

## Online reprocessing simulation for thorium-fueled molten salt breeder reactor

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### INTRODUCTION

The thermal spectrum Molten Salt Reactor (MSR) is an advanced type of reactor that consists of constantly circulating liquid fuel (i.e., mixture of LiF-BeF<sub>2</sub>-ThF<sub>4</sub>-UF<sub>4</sub> or LiF-BeF<sub>2</sub>-ZrF<sub>4</sub>-UF<sub>4</sub>) and solid graphite moderator structures. This liquid fuel form leads to immediate advantages over traditional, solid-fueled, reactors. The molten-salt carrier with dissolved fissile and/or fertile material allows for online refueling and reprocessing. Thus, MSRs can potentially operate years without shutdown, achieving maximum fuel utilization and outstanding neutron economy [1]. Moreover, this type of fuel does not need complex fabrication since the fissile material could be transported from any enrichment plant to the Nuclear Power Plant (NPP) in the form of uranium hexafluoride (UF<sub>6</sub>). Additionally, MSRs have a high level of inherent safety due to their characteristically the strong negative temperature coefficient of reactivity, near-atmospheric pressure in the primary loop, stable coolant, passive decay heat cooling, and small excess reactivity [2].

The thorium-fueled Molten Salt Breeder Reactor (MSBR) was developed in the early 1970s by Oak Ridge National Laboratory (ORNL) specifically to realize the promise of the thorium fuel cycle which allows the use of natural thorium instead of enriched uranium as the fertile element. Thorium breeds the fissile <sup>233</sup>U and avoids uranium enrichment [3]. 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. Consequently, it may significantly increase proliferation resistance when the MSR operates in the breeder regime. The MSRs also could be employed as a converter reactor for transmutation of spent fuel from current Light Water Reactors (LWRs).

Recently, interest in MSRs has resurged, with multiple new companies pursuing commercialization of MSR designs<sup>1</sup>. To further develop these MSR concepts, particularly with respect to their strategies for online reprocessing and refueling, computational analysis methods capturing their unique reactor physics and process chemistry are needed. However, most contemporary nuclear reactor physics software is unable to perform depletion calculations in an online reprocessing regime. Powers *et al.* suggested a novel method for conducting depletion simulations for MSR. This suggested method takes into account fuel salt composition changes due to online reprocessing and refueling based on the deterministic computer code NEWT in SCALE [4]. This approach was later used by Jeong *et al.* to find an equilibrium fuel composition for the MSBR and was validated with a Monte Carlo N-Particle code (MCNP)/CINDER90 model [5].

The current paper presents a single-cell model developed using the continuous-energy Serpent 2 Monte Carlo reactor physics software. It was employed to establish a Serpent-based method for finding the equilibrium core composition and core depletion of the MSBR. All calculations presented in this paper were performed using the Serpent 2 code version 2.1.29 with ENDF/B-VII.0 nuclear data [6, 7]. Serpent 2 is an improvement upon Serpent 1, and contains a complete redesign of memory management using hybrid OpenMP + MPI parallelization. This hybrid parallelization is important in depletion calculations using computer clusters with multiple cores [8]. This work used the built-in Serpent 2 depletion capabilities and its built-in online reprocessing subroutine. Another feature of the MSBR, its circulating liquid fuel and corresponding delayed neutron precursor drift, is not treated here.

### DESCRIPTION OF THE ACTUAL WORK

The MSBR is a thermal spectrum reactor. The reactor vessel has a diameter of 680 cm and a height of 610 cm. It contains a molten fluoride fuel-salt mixture which performs two functions: to generate heat in the moderated region and to transport heat energy from the core to primary heat exchanger using the primary salt pump. The vessel also contains graphite blocks for neutron moderation and reflection. The lithium in the fuel-salt solution is enriched to 99.995% <sup>7</sup>Li because <sup>6</sup>Li is a very strong neutron poison and becomes tritium upon neutron bombardment. In this study, the 0.005% atomic fraction of <sup>6</sup>Li has been taken into account because even such a small amount of isotope with very high absorption cross section can significantly impact the neutron flux energy distribution and, consequently, depletion calculation results. Table I is the summary of the major MSBR parameters used by this model [3].

