

FUEL CYCLE PERFORMANCE OF FAST SPECTRUM MOLTEN SALT REACTOR DESIGNS *

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ABSTRACT

In the search for new ways to generate low-carbon, reliable base-load power, a resurgence of interest in advanced nuclear energy technologies, including Molten Salt Reactors (MSRs), has produced multiple new conceptual MSRs including fast neutron spectrum designs. The fuel cycle performance of four historical fast MSR designs is analyzed using a recently developed SCALE/TRITON 6.2.4 Alpha with a continuous online reprocessing functionality. The fast spectrum and continuous feed and removal of material enable these concepts to have remarkable fuel cycle metrics: (1) resource utilization is approximately 18 times better than for a typical low-enriched thermal spectrum once-through fuel cycle (i.e., from approximately 180 t/GWe-year to 1 t/GWe-year); (2) fast MSRs generate approximately 25 times less nuclear waste than the current once-through fuel cycle. These metrics are consistent with the Evaluation and Screening Study, which produced a technology-agnostic quantification of the characteristic performance of alternate fuel cycles. Additionally, full-core and unit cell transport models were created and compared to verify the viability of using simplified unit cell geometries for long-term depletion simulation. The unit cell approximation provided a speedup of 20 times relative to the full-core simulation, with depleted mass relative error for major isotopes of less than 2%. Additional fast MSRs design and analysis challenges associated with different fuel cycles and the use of MSR technology are addressed and discussed.

KEYWORDS: molten salt reactor, fast reactor, depletion, fuel cycle, salt treatment, salt separations

1. INTRODUCTION

Liquid-fueled molten salt reactor (MSR) concepts are one of many advanced reactor technologies that promise a competitive and sustainable energy source (1). In these MSRs, fissile and/or fertile

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materials are dissolved in a carrier molten salt (e.g., LiF or NaCl), which offers several potential advantages over solid-fueled reactors. These characteristics include improved neutron economy, reduced fuel preprocessing, and the ability to continuously remove fission products and add fissile and/or fertile elements without shutdown (2). Additional beneficial characteristics include a near-atmospheric pressure in the primary loop, relatively high coolant temperature, and the potential for inherent safety.

Historically, operational experience was limited to thermal spectrum MSR concepts with a solid graphite moderator. Oak Ridge National Laboratory (ORNL) operated an \approx 8 MW_{th} Molten Salt Reactor Experiment (MSRE) from 1965 to 1969 to test approaches and materials, demonstrate fissile recycle (both ^{233}U and ^{235}U), and determine generic MSR operational characteristics (3). Experience and promising breakthroughs in reprocessing technologies (4) led ORNL to the simply configured, graphite-moderated (thermal or intermediate spectrum), single-fluid Molten Salt Breeder Reactor (MSBR) by the end of the 1960s. The primary weakness of these single-fluid thermal MSR concepts is that the hundreds of tons (\approx 300 t for MSBR (5)) of expensive, radiologically contaminated, and irradiated graphite require replacement every 4–10 years, raising significant waste and economical issues.

In contrast, consistent with Generation IV International Forum (GIF) sustainability and safety goals (6), unmoderated (no graphite), one- or two-fluid (blanket-equipped) fast spectrum MSR concepts eventually became the Euratom “reference” MSR (7). Fast MSR systems hold additional advantages over conventional light water reactors (LWRs): (1) they can operate as a breeder (i.e., with a conversion ratio (CR)^{*} > 1) (7; 8; 9), fertile-free transuranic (TRU) burner, or U/Th-supported TRU burner (10); (2) they have the potential to be used in both $^{232}\text{Th}/^{233}\text{U}$ and $^{238}\text{U}/^{239}\text{Pu}$ fuel cycles; (3) they can be operated in ways that would generate very little long-lived TRU waste; and (4) they use reduced amounts of natural resources (e.g., natural uranium, natural thorium) per unit energy generated. To quantitatively estimate the benefits of these capabilities, a fuel cycle performance analysis for various fast spectrum MSR concepts is necessary.

Much of the analysis herein uses unit cell representations of four different fast MSR designs: (1) European Molten Salt Fast Reactor (MSFR) (7); (2) Molten Chloride Salt Fast Reactor (MCSFR) (8); (3) REBUS-3700 (9); and (4) Molten Salt Actinide Recycler and Transmuter (MOSART) (10). Some of these designs are two fluid (1, 2), operate in a thorium fuel cycle (1, 4), or use TRU as the start-up fissile material (3, 4). Concepts (2) and (3) use chloride salts,[†] while (1) and (4) use fluorides. This paper discusses the fuel cycle simulation of these concepts to quantify the fuel cycle performance of fast MSRs.

