Modeling and Simulation of Online Reprocessing in the Thorium-Fueled Molten Salt Breeder Reactor

Andrei Rykhlevskii[a], Jin Whan Bae[a], Kathryn D. Huff[a,*]

[a] *Dept. of Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801*

Abstract

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1. Introduction

The Molten Salt Reactor (MSR) is an advanced type of reactor which was developed at Oak Ridge National Laboratory (ORNL) in the 1950s and was operated in the 1960s. More recently, MSR was included in the six advanced reactor concepts that have been chosen by the Generation IV International Forum (GIF) for further research and development. MSRs offer significant improvements "in the four broad areas of sustainability, economics, safety and reliability, and proliferation resistance and physical protection" [1]. To achieve the goals formulated by the GIF, MSRs attempt to simplify the reactor core and improve inherent safety by using liquid coolant which is also a fuel.

In the thermal spectrum MSR, fluorides of fissile and/or fertile materials (i.e. UF_4, ThF_4, PuF_3, TRU[1]F_3) are mixed with carrier salts to form a liquid fuel which is circulated in a loop-type primary circuit [2]. This innovation leads to immediate advantages over traditional, solid-fueled, reactors. These include near-atmospheric pressure in the primary loop, relatively high coolant temperature, outstanding neutron economy, a high level of inherent safety, reduced fuel

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*Corresponding Author
Email address: kdhuff@illinois.edu (Kathryn D. Huff)
[1] Transuranic elements

preprocessing, and the ability to continuously remove fission products and add fissile and/or fertile elements [3]. The thorium-fueled Molten Salt Breeder Reactor (MSBR) was developed in the early 1970s by ORNL specifically to realize the promise of the thorium fuel cycle, which uses natural thorium instead of enriched uranium. With continuous fuel reprocessing, MSBR is very attractive to effecively realize advantages of the thorium fuel cycle because the ^{233}U bred from ^{232}Th is almost instantly [2] being recycled back to the core [4]. The mixture of LiF-BeF$_2$-ThF$_4$-UF$_4$ has a melting point of 499°C, a low vapor pressure at operating temperatures, and good flow and heat transfer properties [5]. In the matter of nuclear fuel cycle, the thorium cycle produces a reduced quantity of plutonium and minor actinides (MAs) compared to the traditional uranium fuel cycle. Finally, the MSR also could be employed as a converter reactor for transmutation of spent fuel from current Light Water Reactors (LWRs).

Modeling liquid-fueled systems with existing neutron transport and depletion tools is challenging because most of these tools are designed for the solid-fueled reactors simulation. The fuel material flows and potential online separations or feeds of specific elements or nuclides are the main challenges of liquid-fueled systems. Furthermore, no established tool for liquid-fueled MSR neutronics and fuel cycle evaluation exist, though internally developed tools from universities and research institutions can approximate online refueling [6]. The foundation for these tools was based on early MSR simulation methods at ORNL, which integrated neutronics and fuel cycle codes (i.e., ROD [7]) into operational plant tools (i.e., MRPP [8]) for MSR and reprocessing system design. More recent research efforts in Europe and Asia mainly focus on fast spectrum reactor fuel cycle analysis and couple external tools to neutron transport and depletion codes take into account continuous feeds and removals in MSRs. Four of these efforts are listed in table 1.

[2] ^{232}Th transmutes into ^{233}Th after capturing a neutron. Next, this isotope decays to ^{233}Pa ($\tau_{1/2}$=21.83m), which finally decays to ^{233}U ($\tau_{1/2}$=26.967d).

Table 1: Tools and methods for fast spectrum system fuel cycle analysis.

