A NEW APPROACH FOR MODELING AND ANALYSIS OF MOLTEN SALT REACTORS USING SCALE

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ABSTRACT

The Office of Fuel Cycle Technologies (FCT) of the DOE Office of Nuclear Energy is performing an evaluation and screening of potential fuel cycle options to provide information that can support future research and development decisions based on the more promising fuel cycle options. [1] A comprehensive set of fuel cycle options are put into evaluation groups based on physics and fuel cycle characteristics. Representative options for each group are then evaluated to provide the quantitative information needed to support the valuation of criteria and metrics used for the study. Included in this set of representative options are Molten Salt Reactors (MSRs), the analysis of which requires several capabilities that are not adequately supported by the current version of SCALE or other neutronics depletion software packages (e.g., continuous online feed and removal of materials). A new analysis approach was developed for MSR analysis using SCALE by taking user-specified MSR parameters and performing a series of SCALE/TRITON calculations to determine the resulting equilibrium operating conditions. This paper provides a detailed description of the new analysis approach, including the modeling equations and radiation transport models used. Results for an MSR fuel cycle option of interest are also provided to demonstrate the application to a relevant problem. The current implementation is through a utility code that uses the two-dimensional (2D) TRITON depletion sequence in SCALE 6.1 but could be readily adapted to three-dimensional (3D) TRITON depletion sequences or other versions of SCALE.

Key Words: Molten salt reactor, fuel cycle options, thorium, separations, reactor analysis

1. FOREWORD

The Office of Fuel Cycle Technologies (FCT) of the DOE Office of Nuclear Energy is performing an evaluation and screening of potential fuel cycle options to provide information that can support future research and development decisions based on the more promising fuel cycle options. [1] This process includes developing a comprehensive set of fuel cycle options that are categorized into evaluation groups based on physics and fuel cycle characteristics. For each group, representative options are evaluated to provide the quantitative information needed to support the valuation of the criteria and metrics used for the study. This involves developing technical information for the representative options as well as determining material mass flows and characteristics, such as resource requirements and waste information. This set of representative options to be evaluated includes Molten Salt Reactors (MSRs) that involve the use of thorium and three separations levels: full separations involving multiple actinides and fission products, limited separations without protactinium separation, and "once-through" operation without any active separations processes.

2. INTRODUCTION

A thermal spectrum molten salt reactor (MSR) includes a fuel-bearing molten salt (e.g., LiF-BeF$_2$-UF$_4$-ThF$_4$) that simultaneously functions as both fuel and coolant, graphite moderator, and a low-pressure primary system. [2, 3] The primary system is low pressure and operates at temperatures between the salt's melting and boiling points. The salt flows through a heat exchanger, transferring nuclear heat either directly to the secondary loop or through an intermediate loop to the secondary loop.   MSRs can be operated with continuous online fuel addition and salt processing to remove fission products and actinides. Typical MSR concepts have conversion/breeding ratios ranging from 0.9–1.05, may require feed of fertile (e.g., thorium) or fissile (e.g., enriched uranium) materials or both, and have a range of possible treatment and reprocessing options that removes various materials from the fuel salt. Past MSR designs had power ratings ranging from 500 MW$_{th}$ to 2250 MW$_{th}$, though smaller or larger versions are also possible.

Reactor analysis work performed at ORNL often utilizes radiation transport and isotopic depletion calculations performed with the SCALE computer code system. [4] More specifically, the SCALE/TRITON depletion sequence couples NEWT as a two-dimensional (2D) discrete-ordinates radiation transport solver to ORIGEN for isotopic depletion and decay calculations, using ENDF/B-VII nuclear data. [5, 6] These capabilities of SCALE have seen extensive use and validation with light water reactors (LWRs) as well as high-temperature gas reactors (HTGRs) and other reactor designs.