2. METHODS

Full-core and simplified unit cell models of four different fast spectrum MSRs designs are created (Fig. 1) to determine fuel cycle performance parameters. For full-core models, realistic vacuum boundary conditions were applied while unit cell simplified models used reflective boundary

*CR \equiv fissile generated/fissile consumed: if CR < 1 , the reactor is a “converter”; CR $\equiv 1$, an “isobreeder”; CR > 1 , a “breeder.”

[†]The chlorine in the MCSFR is fully enriched in ^{37}Cl because ^{35}Cl (76% abundance) is a very strong neutron poison in the fast-neutron energy range.

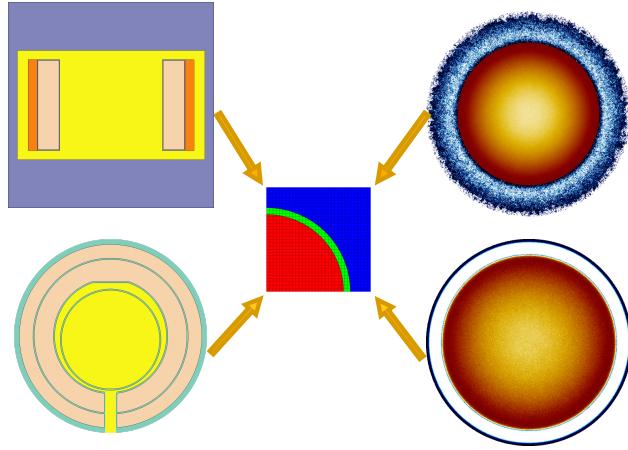


Figure 1: Full-core 3D models of MSFR (upper left), MCSFR (lower left), REBUS-3700 (upper right), and MOSART (lower right) and 2D representative unit cell model (center) showing fuel salt (red), fertile salt (green), and structural material (blue).

conditions that do not take into account neutron leakage (infinite reactor was assumed). Table 1 contains a summary of the principal data of these designs. Fuel cycle performance analysis requires a depletion simulation for each reactor concept over the system lifetime, which in this work is assumed to be 60 years.[†] Full-core 60-year depletion calculations for MSRs are computationally prohibitive. Computation time can be significantly reduced by performing depletion simulations for representative simplified unit cells instead of complex full-core geometry.

2.1. Tool description

All salt treatments and separation in this work are performed as truly continuous (online) processes using SCALE/TRITON version 6.2.4 Alpha (11) with the 238-group ENDF-B/VII.0 cross-section library (12). Some comparisons to simulations using an internal 300-group fast spectrum library yielded similar results at a greater computational cost. Several earlier studies used a batch-wise approach because most reactor physics depletion tools are designed to model batch-fueled reactors with solid fuel (13; 14; 15). This approach requires short depletion time steps to minimize the impact of material feeds and removals during a given time step, leading to a large number of depletion time steps (i.e., transport calculations). This makes lifetime-long depletion calculations computationally expensive. ORIGEN (16) supports continuous feeds and removals but does not track the feed and discharge materials that are important to fuel cycle performance analysis. Thus, the version of SCALE/TRITON currently under testing at ORNL enables the simulation of continuous processes and the tracking of removed materials over the MSR lifetime at a reasonable computational cost.

SCALE/TRITON Alpha currently supports only constant or piecewise feed and removal rates. Therefore, these rates must be determined for each feed and removal before a simulation is run and cannot be adjusted during simulation. One of the features of fast spectrum MSRs is the control

[†]The lifetime of a fast MSR is limited by neutron damage on the reactor vessel. Further R&D activities should be performed to evaluate the maximum fast-neutron fluence and assess structural materials choice.

Table 1: Principal data of selected fast spectrum MSR designs.

Parameter	MSFR (7)	MCSFR (8)	REBUS-3700 (9)	MOSART (10)
Thermal power, MW	3,000	6,000	3,700	2,400
Fuel salt volume (in/out), m ³	18 (9/9)	38 (16/22)	55.6 (36.9/18.7)	49.05 (32.7/16.35)
Fertile salt volume (in/out), m ³	7.3 (7.3/0)	75 (55/22)	—	—
Fuel and fertile salt initial composition, mol%	LiF-ThF ₄ - ²³³ UF ₄ (77.5- 19.9-2.6) LiF-ThF ₄ (77.5-22.5)	NaCl-UCl ₃ - ²³⁹ PuCl ₃ (60-36-4) NaCl-UCl ₃ (60-40)	55mol%NaCl+ 45mol%(natU+ 16.7at.%TRU)Cl ₃	LiF-BeF ₂ - ThF ₄ -TRUF ₃ (69.72-27- 1.28)
Fuel cycle	Th/ ²³³ U	U/Pu	U/TRU	Th/ ²³³ U
Initial fissile inventory, t	7.726	9.400	18.061	9.637
Fissile/fertile salt temperature, K	973/973	1008/923	900	933

of long-term reactivity through the fertile material feed rate, avoiding the use of classic reactivity control devices (e.g., reactivity control rods, burnable poisons, soluble poisons). Overall, reactivity control for fast spectrum MSRs presents an engineering challenge and is outside the scope of this paper. Another feature of MSRs, delayed neutron precursor drift corresponding to its circulating liquid fuel, is very important for safety transient analysis but has a negligible effect on long-term depletion calculations and is not treated here (11).