#	Neutronic code	Depletion code	Authors	Spectra
1	MCNP [9]	REM [10]	Doligez *et al.*, 2014; Heuer *et al.*, 2014 [11, 12]	fast
2	ERANOS [13]	ERANOS	Fiorina *et al.*, 2013 [14]	fast
3	KENO-IV [15]	ORIGEN [16]	Sheu *et al.*, 2013 [17]	fast
4	SERPENT 2 [18]	SERPENT 2	Aufiero *et al.*, 2013 [19]	fast
5	MCODE [20]	ORIGEN2 [21]	Ahmad *et al.*, 2015 [22]	thermal
6	MCNP6	CINDER90 [23]	Park *et al.*, 2015; Jeong *et al.*, 2016 [24, 25]	thermal
7	SCALE [26]	SCALE/ ChemTriton [27]	Powers *et al.*, 2014; Betzler *et al.*, 2017 [27, 28, 29]	thermal
8	SERPENT 2	SERPENT 2	Rykhlevskii *et al.*, 2017 [30]	thermal
9	MCNP	REM	Nuttin *et al.* [31]	thermal

Most of these methods are also applicable to thermal spectrum MSRs. Additional tools developed specifically for thermal MSR applications are also listed in table 1.

Methods (1, 3, 4) provide some form of reactivity control, and methods (1, 4, 5, 6, 8, 9) use a set of all nuclides in depletion calculations.

Liquid-fueled MSR designs have online separations and/or feeds, where material is moved to or from the core at all times (continuous) or at specific time steps (batch). To account for batch discharge, a depletion tool must remove some or all material at specified intervals. This requires the burn-up simulation to stop at a given time and restart with a new liquid fuel composition (after removal of discarded materials and addition of fissile/fertile materials). Accounting for a continuous removal or addition is more difficult because it requires adding a term to the Bateman equations. In SCALE [26], ORIGEN [16] solves a set of Bateman equations using spectrum-averaged fluxes and cross sections generated from a deterministic transport calculation. Methods (1, 4, 8) model true continuous feeds and removals, while other methods employ a batch-wise approach. ORNL researchers have developed ChemTriton, a Python-based script for SCALE/TRITON which uses a semi-continuous batch process to simulate a continuous reprocessing. This tool models salt treatment, separations, discharge, and refill using a unit-cell MSR SCALE/TRITON model over small time steps to simulate continuous reprocessing and deplete the fuel salt [27].

Thorium-fueled MSBR-like reactors similar to the one in this thesis are described in (6, 7, 8, 9). Nevertheless, most of these efforts considered only simplified unit-cell geometry because depletion computations for a many-year fuel cycle are computationally expesive even for simple models.

Nuttin *et al.* broke up reactor core geometry into tree MCNP cells: one for salt channels, one for two salt plena above and below the core and the last cell for the annulus, consequently, two-region reactor core was approximated by one region with averaged fuel/moderator ratio [31]. A similar approach was used by Powers *et al.*, Betzler *et al.*, and Jeong *et al.* [27, 28, 4, 29, 32, 25] and clearly misrepresent the two-region breeder reactor concept. The unit-cell or one-region models may produce reliable results for homogeneous reactor cores (i.e. Molten Salt Fast Reactor (MSFR), Molten Salt Actinide Recycler and Transmuter (MOSART)) or for one-region single-fluid reactor designs (i.e. Molten Salt Reactor Experiment (MSRE)). A two-region MSBR must be simulated using a whole-core model to represent different neutron transport in the inner and outer regions of the core, because most fissions happens in the inner region while breeding occurs in the outer zone.

Aufiero *et al.* extended the Monte Carlo burnup code SERPENT 2 and employed it to study the material isotopic evolution of the MSFR. The developed extension directly takes into account the effects of online fuel reprocessing on depletion calculations and features a reactivity control algorithm. The extended version of SERPENT 2 was assessed against a dedicated version of the deterministic ERANOS-based EQL3D procedure [13] and adopted to analyze the MSFR fuel salt isotopic evolution. We employed this extended SERPENT 2 for a simplified unit-cell geometry of thermal spectrum thorium-fueled MSBR and obtained results which contradict existing MSBR depletion simulations [25].