As a consequence of the characteristics noted above and their differences relative to LWRs, MSR analysis requires several functions and features that are not supported in the current version of SCALE. MSR unique features include a flowing fuel that provides constant mixing and some power production in fuel outside of the traditional "core" (moderated) region. Modeling capabilities required for MSR analysis include the continuous online feed and removal of various materials at rates that depend upon the species involved and desired operation mode.   ORIGEN [7] provides the capability of continuous and/or batch feed and removal of different materials, as well as other desired features such as mixing different material compositions together, but these capabilities are not available within SCALE/TRITON. These shortcomings have also been noted elsewhere for other codes outside of SCALE. [8]

In order to use SCALE for simulating MSRs, an approach was developed at ORNL that takes user-specified MSR parameters and performs a series of SCALE/TRITON calculations to determine the resulting equilibrium operating conditions. Details of the approach, including the modeling equations used and how they were implemented, are provided below. The current implementation of this work uses the 2D TRITON depletion sequence in SCALE 6.1 but could be readily adapted to three-dimensional (3D) TRITON depletion sequences or other versions of SCALE. The primary purpose of this utility code is to take initial parameters and simulate chemical processing and power operations of the system over a period of time long enough to establish an equilibrium state, which is essential for evaluating fuel cycle options.

3. MODELING APPROACH

Analysis of the thorium-fueled MSR systems of interest in this work requires salt processing calculations that must provide several important possible functions related to material feed and removal: possible direct discard of fuel salt, salt treatment processes to remove fission products that would otherwise not stay in solution such as gases and noble metals, MSR separations work that actively separates or extracts from the fuel salt species such as protactinium (Pa) or rare earth element (REE) fission products (FP), and thorium addition. Protactinium separation allows Pa to decay to U-233 outside the reactor, which increases the efficiency of breeding U-233 fissile material from thorium by minimizing parasitic neutron absorption reactions. As a result of these material flow processes, some fuel salt properties [e.g., heavy metal (HM) density] need to be adjusted to account for materials that were added or removed. These processes could be applied in sequence or in parallel depending upon their implementation; at present, they are applied sequentially with separations of U and Pa first, followed by salt treatment and separations, and then any desired salt discard.

All of the salt processing functions discussed above could be performed as either batchwise or continuous (online) processes, depending upon the operational desires and needs. SCALE/TRITON is designed to model batch-fueled reactors with solid fuel (e.g., LWRs) and does not provide access to the continuous material feed and removal capabilities or the specification of process separations within ORIGEN. In addition, the current implementation of continuous feed and removal in ORIGEN does not track the feed and discharge materials that are important to understand feed and waste streams. One important consequence of these limitations is that short depletion time steps are required in order to minimize the impact of no feed or removal during the time step; this requires a large number of depletion time steps to simulate longer periods of time, which makes long depletion calculations computationally expensive. This situation is far from ideal but offers the best trade-off at the present time for providing the capability of meeting all functional requirements while also obtaining reasonably accurate results.

3.1. Salt Treatment and Separations

Salt processing calculations for material removal during salt treatment and MSR separations require user-specified elemental removal rates, for which definitions and representative values may be found in Molten Salt Breeder Reactor (MSBR) reports. [2, 3] Elemental removal rates are given in the form of "cycle time", defined as the amount of time to completely remove an element from the salt, rather than separations efficiencies. [2, 3] This is due to the fact that separations efficiencies for each process are not meaningful because many of the materials are returned to the reactor after processing. Removal fractions for each element of interest are calculated according to

$$\mathrm{RF}_j = \frac{t_{depl}}{t_j^{cycle}} \, , \tag{1}$$

where

RF_j is the removal fraction for element j,

t_{depl} is the depletion time step length [days], and

t_j^{cycle} is the effective cycle time for element j [days].

The method of calculating removal fractions has been chosen to match the definition that 100% of element j would be removed ($RF_j = 1$) during a time period (t_{depl}) equal to the cycle time for that element (t_j^{cycle}). Given the form of Eqn. (1), elemental removal fractions will vary directly with the duration of depletion time step and inversely with cycle times. Table I provides a practical illustration of this relationship, showing representative cycle times [2] and their corresponding removal fractions for a depletion time step of 3 days, which matches the cycle time for Pa. Removal fractions greater than one are nonphysical; they are an artifact of that element's cycle time being less than the length of the depletion step. The current implementation sets any removal fraction with a value greater than one to a value of one.