2.2. Models description

In contrast with thermal MSRs, fast spectrum concepts do not have a channel or assembly structure but instead use a cylindrical or spherical vessel to contain the homogenized fuel mixture. Two-fluid systems might have a cylindrical (e.g., MSFR) or spherical (e.g., MCSFR) blanket with fertile salt to reduce neutron leakage and enhance fissile material breeding (Fig. 1). Details regarding the configuration of each reactor can be found in Refs. (7; 8; 9; 10). For two-fluid concepts, the 2D unit cell model contains a cylindrical fuel salt channel with a thin outer layer of fertile salt inside a square block of structural material (Hastelloy N). The unit cell model for the single-fluid REBUS-3700 has fuel salt and structural material only; the MOSART simplified model consists of a fuel salt square block with a graphite cylinder in the center to represent the 0.2 m graphite reflector needed to increase ²³³U breeding from thorium (17).

To show the performance of unit cell models for depletion simulations, high-fidelity full-core models were developed using the Monte Carlo code SERPENT2 (16 million neutron histories per state point) with the ENDF/B-VII.1 library (18; 19). For average unit cell models, the geometry and size are optimized to obtain a sufficiently accurate multiplication factor and neutron energy spectrum in a reasonable time using specific metrics:

1. Eigenvalue discrepancy between full-core and unit cell models less than 300 pcm[§] at beginning of life.
2. Pearson correlation coefficient r (Eq. 1) for neutron spectrum normalized by lethargy more than 0.995:

$$r = \frac{\sum_{i=1}^N (\Phi_i^f - \bar{\Phi}^f)(\Phi_i^u - \bar{\Phi}^u)}{\sqrt{\sum_{i=1}^N (\Phi_i^f - \bar{\Phi}^f)^2 \sum_{i=1}^N (\Phi_i^u - \bar{\Phi}^u)^2}} > 0.995 \quad (1)$$

Φ_i^f, Φ_i^u = the neutron flux for i^{th} energy group for full core and unit cell

$\bar{\Phi}^f, \bar{\Phi}^u$ = the neutron flux averaged over N energy groups for full core and unit cell

(2)

3. Approximation error δ (Eq. 3) in total neutron flux less than 3%:

$$\delta = \frac{\sum_{i=1}^N (\Phi_i^f - \Phi_i^u)}{\sum_{i=1}^N \Phi_i^f} \times 100\% < 3\% \quad (3)$$

Additionally, short-time[¶] depletion calculations without online reprocessing were conducted using SERPENT2 for both the full-core and the unit cell model. Next, the depleted mass approximation error for all isotopes was calculated to validate depletion calculation results. Finally, the approximation error for representative isotopes was plotted for each reactor type to demonstrate the viability of the unit cell model for depletion simulations. Reactor symmetry was leveraged to simplify the geometries to quarter-cell models. For this optimization, a 16×16 spatial mesh for the NEWT neutron transport calculation was used in SCALE/TRITON.

2.3. Fuel cycle performance metrics

The main objective of this work is to analyze fast MSR systems and fuel cycles in support of the Systems Analysis and Integration Campaign of the US Department of Energy, Office of Nuclear Energy (DOE-NE). DOE established an Evaluation and Screening Team (EST) consisting of national laboratory and industry experts in nuclear fuel cycles to develop the evaluation metrics for 40 representative nuclear fuel cycle options (i.e., Evaluation Groups). From the continuous reprocessing depletion simulations conducted for the four selected fast MSR designs, the following evaluation metrics were determined:

1. Natural uranium per energy generated (for MCSFR, REBUS-3700).
2. Natural thorium per energy generated (for MSFR, MOSART).
3. Mass of SNF+HLW^{||} disposed per energy generated.
4. Mass of DU+RU+RTh^{**} disposed per energy generated.

For fuel cycle metric computations, a few assumptions were made: (1) fission products are separated from the fuel salt and disposed of, (2) the remaining heavy metals are separated from the fuel salt and can be reused to start up additional reactors, and (3) the carrier salt can also be reused. Results are compared with metric data for respective Evaluation Groups using representative fuel cycle technologies from the Nuclear Fuel Cycle E&S study (20).