The present work introduces the online reprocessing simulation code, SaltProc, which expands the capability of the continuous-energy Monte Carlo Burnup calculation code, SERPENT 2 [18], for simulation liquid-fueled MSR operation [33]. It is also reports the application of the coupled SaltProc-SERPENT 2 system to the MSBR, which represents the continuation of the work presented in [34, 30]. The major objective of the work herein is to analyze MSBR neutronics and fuel cycle to find the equilibrium core composition and core depletion. The additional objective is to compare predicted operational and safety parameters of the MSBR at both the initial and equilibrium states. Finally, ^{232}Th feed rate will be determined and MSBR fuel cycle performance will be analyzed.

The MSBR complex geometry is hard to describe in software input, and, usually, researchers make significant geometric simplifications to model it [24]. Note that this thesis leverages extensive computational resources to avoid these geometric approximations and accurately capture breeding behavior. This article only discusses liquid-fueled MSRs. Another challenge of the MSRs, its delayed neutron precursor drift related to circulating liquid fuel, is not treated here.

2. Methods

Liquid-fueled system modeling with contemporary reactor physics codes is difficult because most of these codes were developed for analyzing solid-fueled reactors. In fact, world-wide commercial reactors fleet today is represented by systems with solid fuel. Liquid-fueled systems ability to continuously remove fission products and add fissile and/or fertile elements is the main challenge for depletion simulations. SaltProc takes into account online separations and feeds using SERPENT 2 continuous-energy Monte Carlo neutron transport and depletion code.

2.1. Molten Salt Breeder Reactor design and model

The 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 salt flows through channels in moderating and reflecting graphite blocks. Salt at about 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 demonstrate the configuration of the MSBR vessel, core configuration, "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. Zone I composed of 1,320 graphite cells, 2 graphite control rods, and 2 safety[3] 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. 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.

Since reactor graphite experiences significant dimensional changes due to neutron irradiation, the reactor core was designed for periodic replacement. Based on the irradiation experimental data from MSRE, core graphite lifetime

[3]These rods needed for emergency shutdown only.

is about 4 years and reflector graphite lifetime is 30 years [5].

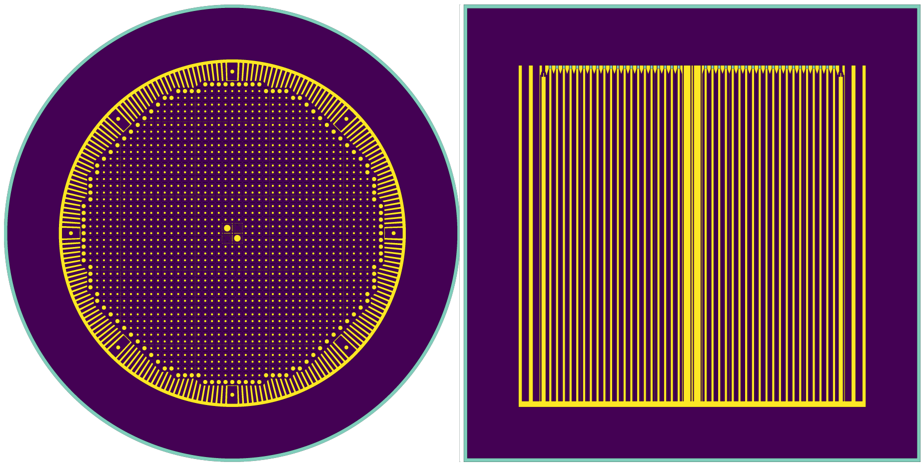

Figure 1: Plan and elevation view of SERPENT 2 MSBR model developed in this work.

Moreover, it was decided to remove and install the core graphite as an assembly rather than by individual blocks, because it is relatively easier for maintenance personnel and has lower probability of radioactive elements escape due to used blocks damage during removal. In addition, handling the core as an assembly also allows the replacement core to be carefully preassembled and tested under factory conditions.

There are eight symmetric graphite slabs with a width of 15.24 cm in zone II, 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 demonstrates the 5.08-cm-wide annular space between the removable core graphite in zone II-B and the permanently mounted reflector graphite. This annulus constists entirely of fuel salt, provides space for moving the core assembly, helps compensate the elliptical dimensions of the reactor vessel, and serves to reduce the damaging flux at the surface of the graphite reflector blocks. In this work, all figures of the core were generated using the built-in SERPENT plotter.