The MSBR core consists of two different zones. The central zone, Zone I, in which 13.2% of the volume is fuel salt and 86.8% graphite. Zone I composed of 1320 graphite cells, 2 graphite control rods, and 2 safety rods consisting of boron carbide clad. 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. In this work, only a Zone I element with a 3.42138 cm diameter central fuel channel and molten-salt fuel volume fraction of 0.132 was considered. Figure 1 shows the geometry of the unit cell model. The boundary condition of the unit cell is periodic. The density of the fuel salt is 3.3304 g/cm<sup>3</sup> and the graphite density is 1.843 g/cm<sup>3</sup>. The temperature of fuel and graphite is fixed at 908K for the whole reprocessing cycle [3].

<sup>1</sup>Examples include both liquid-fueled molten salt designs from Transatomic, Terrapower, Terrestrial, and Thorcon.

TABLE I: Summary of principal data for MSBR.

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	0.132
Salt volume fraction in outer core zone	0.37
Fuel-salt inventory (Zone I)	8.2 m <sup>3</sup>
Fuel-salt inventory (Zone II)	10.8 m <sup>3</sup>
Fuel-salt inventory (annulus)	3.8 m <sup>3</sup>
Fuel salt components	LiF-BeF <sub>2</sub> - ThF <sub>4</sub> - <sup>233</sup> UF <sub>4</sub> - <sup>239</sup> PuF <sub>3</sub>
Fuel salt composition	71.767-16-12- 0.232-0.0006 mole%

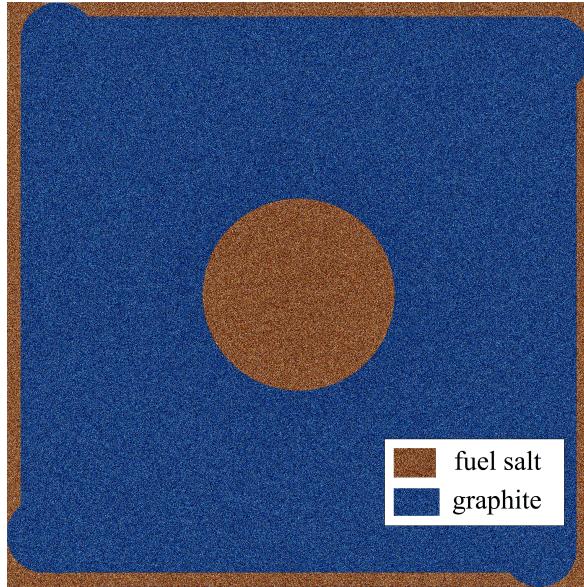


Fig. 1: Molten Salt Breeder Reactor (MSBR) unit cell of Zone I geometry.

### Online reprocessing method

Currently, researchers investigating the MSRs typically develop custom supporting tools and scripts to simulate online reprocessing and refueling. Most utilize stochastic (i.e. MCNP) or deterministic (i.e. SCALE) methods for depletion and supplement these with custom Python scripts to capture the impacts of reprocessing [5, 9]. Serpent 2 is the first commonly used Monte Carlo software to support continuous material reprocessing. It does so by allowing the user to define multiple material flows into and out of the fuel. This work applies this Serpent 2 feature to the MSBR.

The MSBR has the capability to remove all poisons (e.g.

<sup>135</sup>Xe), noble metals, and gases (e.g. <sup>75</sup>Se, <sup>85</sup>Kr) every 20 seconds. The <sup>232</sup>Th in the fuel absorbs thermal neutrons and produces <sup>233</sup>Pa which then decays into the fissile <sup>233</sup>U. Protactinium presents a challenge, since it has a large absorption cross section in the thermal energy spectrum. Accordingly, <sup>233</sup>Pa is continuously removed from the fuel salt into a protactinium decay tank and allowing <sup>233</sup>Pa to decay to <sup>233</sup>U without poisoning the reactor. The reactor reprocessing system is designed to separate <sup>233</sup>Pa from the molten-salt fuel over 3 days, hold it while <sup>233</sup>Pa decays into <sup>233</sup>U, and return it back to the primary loop. This feature allows the reactor to avoid neutron losses to protactinium, keeps fission products to a very low level, and increases the efficiency of <sup>233</sup>U breeding [3].