[§] 1 pcm = $10^{-5} \Delta k_{eff} / k_{eff}$.

[¶] 1140 days ≈ 3 years has been selected because of SERPENT2 burnup limitations for subcritical systems (if multiplication factor k_{eff} becomes too low the simulation is interrupted).

^{||} spent nuclear fuel (SNF)+high level waste (HLW).

^{**} depleted uranium (DU) + recovered uranium (RU) + recovered thorium (RTh).

3. RESULTS

This section presents calculation results, such as neutron flux spectrum for full-core and unit cell models, the depletion calculations key findings, and the E&S evaluation metrics.

3.1. Full-core and unit cell neutron spectra

Figures 2 and 3 show the neutron flux energy spectra for full-core 3D models obtained with SERPENT2 and simplified unit cell 2D models obtained with SCALE/TRITON. The calculated correlation coefficient (r) and approximation error (δ) in the total neutron spectrum for the unit cell model is indicated in the left upper corner of each plot. The most accurate approximation is obtained for the MOSART, while the MCSFR has the worst accuracy. The main reason for the discrepancies is the low-energy-group resolution at fast-neutron energies in the 238-group nuclear data library, which was developed for thermal reactors. This issue could be resolved by using problem-oriented nuclear data for fast reactors, but it is computationally expensive.

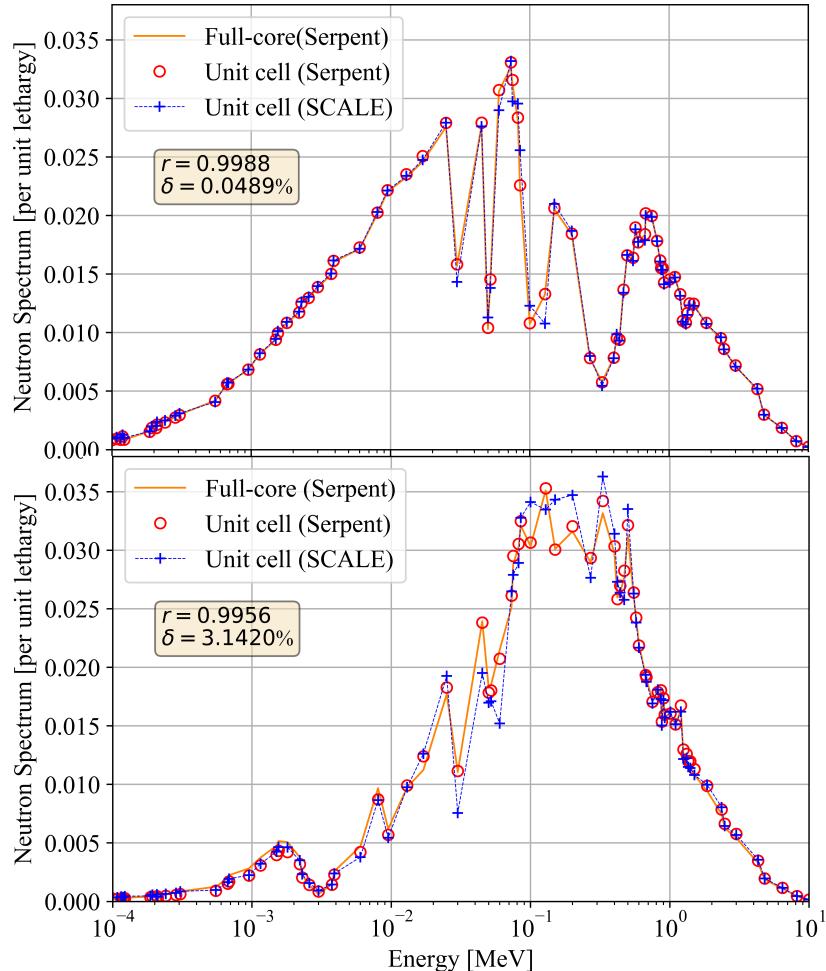


Figure 2: Neutron flux energy spectrum for full-core and unit cell models for two-fluid MSFR (top) and MCSFR (bottom).