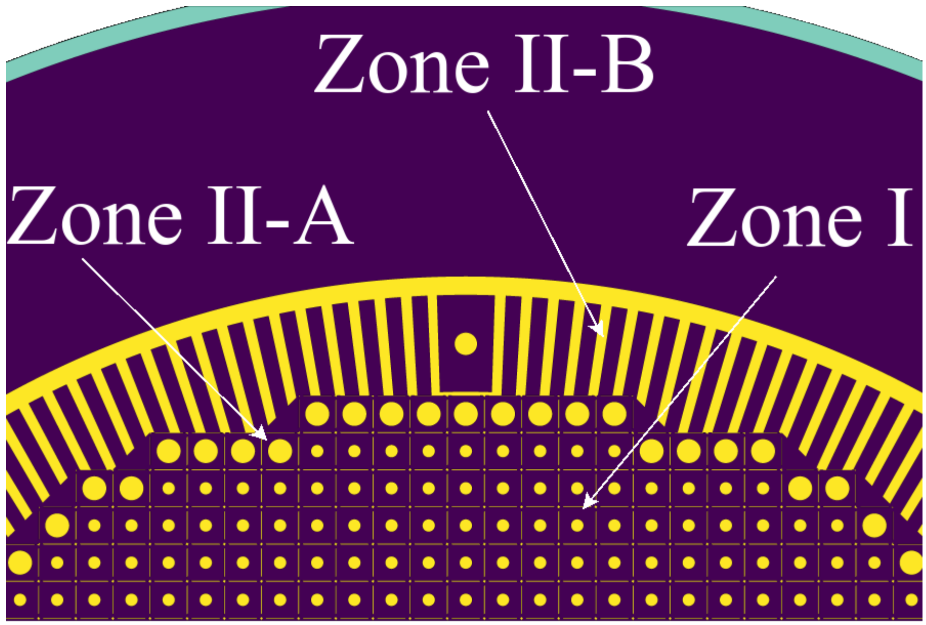

Figure 2: Detailed view of MSBR zone II model.

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. In zone I, 13% of the volume is fuel salt and 87% is graphite. Zone I is composed of 1,320 graphite cells and 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.

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. Zone I is well-moderated to achieve the desired fission power density. Figure 3 demonstrates the elevation and sectional views of graphite elements of zone I [5] and their SERPENT model [34].

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. This zone is formed of two kinds of elements: large-diameter fuel channels (zone II-A) and radial 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 and have elliptical-shaped dowels running axially between the prisms and needed to isolate the fuel salt flow in zone I from that in zone II. Figure 4 shows shape 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 **??**. Zone II serves as a blanket to achieve the best performance: a high breeding ratio and a low fissile inventory. The neutron energy spectrum in zone II is made harder to enhance 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 [5].

The main challenge was to accurately represent zone II-B because it has irregular elements with sophisticated shapes. From the ORNL report [5], the suggested design of zone II-B has 8 irregularly-shaped graphite elements every 45° as well as salt channels (figure **??**). 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 37%, so that this simplification did not considerably change the core neutronics. This is the only simplification made to the MSBR geometry in this work.