Table I. Cycle times used for material removal in salt treatment and separations processes

Processing Type	Processing Group	Elements	Cycle time	Removal Fraction (t_{depl} = 3 days)
Salt Treatment	Volatile Gases	Xe, Kr	20 sec	1.3×10^4
	Noble Metals	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te	20 sec	1.3×10^4
	Seminoble Metals	Zr, Cd, In, Sn	200 days	0.015
	Volatile Fluorides	Br, I	60 days	0.05
Salt Processing	Protactinium	Pa	3 days	1.0
	Rare Earth Elements	Y, La, Ce, Pr, Nd, Pm, Sm, Gd	50 days	0.06
		Eu	500 days	0.006
	Discard	Rb, Sr, Cs, Ba	3435 days	8.73×10^{-4}

Modeling the salt treatment and MSR separations as batch processes between depletion time steps fails to fully capture the reality of a continuous online removal process; however, any error introduced to this process can be minimized by using small depletion steps and should generally yield eigenvalues lower than would be seen with a more realistic continuous removal model given the elements currently being removed. As noted above, for current MSR work ORNL has chosen to use a depletion time step of 3 days in order to avoid exceeding the Pa cycle time.

As shown below, elemental masses are calculated using the summation of the isotopic mass for each isotope i that belongs to element j. Using these isotopic and elemental masses, along with isotopic number densities and the removal fractions determined above, isotopic and elemental masses for the resulting fission product waste streams and number densities for fission products remaining in the fuel salt are calculated as

$$m_i(t_n) = \frac{N_i(t_n)M_i V_{salt}}{(10^{-24}\ cm^2/b)N_A} \tag{2}$$

$$m_i^{rem}(t_n) = RF_{j \ni i} m_i(t_n) \tag{3}$$

$$m_j^{rem}(t_n) = \sum_{i \in j} m_i^{rem}(t_n) \tag{4}$$

$$N_i'(t_n) = N_i(t_n)\left(1 - RF_{j \ni i}\right) \tag{5}$$

$$m_{tot}^{rem}(t_n) = \sum_j m_j^{rem}(t_n)\ , \tag{6}$$

where

$m_i(t_n)$	is the mass of isotope i at time t_n [g],
$N_i(t_n)$	is the atomic number density of isotope i at time t_n [a/b-cm],
M_i	is the molar mass of isotope i [g/mol],
V_{salt}	is the total volume of fuel salt [m^3],
N_A	is the Avogadro constant (6.022×10^{23}) [atoms/mol],
$m_i^{rem}(t_n)$	is the mass of isotope i removed at time t_n [g],
$RF_{j \ni i}$	is the removal fraction for element j to which isotope i belongs,
$m_j^{rem}(t_n)$	is the mass of element j removed at time t_n [g],
$N_i'(t_n)$	is the atomic number density of isotope i at time t_n after salt treatment and processing [a/b-cm], and
$m_{tot}^{discard}(t_n)$	is the total mass of fuel salt discarded at time t_n [g].

Separation of protactinium and U-233 content in the fuel salt are somewhat more complicated due to the U-233 produced by Pa decay adding to an existing amount of U-233 in the fuel salt. The resultant masses and number densities for Pa and U-233 are modeled according to

$$m_{Pa}^{sep}(t_n) = \frac{RF_{Pa}N_{Pa233}(t_n)M_{Pa233}V_{salt}}{(10^{-24}\ cm^2/b)\ N_A} \tag{7}$$

$$N_{Pa233}'(t_n) = N_{Pa233}(t_n)(1 - RF_{Pa}) \tag{8}$$

$$N_{U233}(t_{n+1}) = N_{U233}(t_n) + RF_{Pa}N_{Pa233}(t_n)\ , \tag{9}$$

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where

$$m_{Pa}^{sep}(t_n)$$ is the mass of Pa separated at time t_n [g],
$$N_{U233}(t_{n+1})$$ is the atomic number density of U-233 at time t_{n+1} [a/b-cm],

and all other terms have been defined above or follow the form of previously defined terms.

