%	1.88%

ORNL report data [1]. By propagating the k_{eff} statistical error provided by SERPENT2, uncertainty for each temperature coefficient was obtained and appears in Table 4. Other sources of uncertainty are neglected, such as cross section measurement error and approximations inherent in the equations of state providing both the salt and graphite density dependence on temperature.
165 The main physical principle underlying the reactor temperature feedback is an expansion of heated material. When the fuel salt temperature increases, the density of the salt decreases, but at the same time, the total volume of fuel

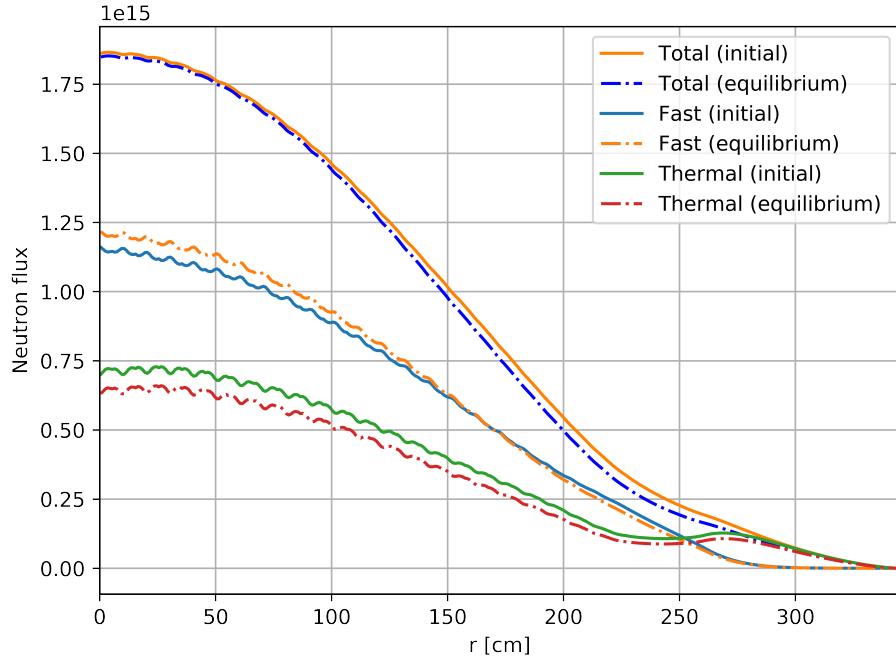


Figure 11: Radial neutron flux distribution for initial and equilibrium fuel salt composition.

salt in the core remains constant because it is bounded by the graphite. When
 170 the graphite temperature increases, the density of graphite decreases, creating additional space for fuel salt. To determine the temperature coefficients, the cross section temperatures for the fuel and moderator were changed from 900K to 1000K. Three different cases were considered:

1. Temperature of fuel salt rising from 900K to 1000K.
- 175 2. Temperature of graphite rising from 900K to 1000K.
3. Whole reactor temperature rising from 900K to 1000K.

In the first case, changes in the fuel temperature only impact fuel density. In this case, the geometry is unchanged because the fuel is a liquid. However,
 180 when the moderator heats up, both the density and the geometry change due to thermal expansion of the solid graphite blocks and reflector. Accordingly, the new graphite density was calculated using a linear temperature expansion