Since removal rates vary among nuclides in this reactor concept, the Serpent 2 reprocessing method is unable to capture the desired reprocessing strategy. The removal rates also 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, serious impacts of short lived fission products are not captured in a manner that is faithful MSBR conceptual design. To compromise, the time interval for depletion calculations in this model was selected as 3 days to correlate with the removal interval of <sup>233</sup>Pa and Thorium was continuously added to maintain the initial mass fraction of <sup>232</sup>ThF<sub>4</sub>.

## RESULTS AND ANALYSIS

Using the methodology described previously, the MSBR unit cell depletion analysis was performed to find equilibrium core conditions. Calculation results reported in this section include multiplication factor, neutron flux energy spectrum, and atomic density of major isotopes.

### Equilibrium state analysis

This analysis models a single representative unit cell rather than the whole MSBR core. Consequently, it does not take into consideration different fuel-moderator volume ratios for Zone I, Zone II, the annulus, and the reflector. The initial multiplication factor during depletion calculation is selected for a state with fully withdrawn control rods, which gives considerable excess reactivity in the beginning of the cycle (approximately 5000 pcm). The standard deviation for these calculations is approximately 100 pcm. Figure 2 shows the infinite multiplication factor for 1200-days reprocessing cycle calculated by Serpent 2 with ENDF/B-VII.0 nuclear data. A significant standard deviation causes the multiplication factor fluctuations visible in this plot.

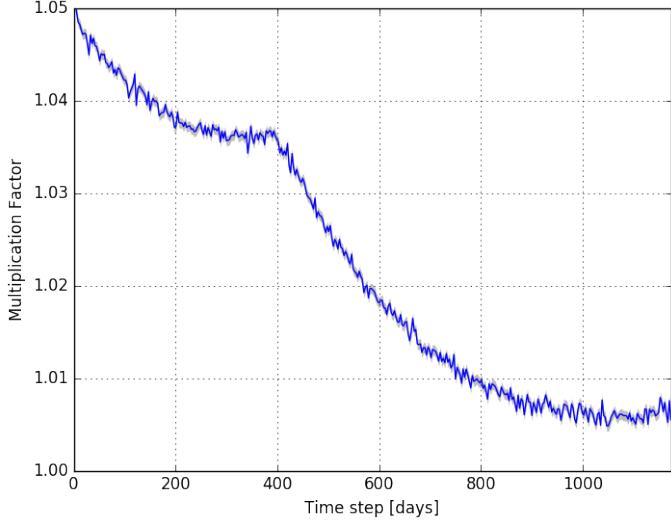


Fig. 2: Infinite multiplication factor during a 1200 days depletion simulation. The confidence interval  $\pm\sigma$  is shaded.

Protactinium-233 is continuously removing into the tank for protactinium decay.  $^{233}\text{Pa}$  has a half-life of 27 days and beta decays into  $^{233}\text{U}$  which as fresh fuel goes back to the reactor core. The infinite multiplication factor decreases first 400 days of depletion due to strong absorbers (e.g.  $^{233}\text{Th}$ ,  $^{234}\text{U}$ ) accumulation which causes relatively high fuel ( $^{233}\text{U}$ ) refill inflow to keep reactor critical. During reactor operation producing fissile materials other than  $^{233}\text{U}$  in the core (e.g.  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ) which makes it possible to decrease fresh fuel refill rate after 1 year of operation.