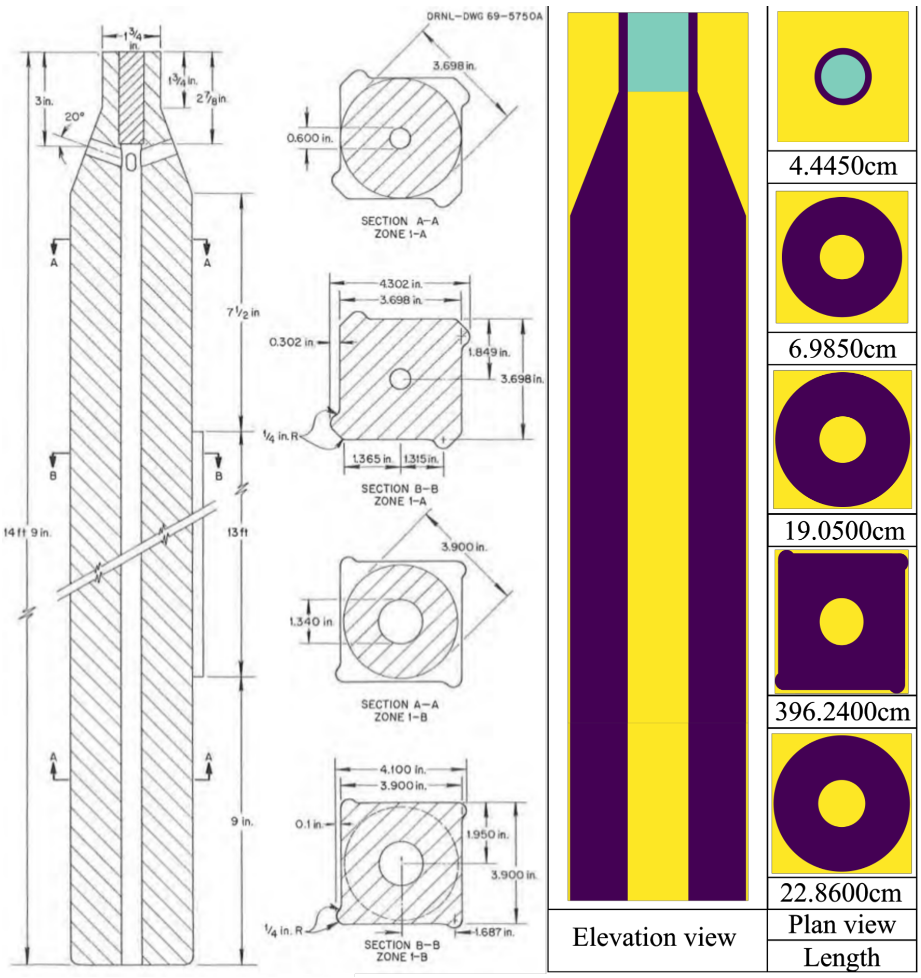

Figure 3: Graphite moderator elements for zone I [5, 34].