3.2. Salt Discard

Direct discard of fuel salt removes the specified volume fraction of salt, including heavy metals, and creates this as a waste stream. Many MSR concepts don't discard any fuel salt in this manner, due to reduced resource utilization and the disposal of useful fissile materials, but some potential fuel cycle options might consider this feature to eliminate the need for separations. New isotopic number densities, elemental masses discarded, and total mass of discarded salt are calculated according to

$$m_j^{discard}(t_n) = DF \sum_{i \in j} m_i(t_n) \tag{10}$$

$$N_i''(t_n) = N_i'(t_n)(1 - DF) \tag{11}$$

$$N_i(t_{n+1}) = N_i''(t_n) \tag{12}$$

$$m_{tot}^{discard}(t_n) = \sum_j m_j^{discard}(t_n) \ ,$$

where

$$m_j^{discard}(t_n)$$ is the mass of element j discarded at time t_n [g],
DF is the salt discard fraction,
$$N_i''(t_n)$$ is the atomic number density of isotope i at time t_n after salt discard [a/b-cm],
$$N_i(t_{n+1})$$ is the atomic number density of isotope i at time t_{n+1} [a/b-cm],

and all other terms have been defined above or follow the form of previously defined term. As previously stated, it should be noted that any salt discard occurs after that stream of salt has undergone U and Pa separations and then salt treatment and separations. If salt discard is not desired, the elemental and total discard masses are set to 0 and values for $N_i''(t_n)$ and $N_i(t_{n+1})$ can calculated accordingly.

3.3. Thorium Feed

In addition to the material removal processes, the previously defined fuel cycle options of interest require the feed of additional thorium to act as makeup material and maintain an equilibrium state. While a precise feed rate could be specified, the current methodology simply conserves Th number density during the entire depletion. This approach implies that the Th loading for the first depletion step is the value at desired equilibrium state. The time-dependent thorium feed mass and number densities are calculated as

$$m_{Th232}^{feed}(t_n) = \frac{\left(N_{Th232}(0) - N''_{i\,Th232}(t_n)\right)M_{Th232}V_{salt}}{(10^{-24}\ cm^2/b)\ N_A} \tag{13}$$

$$N_{Th232}(t_{n+1}) = N_{Th232}^0\ , \tag{14}$$

where

$m_{Th232}^{feed}(t_n)$ is the mass of thorium feed required at time t_n [g],
$N_{Th232}(0)$ is the initial Th number density (t=0) [a/b-cm],

and all other terms have been defined or follow the form of previously defined terms.

3.4. Limitations

The methods described above for material feed and removal processes result in several limitations. The most important computational limitation is that short depletion time steps must be used to ensure reasonable removal fractions and approximate a continuous process. In addition, the current model does not check the validity or feasibility of salt density and composition; a more robust approach would ensure realistic salt chemistry by including calculations that ensure solubility limits are not exceeded by any elements (or groups of elements) and perform chemistry control of the salt by ensuring proper makeup or removal of base salt constituents (e.g., F, Li, and Be). These salt chemistry checks would require significantly more complicated calculations and might also require new experimental data for solubility limits and chemistry control methods.

4. IMPLEMENTATION

The new methodology described above has been implemented in this current work as a utility code that wraps around SCALE/TRITON; this utility code provides limited use capability and is not intended to be incorporated into any future versions of SCALE. The utility code in effect performs "restart" SCALE/TRITON calculations, renormalizing properly for the HM content of the system in order to ensure proper power and flux levels and subsequent isotopics. Figure 1 provides a flowchart outlining the process implemented, with various sections explained in more detail below.
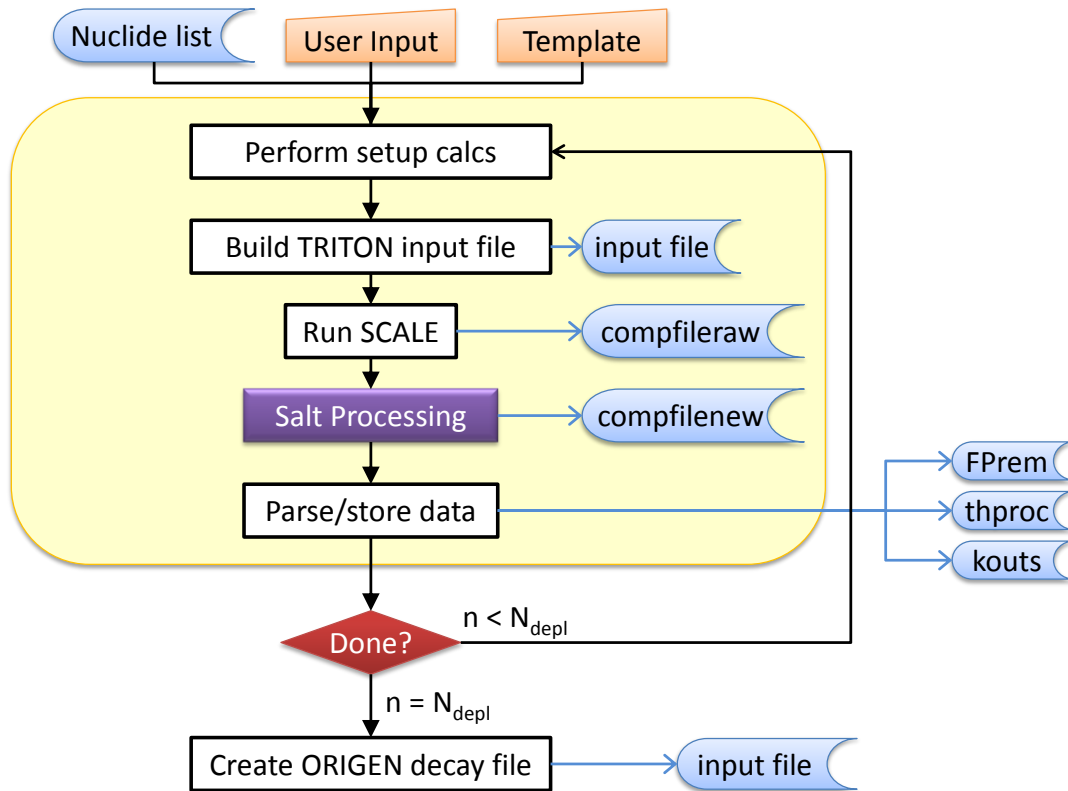
Figure 1. A conceptual flowchart outlining the process implemented