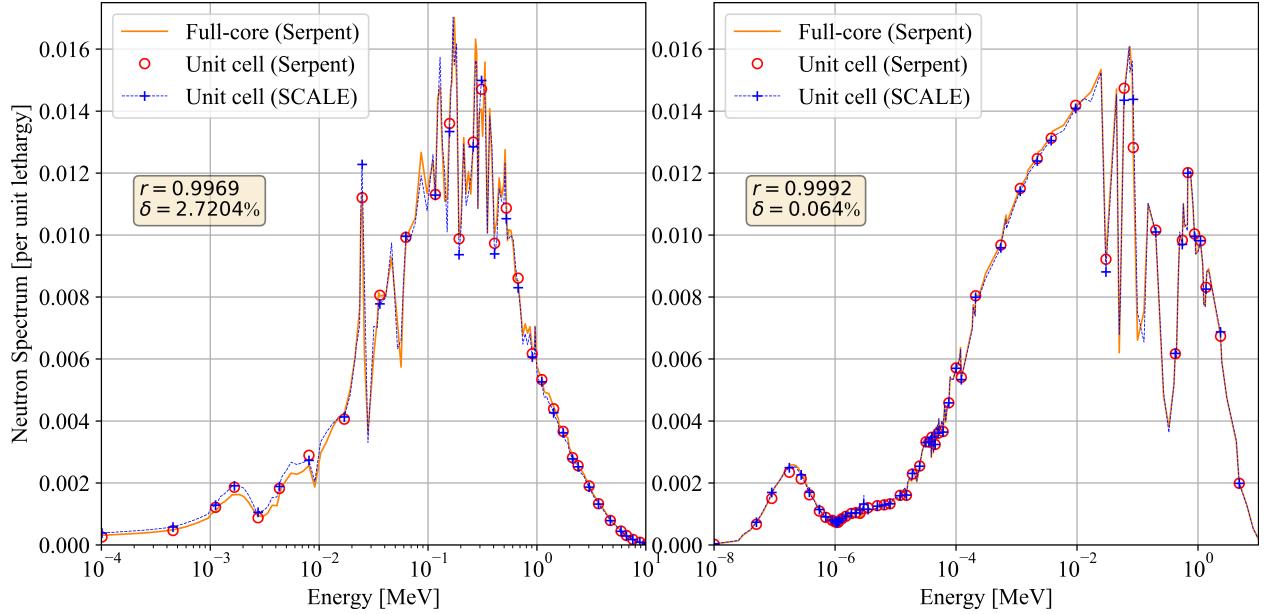


Figure 3: Neutron flux energy spectrum for full-core and unit cell models for single-fluid REBUS-3700 (left) and MOSART (right).

For all four reactor concepts, the neutron energy spectra obtained with the SERPENT Monte Carlo and SCALE/TRITON agree very well as in previous studies (21). Larger differences are observed in the intermediate energy ranges because of cross-section resonances for these energies and to the limited number of energy groups for SCALE/TRITON. Continuous-energy physics is expected to provide better results as it does not suffer from inaccurate cross-section representations in resonances and the fast energy range.

Finally, the unit cell approximation accuracy for depletion simulations was estimated using SERPENT2 burnup capabilities for the full-core and unit cell model of the MSFR. The relative error of unit cell approximation for important isotopes mass after 1140 EFPD (effective full-power days) of salt irradiation is less than 2% and increases during operation. Furthermore, the unit cell approximation has provided a speedup of **20 times** over the original full-core model. Further method optimization (e.g., using problem-oriented cross-section libraries optimized for fast reactors) is needed to achieve better accuracy.

3.2. Fuel cycle performance analysis

Figure 4 demonstrates that all reactors started with the same small excess of reactivity (≈ 2000 pcm) and remained critical throughout the 60-year operational lifetime. The thorium-fueled MSFR and MOSART show a significant reactivity drop during the first few years of operation because of ^{233}Pa accumulation; this is a strong neutron poison that β decays to ^{233}U with a half-life of 27.4 days. For

U/Pu concepts (MCSFR and REBUS), ^{239}Np β decays $\dagger\dagger$ to ^{239}Pu much faster ($\tau_{1/2} = 2.356$ days). Therefore, a lower fissile material feed rate is needed to compensate this breeding delay.

Figures 5 and 6 show the heavy metal inventory evolution for four selected fast MSR designs during 60 years of operation at a 100% power level. The two-fluid MSFR breeds ^{233}U from ^{232}Th in the driver (core) and in the blanket. Fissile material produced in the blanket is continuously removed and then rerouted in two directions: (1) to the driver to compensate fuel burnup and maintain reactor criticality and (2) to fissile material storage. Notably, a significant amount of ^{234}U was accumulated in the core and significantly deteriorates breeding performance because ^{234}U is a neutron absorber. Thus, to avoid this parasitic absorption, ^{233}Pa (which is a precursor for ^{234}U production) could be isolated from the core and allowed to decay to ^{233}U . This approach was suggested by ORNL for the thermal MSBR concept (5) and can also be employed for fast MSRs.