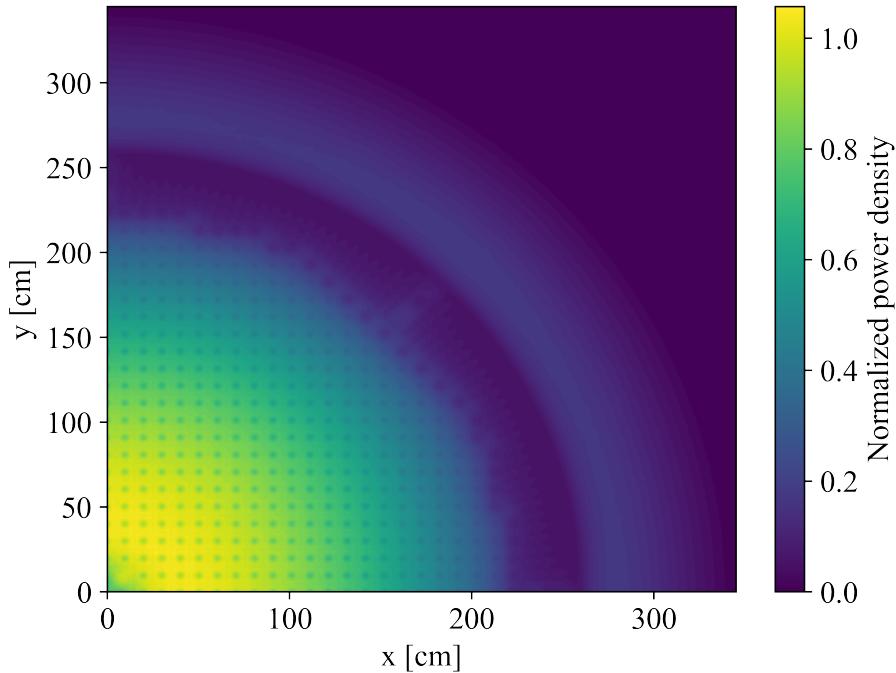


Figure 12: Normalized power density for equilibrium fuel salt composition.

coefficient of $1.3 \times 10^{-6} \text{K}^{-1}$ [1]. A new geometry input for SERPENT2, which takes into account displacement of graphite surfaces, was created based on this information. For calculation of displacement, it was assumed that the interface
 185 between the graphite reflector and vessel did not move, and that the vessel temperature did not change. This is the most reasonable assumption for the short-term reactivity effects because inlet salt is cooling graphite reflector and inner surface of the vessel.

The fuel temperature coefficient (FTC) is negative for both initial and equi-
 190 librium fuel compositions due to thermal Doppler broadening of the resonance capture cross sections in the thorium. A small positive effect of fuel density on reactivity increases from +1.21 pcm/K at reactor startup to +1.66 pcm/K for equilibrium fuel composition which has a negative effect on FTC magnitude during the reactor operation. This is in good agreement with earlier research

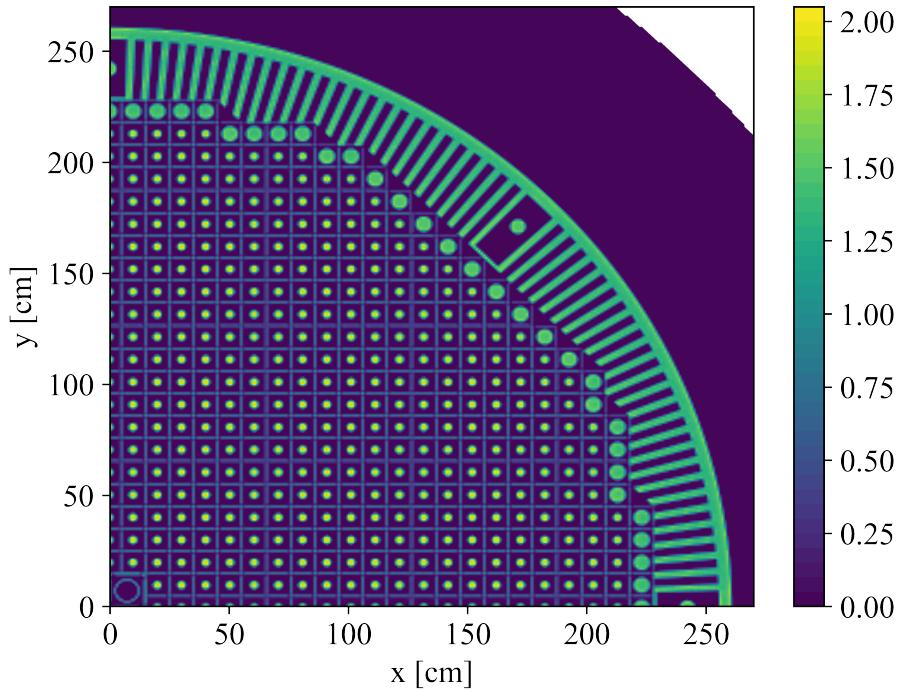


Figure 13: ^{232}Th neutron capture reaction rate normalized by total flux for equilibrium fuel salt composition.