4.1. Setup Calculations and Executing SCALE

The required input files consist of a main Perl script file named according to the user's wishes (e.g., `MSBR.pl`) and an external file that contains lists of nuclides broken down in different ways and formats (`nuclidelists.pl`). The main script contains an input section at the top where the user specifies MSR parameters and salt processing options. MSR parameters include power level, temperatures, initial fuel salt composition, densities, volumes, and geometry parameters for the 2D unit cell used for radiation transport calculations. Salt processing options include parameters governing any desired salt discard and element-specific cycle times for both salt treatment and MSR separations work. The main script also contains a template input file for SCALE/TRITON depletion calculations. Figure 2 shows the geometry for the unit cell calculation that corresponds to this template input file.
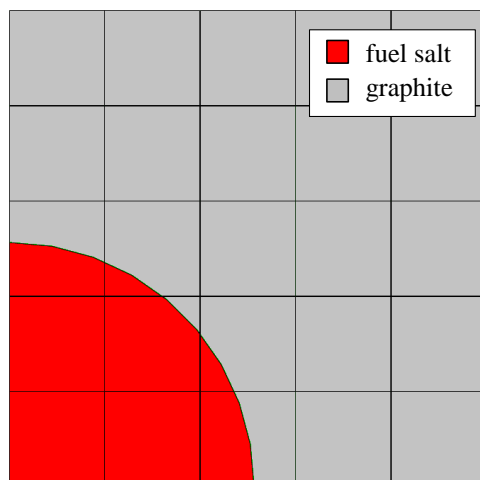
Figure 2. MSR unit cell geometry used for 2D SCALE/TRITON depletion

As seen above, the MSR unit cell modeled by this Perl script is a graphite block with a single flow channel at its center that contains fuel salt. The symmetry in the system is used to simplify the problem into one-quarter of the unit cell geometry, which is subsequently broken into a 5×5 spatial grid for the NEWT neutron transport calculations in SCALE/TRITON. The 5×5 spatial grid was chosen to provide a balance of speed and accuracy after evaluating multiple mesh options. This analysis uses an MSR concept based on square-pitch flow channels; however, the same analysis can be performed on triangular-pitch flow channels or other more complex geometries.

Setup calculations are performed to build an initial SCALE/TRITON depletion calculation input (e.g., `msbr_triton_depl1.inp`). These setup calculations include initial mass of HM, specific power in units of MW$_{th}$ per metric ton (MT) of HM, and other variables needed for internal functions. After building the initial input file, SCALE/TRITON is executed for that time step. Upon completion of the depletion calculation, SCALE writes an output file (e.g., `msbr_triton_depl1.out`) containing results from NEWT and ORIGEN calculations and also produces a file (e.g., `MSBRfuelcmp1raw.txt`) with the raw isotopic composition of the fuel salt at the end of the depletion step.