Similarly, the MCSFR breeds ^{239}Pu from ^{238}U and directly produces a considerable amount of nonfissile ^{240}Pu . The ^{240}Pu has a high absorption cross section, which degrades the neutron economy. The MCSFR at the end of 60-year operation cycle has approximately 28 t of fissile ^{239}Pu left in the storage and driver, and demonstrates the best among four considered designs with respect to doubling time: 17 years (e.g., during 60 years of operation the MCSFR produces enough ^{239}Pu to startup ≈ 3.5 additional reactors of this type).

The single-fluid MOSART is designed as an efficient LWR SNF transmuter (Figure 6). TRU from spent fuel is used as a fissile material (similarly to mixed oxide (MOX) fuel for LWRs) to breed new fissile ^{233}U from ^{232}Th . Notably, MOSART has a graphite reflector to shift the neutron spectrum to the intermediate range (Figure 3) because ^{233}U breeding is more efficient in the thermal neutron energy range. Moreover, total TRU inventory increases during operation to compensate for parasitic neutron absorption (by ^{240}Pu , ^{234}U , etc). Overall, over 60 years of operation MOSART consumed 43.68 t of TRU from LWR SNF and 26.7 t of ^{232}Th , while producing 66.4 GWe-year and having 2.3 t of fissile ^{233}U left in the core at the end of the lifetime.

Similarly to the MOSART, REBUS-3700 also uses TRU from LWR as a fuel but operates with the $^{238}\text{U}/^{239}\text{Pu}$ fuel cycle. This fast MSR concept maintained a nearly constant TRU and natural uranium inventory over 60 years without any issues related to neutron poison accumulation, which is possible because of the very hard neutron spectrum of this MSR concept (Figure 3). Notably, REBUS-3700 does not need continuous fissile material feed (only fertile material feed). Over 60 years of operation, REBUS-3700 would consume 85 t of natural uranium and 18 t of TRU as start-up fuel, while producing 102 GWe-year and having 10.5 t of fissile ^{239}Pu inventory at the end of the lifetime.

3.3. E&S Evaluation Metrics

Evaluation metrics were calculated using the E&S guide (20) to evaluate fast MSR fuel cycle performance and compare it with other designs (Table 2). Normalized per GWe-year, the MOSART and MSFR concepts required only 0.402 and 0.663 t of natural thorium, respectively. Natural uranium utilization per energy generated for the U/Pu cycle concepts MCSFR and REBUS is less

$\dagger\dagger$ ^{238}U captures a neutron to form ^{239}U , which almost immediately β decays to ^{239}Np .

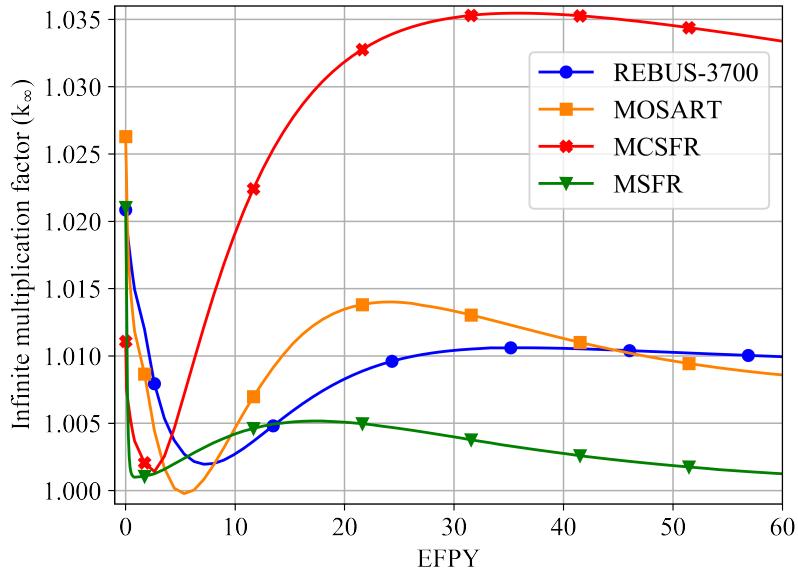


Figure 4: Infinite multiplication factor for four reactor designs during 60 years of operation.