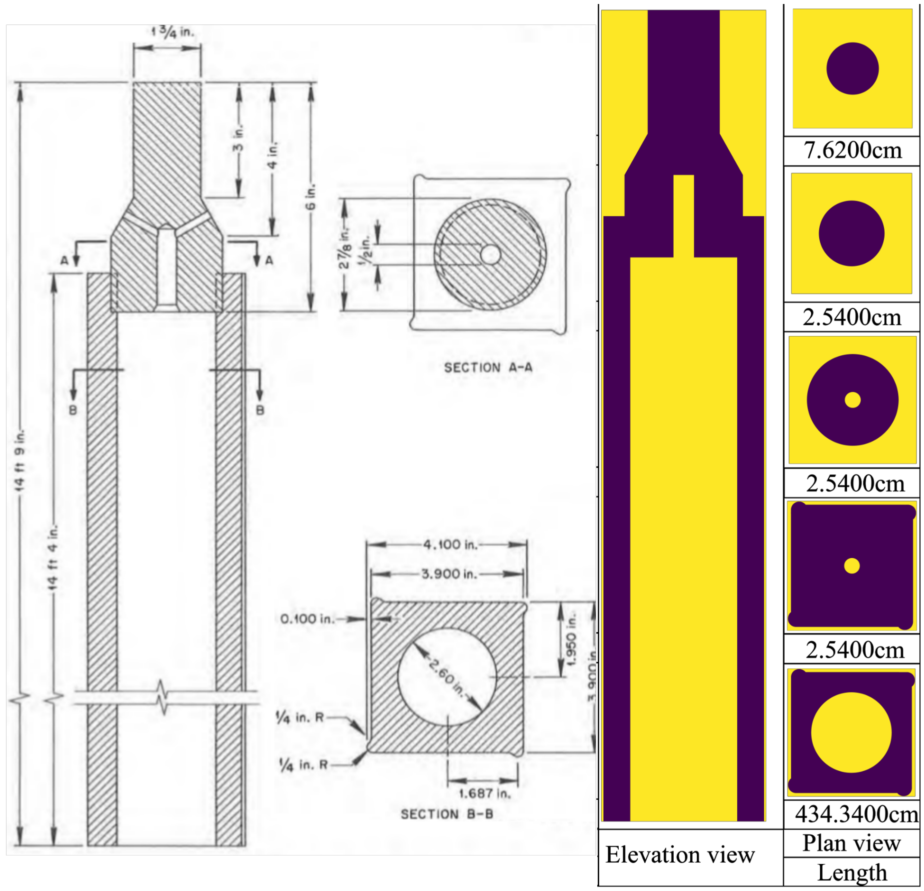

Figure 4: Graphite moderator elements for zone II-A [5, 34].

2.1.3. Material composition and normalization parameters

The fuel salt, the reactor graphite, and the modified Hastelloy-N[4] are materials unique of the MSBR and were created at ORNL. The initial fuel salt used the same density (3.35 g/cm^3) and composition LiF-BeF$_2$-ThF$_4$-^{233}UF$_4$ (71.8-16-12-0.2 mole %) as the MSBR design[5]. The lithium in the molten salt fuel is fully enriched in ^{7}Li because ^{6}Li is a very strong neutron poison and becomes tritium upon neutron capture.

[4] Hastelloy-N is very common in reactors now but have been studied and developed at ORNL in a program that started in 1950s.

For cross section generation, JEFF-3.1.2 neutron library was employed [35]. The specific temperature was fixed for each material to correctly model the Doppler-broadening of resonance peaks when SERPENT generates the problem-dependent nuclear data library. The isotopic composition of each material at the initial state was described in detail in the MSBR conceptual design study [5] and has been applied to SERPENT model without any modification. Table 2 is a summary of the major MSBR parameters used by this model [5].

Table 2: Summary of principal data for MSBR [5].

Thermal capacity of reactor	2250 MW(t)
Net electrical output	1000 MW(e)
Net thermal efficiency	44.4%
Salt volume fraction in central core zone	0.13
Salt volume fraction in outer core zone	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	71.85-16-12-0.25 mole%
Fuel salt density	3.35 g/cm^3

Removing specific chemical elements from a molten salt is a complicated task that requires intelligent design (e.g., chemical separations equipment design, fuel salt flows to equipment) and has a considerable economic cost. This section contains MSBR chemical processing plant and gas separation system brief overview.

- MSBR design description.

- SERPENT2 very short overview (couple paragraphs).
- Full-core model description.
- Online reprocessing method. Advantages and disadvantages of batch-wise approach. SaltProc capabilities description.

3. Results

- K-eff over 40 years of simulation with <15pcm uncertainty.
- Dynamics isotope composition from initial to equilibrium composition.
- Dynamics of fissile vs non-fissile isotopes over 40 years.
- Neutron spectrum for both states and separately for Zone I and Zone II (probably, even Zone II-A and II-B).
- Power distribution plot (without breeding, too much pics).
- Control rod worth & Six factor analysis & temperature coefficients.
- ^{232}Th refill rate.
- Bried discussion (2-3 paragraph).

4. Conclusion

Condensed copy-paste of the thesis conclusion.

- Full-core model which is better the most exists. Importance of full-core approach for multi-region designs.
- K-eff dynamics and explanation of this dynamics.
- Spectral shift explanation.
- Why spectral shift causes safety parameters worsening and power profile changes.
- ^{232}Th rate is in a good agreement with references.

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Kathryn D. Huff directed and supervised the work, conceived and designed the simulations, wrote the paper, 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 Verfication Technologies and the Consortium for Nonproliferation Enabling Capabilities, and the International Institute for Carbon Neutral Energy Research (WPI-I2CNER), sponsored by the Japanese Ministry of Education, Culture, Sports, Science and Technology.

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