4.2. Salt Processing

Salt processing calculations start with the SCALE/TRITON output file and the raw material composition file for a given depletion time step and then need to model several important possible functions for an MSR system related to material feed and removal: possible direct discard of fuel salt, salt treatment and MSR separations processes, and thorium addition. Section 3 provides detailed descriptions of the methodology used for modeling these material flow processes. As noted above, real MSR systems could perform these salt processing functions as batchwise or continuous processes, but the methodology used in this paper requires batchwise modeling and approximates a continuous process by using sufficiently small time steps.

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Salt processing begins with salt treatment and MSR separations calculations using the raw fuel salt composition provided by SCALE/TRITON as a starting point and removal fractions based upon user-specified depletion step lengths and elemental cycle times (Section 3.1). If salt discard is requested, those calculations are performed using the number densities after treatment and separations. Lastly, thorium feed calculations are performed.

The physical reality of how the salt discard, salt treatment, and MSR separations processes would likely be sequenced directly interacts with the order and manner in which their models should be applied. If future methods enable continuous online salt treatment and separations, the order in which different processes are applied could be somewhat mitigated in that parallel processing would be used for some operations instead of fully sequential processing. It is also worth noting that the salt treatment described in this document uses passive processes to remove species that might otherwise not stay in solution, such as gases and noble metals. This can be thought of as somewhat analogous to coolant chemistry processes such as lithium hydroxide control in LWRs, in that it addresses coolant chemistry issues related to removing gases from the salt and controlling where solubility-limited species will plate out, but it also provides a substantial benefit to improved reactor performance and might be optimized for this purpose.

Having determined all of the masses for material feed and removal and the resultant isotopic number densities in the fuel salt at the beginning of the next depletion step, a new composition file (e.g., `MSBRfuelcmp1.txt`) is stored for the next depletion step.

4.3. Output Parsing and Data Storage

The steps described in Sections 4.1 and 4.2 include a SCALE/TRITON depletion calculation for a single time step, the calculation of numerous material feed and removal terms, and writing a fuel salt composition for the next depletion step. Before proceeding to the next depletion step, some parsing and data storage occurs.

First, the output file is parsed to collect the eigenvalues calculated for the MSR and append these values to a single file (e.g., `MSBRkouts.txt`) that stores all eigenvalues calculated during the full depletion loop. Next, isotopic masses from material removal processes are normalized to the amount of time in the depletion step in order to yield mass flow rates (in units of grams per second) and are stored to files; isotopic mass flow rates from salt discard are written to one file (e.g., `MSBRdiscarded1.txt`), while salt treatment and separations isotopic mass flow rates are written to another file (e.g., `MSBRremoved1.txt`). Integrated mass flow rates for thorium feed (Equation (12)), Pa separated from the fuel salt (Equation (9)), total fission products removed from the salt by treatment and separations (Equation (8)), and salt discarded (Equation (3)) are all appended to a file (e.g., `MSBRthorproc.txt`) that stores all integral mass flow rates calculated during the full depletion loop.

4.4. Loop Calculations

Upon successfully writing the data files described above, all calculations for the current depletion step have been completed. This establishes a loop of steps performed for each depletion step, shown in Figure 1 as a yellow box; upon completion of calculations and data

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storage for a given depletions step, the utility code returns to the top of the loop and starts the next depletion step by performing setup calculations using the new composition file (after material removal and addition) along with user-specified parameters and the template input file. This loop is repeated until for the user-specified total number of depletion steps.

4.5. Decay Heat Calculation Setup

After completing all desired depletion steps, the final composition file is used to create an ORIGEN input file for calculating decay heat (e.g., `MSBREOLdecay.inp`), which does not run automatically but can be easily executed if desired. Decay heat calculations are a key aspect of evaluating nuclear fuel cycle options. If needed, other parameters (e.g., activity and radiotoxicity) could easily be added to this stand-alone ORIGEN input file in the future.

5. RESULTS

Calculations performed to conduct a parametric analysis of a thorium-fueled MSR system with full separations work and determine key fuel cycle parameters demonstrate the application of this new method to a relevant problem.