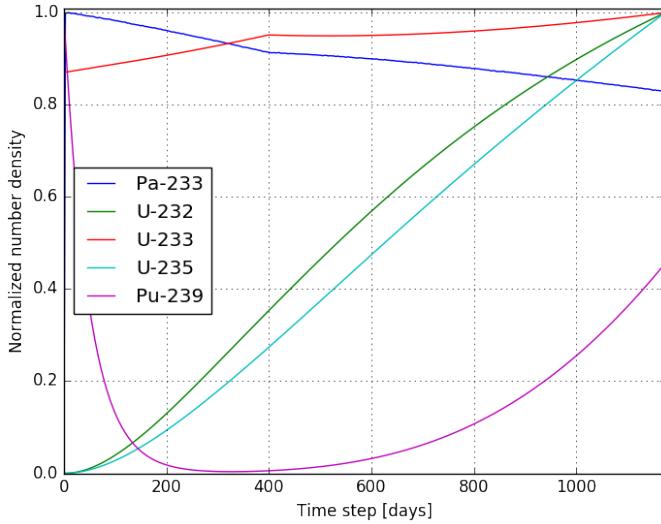


Fig. 3: Normalized number density of major isotopes during 1200 days of depletion.

The analysis of the fuel salt composition variation gives clearer information about the equilibrium core state. Figure 3 shows the normalized number density of isotopes influential to core neutronics at the beginning of each depletion time interval. The number density of protactinium is very low (less

than  $10^{16} \text{ 1/cm}^3$ ) but some small amount of it is produced during the 3-day reprocessing period. In this assessment, the multiplication factor stabilizes after approximately 950 days. Figure 4 represents the rates of online reprocessing material flows over the 4-year depletion calculation. In Figure 4

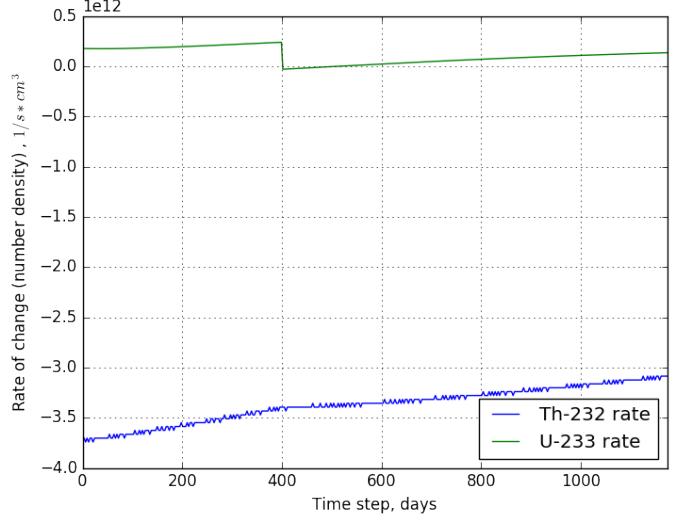


Fig. 4: Materials flows rates during online reprocessing.

we can see that, to keep the reactor critical, a higher  $^{233}\text{U}$  flow rate from the protactinium decay tank was required for first 400 days of operation. After that, the  $^{233}\text{U}$  flow rate can be reduced. The  $^{232}\text{Th}$  rate slightly decreases over 4 years of operation due to other than  $^{233}\text{U}$  fissile materials accumulation.

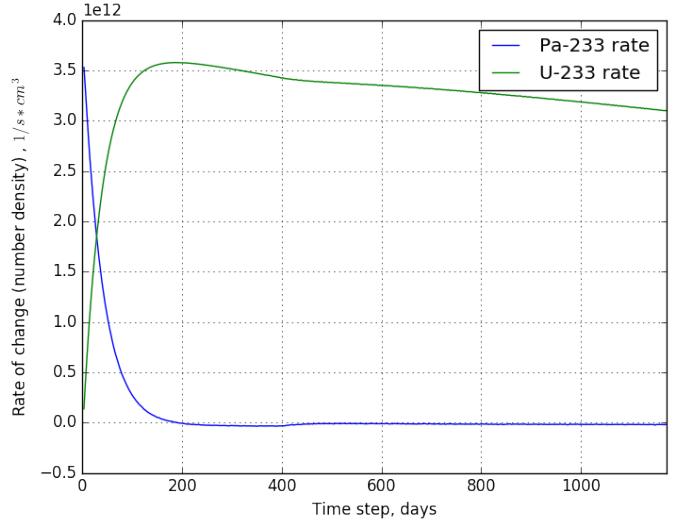


Fig. 5: Materials flows rates for the protactinium decay tank during MSBR online reprocessing.