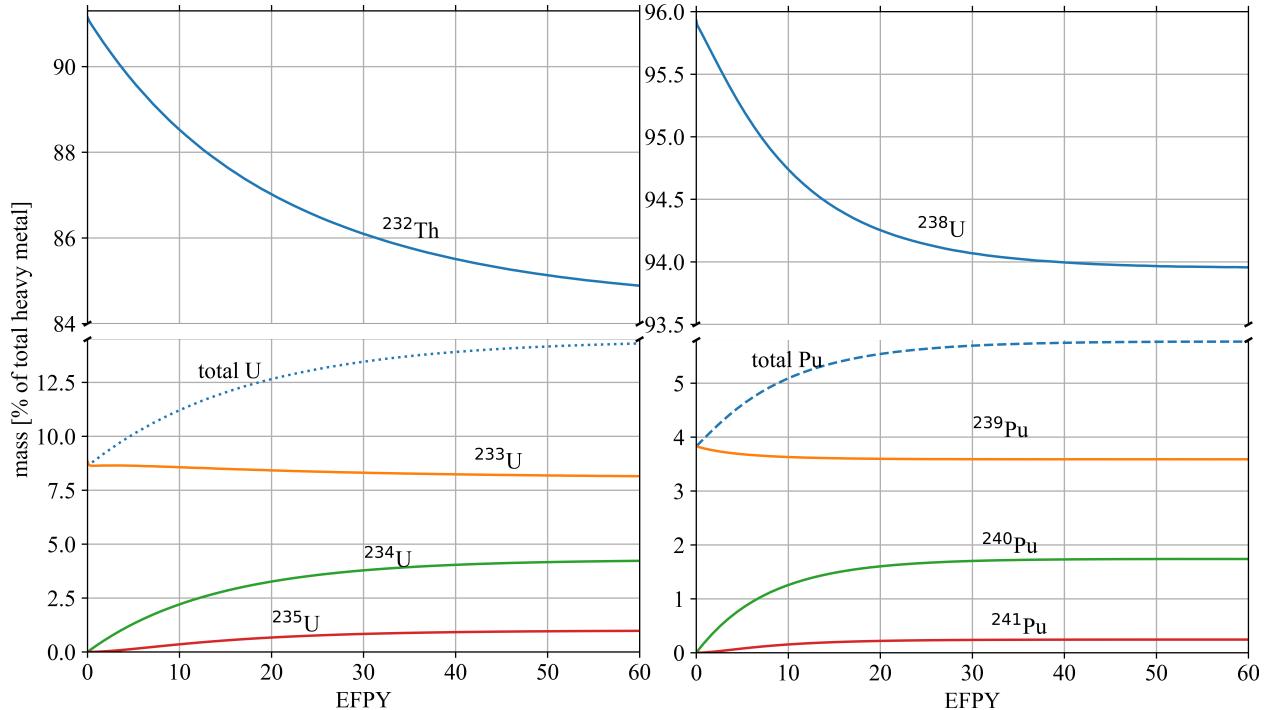


Figure 5: MSFR (left) and MCSFR (right) heavy metal isotopic salt content during operation calculated with the unit cell model (238-group transport).

than 0.9 t/GWe-year. The considered reactor designs were operated in a continuous recycle fuel cycle and did not dispose of DU, RU, or RTh.

Low natural resource consumption results in a total waste reduction (SNF+HLW) to less than 0.9