[1?]. The moderator temperature coefficient (MTC) is positive for the startup composition and decreases during reactor operation because of spectrum hardening with fuel depletion. Finally, the total temperature coefficient of reactivity is negative for both cases, but decreases during reactor operation due to spectral shift. In summary, even after 20 years of operation the total temperature coefficient of reactivity is relatively large and negative during reactor operation (comparing with conventional PWR which has temperature coefficient about $-1.71 \text{ pcm}/^{\circ}\text{F} \approx -3.08 \text{ pcm/K}$ [?]), despite positive MTC, and affords excellent reactor stability and control.

Table 4: Temperature coefficients of reactivity for initial and equilibrium state.

Reactivity coefficient	Initial [pcm/k]	Equilibrium [pcm/k]	Reference (initial/equilibrium)[2]
Doppler in fuel salt	-4.70 ± 0.159	-5.30 ± 0.186	
Fuel salt density	$+1.19 \pm 0.155$	$+2.93 \pm 0.186$	
Total fuel salt	-3.67 ± 0.157	-2.62 ± 0.189	
Doppler in graphite	$+2.00 \pm 0.158$	$+0.85 \pm 0.188$	
Total moderator (graphite)	$+2.00 \pm 0.158$	$+0.85 \pm 0.188$	
Total core	-1.45 ± 0.159	-2.04 ± 0.186	$-2.0 / -1.8$

3.7. Six Factor Analysis

The effective multiplication factor can be expressed using the following formula:

$$k_{eff} = k_{inf} P_f P_t = \eta \epsilon p f P_f P_t$$

205 Table 5 summarizes the six factors for both initial and equilibrium fuel salt composition. Using SERPENT2 and SaltProc, these factors and their statistical uncertainties have been calculated for both initial and equilibrium fuel salt composition (see Table 1). The fast and thermal non-leakage probabilities remain constant despite the evolving neutron spectrum during operation. In 210 contrast, the neutron reproduction factor (η), resonance escape probability (p), and fast fission factor (ϵ) are considerably different between startup and equilibrium. As indicated in Figure 9, the neutron spectrum is softer at the beginning of reactor life. Neutron spectrum hardening causes the fast fission factor to increase through the core lifetime. The opposite is true for the resonance 215 escape probability. Finally, the neutron reproduction factor decreases during reactor operation due to accumulation of fissile plutonium isotopes.

3.8. Thorium refill rate and U233 production

Table 5: Six factors for the full-core MSBR model for initial and equilibrium fuel composition.

Factor	Initial	Equilibrium
Neutron reproduction factor (η)	$1.3960 \pm .000052$	$1.3778 \pm .00005$
Thermal utilization factor (f)	$0.9670 \pm .000011$	$0.9706 \pm .00001$
Resonance escape probability (p)	$0.6044 \pm .000039$	$0.5761 \pm .00004$
Fast fission factor (ϵ)	$1.3421 \pm .000040$	$1.3609 \pm .00004$
Fast non-leakage probability (P_f)	$0.9999 \pm .000004$	$0.9999 \pm .000004$
Thermal non-leakage probability (P_t)	$0.9894 \pm .000005$	$0.9912 \pm .00005$

4. Discussion and conclusions

5. Acknowledgments

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²²⁵ The authors contributed to this work as described below. Osama Ashraf conceived and designed the simulations, wrote the paper, prepared figures and/or tables, performed the computation work, and reviewed drafts of the paper. Osama Ashraf is supported by

²³⁰ Andrei Rykhlevskii conceived and designed the simulations, wrote the paper, prepared figures and/or tables, performed the computation work, contributed to the software product, and reviewed drafts of the paper. Andrei Rykhlevskii is supported by DOE ARPA-E MEITNER program award 1798-1576.

²³⁵ Kathryn D. Huff directed and supervised the work, conceived and designed the simulations, contributed to the software product, and reviewed drafts of the paper. Prof. Huff is supported by the Nuclear Regulatory Commission Faculty Development Program, the National Center for Supercomputing Applications, the NNSA Office of Defense Nuclear Nonproliferation R&D through the Consortium for Verification Technologies and the Consortium for Nonproliferation Enabling Capabilities, the International Institute for Carbon Neutral Energy Research (WPI-I2CNER), sponsored by the Japanese Ministry of Education, Culture, Sports, Science and Technology, and DOE ARPA-E MEITNER program award 1798-1576.

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