As shown in Figure 5, the tank for protactinium decay accumulates  $^{233}\text{Pa}$  for approximately 200 days. Fresh  $^{233}\text{U}$  fuel flow is also established after 200 days. Uranium produced in the tank by protactinium decay is separated by circulation of the salt through a flourinator. The fully processed molten salt, on its way back to the primary loop, has uranium added

at the rate required to maintain or adjust the fissile material concentration and, hence, the reactivity, in the reactor core as desired.

### Neutron spectrum

Figure 6 represents the neutron flux per lethargy energy distribution of the initial and the equilibrium core compositions. The spectrum for the equilibrium state is harder than the initial state due to heavy fission products.

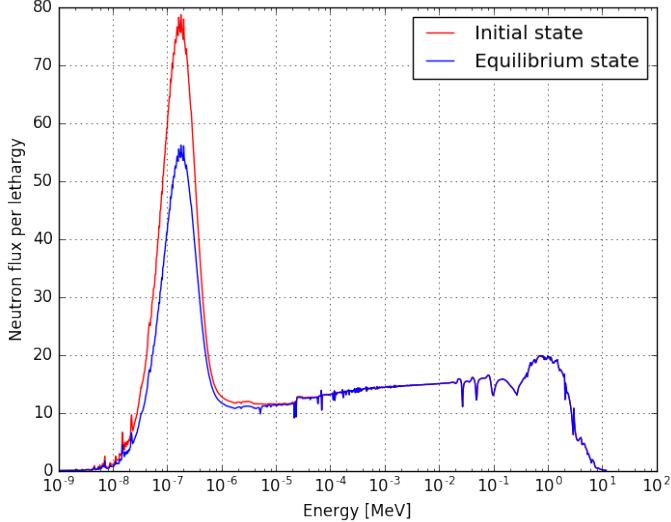


Fig. 6: Neutron spectrum for initial and equilibrium composition (normalized per lethargy).

In this work, only the Zone I unit cell, where the fuel salt volume fraction is 13.2%, was considered. The neutron spectrum for Zone II cell, where the fuel salt volume fraction is 37% is expected to be harder and the peak in thermal energy region is predicted to be much lower. To obtain a high-fidelity neutron energy spectrum, a full-core MSBR analysis is required.

### CONCLUSIONS

The depletion calculation of the MSBR unit cell model for finding the equilibrium states was performed using the Serpent 2 Monte Carlo code to simulate simplified case of the online reprocessing and refueling to find equilibrium material composition. When running depletion calculation, the fission products are removed and fertile/fissile materials are added to fuel salt every 3 days. The important MSR feature, online reprocessing & refueling is implemented in the Serpent 2 material burnup routine. The results of this study indicate that from the depletion calculation the multiplication factor slowly decreases and reaches to the equilibrium state. The most obvious finding to emerge from the analysis of initial and equilibrium materials composition is that neutron energy spectrum is harder for equilibrium state because significant amount of heavy fission products were accumulated in the MSBR core.

These results are contrary with those of Jeong and Park

(2016) who suggested two different unit cell models and uses MCNP with Python-script to simulate MSBR online reprocessing to find equilibrium composition. This inconsistency may be due to different fuel fraction in the unit cell (Jeong and Park selected 20.6% salt fraction). To obtain better results for this online reprocessing simulation, many future efforts are planned. First, a depletion simulation will be performed using a full-core, three-dimensional, high-fidelity model of MSBR that has been developed in Serpent 2. In this case, different fuel-moderator volume ratios for the reactor Zone I, Zone II, annulus, and reflectors will be taken into account to find accurate multiplication factor, neutron spectrum and, hence, depleted composition. Secondly, an additional Serpent 2 flow control system subroutine should be developed to simulate adjusting material flows (e.g. rate of removing  $^{233}\text{Pa}$  from the salt and adding fissile  $^{233}\text{U}$  from the tank for protactinium decay) depending upon the instantaneous reactivity value, which is a more promising reactivity control method than moving control rods. Finally, the temperature effect of reactivity for both fuel salt and graphite should be calculated to find optimal effective multiplication factor range.

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