The Molten Salt Breeder Reactor (MSBR) project provides the basis for these current calculations. [2, 3] The MSBR power level of 2250 MW_{th} and total salt volume of 48.7 m^3 are maintained, but the MSBR two-zone design is replaced by a single fuel zone design in this model and analysis. The unit cell geometry consists of a graphite block with a fuel salt flow channel in the center. The flow channel radius is chosen such that the volume fraction of molten salt in the unit cell (20.6%) represents the volume-weighted average of salt volume fractions in Zone I (13%) and Zone II (37%) of the MSBR. [2] Optimization work could improve this molten salt unit cell, but this serves as a representative starting point suitable for the level of accuracy required in this study. Depletion time steps in SCALE/TRITON were set to 3 days. The fuel salt temperature was assumed to be 700°C and the graphite block temperature was assumed to be 900°C, based upon MSBR analyses of gamma heating in the graphite blocks and subsequent cooling of the graphite blocks by the molten salt. The fuel salt composition starts off as 71.8 LiF – 16 BeF_2 – 12 ThF_4 – 0.2 UF_4, roughly the same salt composition as was used for the MSBR design [2]; at 700°C, this yields a density of 3.28 g/cm^3 using the salt density data generated during the MSBR project. [9, 10] Salt processing options included no salt discard and treatment and separations processes that used cycle times specified in Table I; active separations work included the separation of Pa.

The fuel cycle option of interest represents a single-stage fuel cycle option that utilizes an MSR with full continuous recycling. The objective of this fuel cycle option is self-sustained operation in a critical thermal neutron spectrum system with equilibrium requirements of no enrichment and only a thorium feed. Self-sustaining operation requires the conversion of thorium to fissile uranium to be in balance with the rate at which fission consumes uranium.

Calculations established near-equilibrium conditions after 20 years of operation based upon several parameters including eigenvalues, mass flow rates, and isotopic number densities in the fuel salt. Figure 3 shows the infinite multiplication factor converging to a steady equilibrium

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value of 1.022. Table II summarizes nominal equilibrium results for this scenario. These results, which include adequate equilibrium reactivity and conversion ratio as well as reasonable mass flow rates, indicate that a thorium-fueled MSR with full separations offers a viable fuel cycle option.
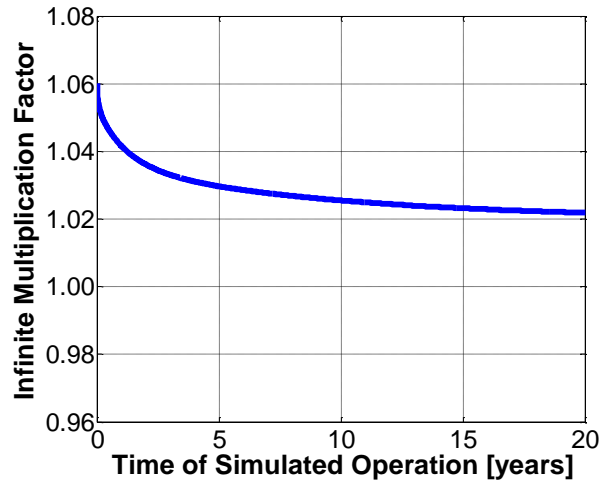
Figure 3. Infinite multiplication factor converging to equilibrium after 20 years of simulated operation for a Th-fueled MSR with full separations

Table II. Key equilibrium fuel cycle parameters for a Th-fueled MSR with full separations

Parameter	Value	Units
Conversion Ratio	1.001	-
Th Feed	926.0	kg/yr
Fissile Mass in Salt	1303.7	kg
Th Mass in Salt	69737.2	kg

6. CONCLUSIONS

A new methodology developed at ORNL enables the analysis of MSR systems using the SCALE computer code system in order to support current DOE fuel cycle work. This methodology incorporates user-specified parameters and models for several material feed and removal processes and performs a series of SCALE/TRITON calculations to establish equilibrium operating conditions for the desired concept and scenario. Results for an MSR fuel cycle option of interest demonstrate the applicability of this new method to a relevant problem and confirm that a thorium-fueled MSR with full separations offers a viable fuel cycle option. The current implementation of this work, a utility code that will not be incorporated into SCALE, uses the 2D TRITON depletion sequence in SCALE 6.1 but could be readily adapted to 3D TRITON depletion sequences or other versions of SCALE.

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