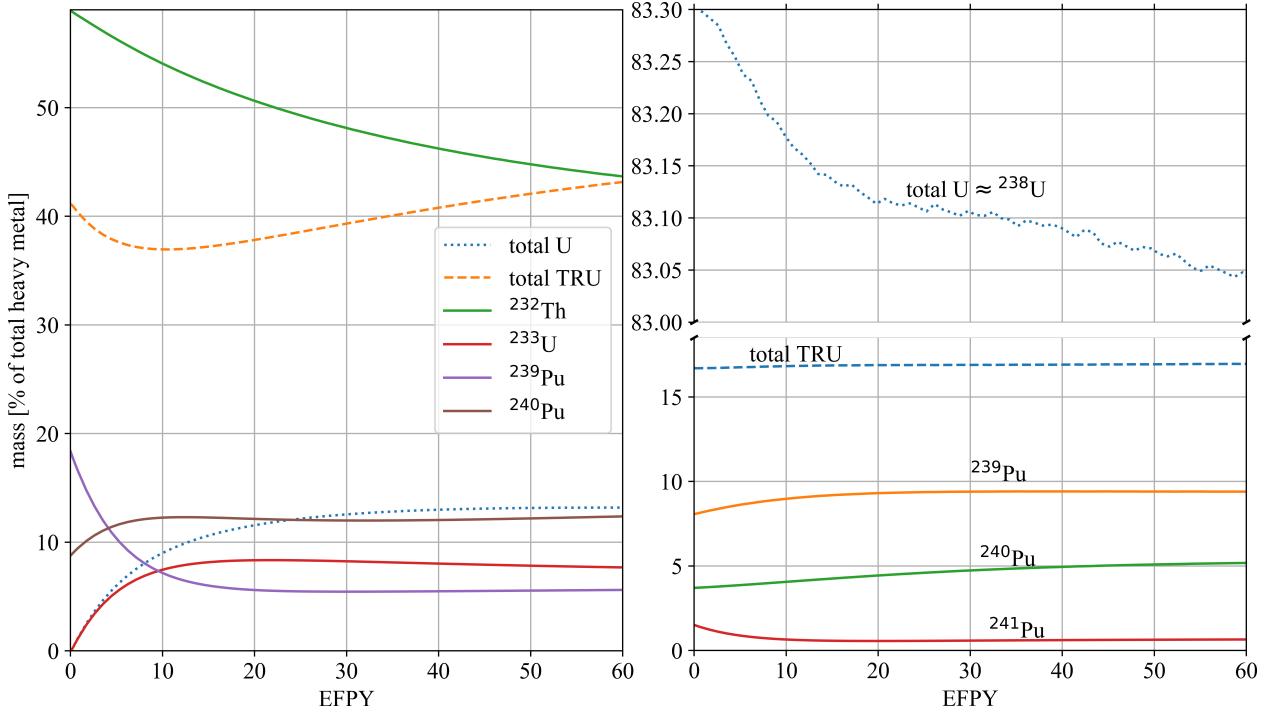


Figure 6: MOSART (left) and REBUS-3700 (right) heavy metal isotopic salt content during operation calculated with the unit cell model (238-group transport).

Table 2: The E&S evaluation metrics of selected fast spectrum MSR designs.

Parameter	MSFR	MCSFR	REBUS	MOSART
Evaluation Group	EG28	EG23	EG24	EG28
Natural U or Th Utilization, t/GWe-yr	0.663(Th)	0.973 (U)	0.834 (U)	0.402(Th)
Mass of SNF+HLW disposed, t/GWe-yr	0.866	0.894	0.813	0.820
Mass of DU+RU+RTh disposed, t/GWe-yr	0.0	0.0	0.0	0.0
Products from Reprocessing/Separation technology, t: RU/RTh/TRU/Fission Product (FP)	8.7/41.9/ 0.36/69.51	83.2/0.0/ 32.8/140.3	92.6/0/ 18.9/79.6	3.9/12.9/ 12.9/54.1

t/GWe-year. In addition, MOSART and REBUS propose using TRU separated from LWRs SNF as fissile material, reusing the SNF from 60 years of nuclear power generation. Notably, the lower natural resource utilization does not directly correlate with the mass of disposed waste because initial TRU from LWR SNF to fuel the MOSART and REBUS was considered as “free” resource and was not included in natural uranium/thorium mass. Overall, the fuel cycle performance of the considered MSR designs is consistent with fast spectrum continuous recycle fuel cycles evaluated under the E&S Study (EG23, EG24, EG28) (20).

4. CONCLUSIONS

The fuel cycle performance of four fast spectrum MSR designs was analyzed to determine key fuel cycle metrics. Full-core SERPENT Monte Carlo transport models and 2D unit cell SCALE/TRITON deterministic transport models were created to demonstrate the viability of using simplified unit cell models for long-term (i.e., 60 years) fuel cycle simulations to reduce computational burdens. Comparison between full-core and unit cell fluxes shows a relative error of less than 3.2% and a correlation coefficient of 0.9956, while the unit cell geometry approximation has provided a $20\times$ speedup.

Unit cell depletion simulations with continuous fission product removal and constant fertile/fissile material feeds show that with a startup infinite multiplication factor of approximately 1.02, all concepts remain critical during 60 years of operation. Natural uranium or thorium utilization varies from 0.402 t/GWe-yr for the thorium-fueled MOSART to 0.973 MTU/GWe-yr for the U/Pu MCSFR. SNF+HLW generation normalized per GWe-yr for all four designs is consistent with fast spectrum fuel cycle technologies.

The unit cell time-dependent analyses that were performed predict the fuel cycle performance for fast spectrum MSRs concepts and provide direction for future performance studies and design improvement. Additional validation of SCALE/TRITON Alpha against another continuous reprocessing code (e.g., SERPENT2) and/or batch-wise Python package SaltProc (22) is required to provide more confidence in the results.

Continued research into fast MSRs could progress in a number of different directions. First and foremost, efforts should be made to simulate the MSFR with additional protactinium isolation system which enhance ^{233}U breeding for two startup fissile compositions: (1) TRU to evaluate the MSFR performance as a waste burner; (2) new enriched uranium because it promises better fuel cycle performance. Secondly, the MCSFR design might be optimized to operate with startup composition based on enriched uranium instead of plutonium. An additional area to explore is the accident safety analysis, which may benefit from the deployment of a multi-physics model of investigated fast MSR designs with a coupled neutronics/thermal-hydraulics code, such as Moltres (23).

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