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INTRODUCTION

Recent developments in the commercial reactor sector have accelerated the need for modeling and simulation capabilities for design and safety analyses for the upcoming deployment of advanced experimental reactors. As part of this quicker timeline there is an active effort to utilize the Department of Energy's (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) codes to support near-term deployment.

One such effort funded by the DOE's Molten Salt Reactor (MSR) Campaign has focused on applying NEAMS codes to model liquid-fueled reactors, which will either have an active off-gas system or at the very least an off-gas plenum where volatile fission gases (e.g., Xe & Kr) and other insoluble fission products will accumulate during reactor burnup. This spatial mobility of fuel and fission products complicates depletion and source term calculations, especially when certain insoluble isotopes migrate to an off-gas region separated from the main fuel flow through the core of the reactor.

This is truly a multiphysics process where neutronics, chemistry, thermal-hydraulics, and mass transfer/reaction kinetics drive the volatilization of chemical species and transport through the reactor system. To accurately model this effect for future experimental reactor demonstrations and for the design of chemical species measurement devices used in future off-gas systems, a multiphysics informed approach using the NEAMS code suite is desired.

This work reports the first step in such an analysis, i.e., the implementation and verification of isotope removal capability for multi-region MSR depletion in Griffin the NEAMS multi-fidelity neutronics code [1]. This summary highlights previous work done for this problem and the importance of a multiphysics approach to solving this problem moving forward and why it is well suited for the NEAMS code suite utilizing the MOOSE framework [2]. Next, a brief review of the Bateman equations including the isotopic removal term is provided which describes how this removal term is added to the CRAM solver in Griffin. An example depletion problem of a homogenous medium MSR core that is used in the confirmatory studies is described. The results of confirmatory studies comparing Griffin depletion with isotope extraction versus SERPENT [3] for the example problem are showcased. Lastly, ongoing work incorporating the chemical equilibrium of the fuel-salt to determine the volatilization of species is also discussed.

PREVIOUS WORK AND THE VALUE OF NEAMS MULTIPHYSICS

To the best of the authors knowledge, the two primary reactor analysis codes or code suites that can model multi-region depletion with isotope removal are the Monte Carlo code SERPENT and the reactor analysis code suite SCALE (ChemTriton) [3,4]. The former is computationally expensive, and the latter lacks the ability to easily integrate with a multiphysics framework necessary to accurately resolve the spatial and time dependent extraction rates expected in these systems. This is where the NEAMS code suite, specifically the multiscale and multiphysics MOOSE framework can offer a less computationally expensive but still rigorously informed solution [2].

Utilizing NEAMS codes, a multiphysics approach can be modeled where the removal rates of isotopes can be accurately determined at each time step by incorporating the chemical equilibrium, thermal-hydraulics, and mass transfer/reaction kinetics occurring in the reactor system. Specifically, tools like Griffin for neutronics and depletion, Thermochimica [5] or Yellowjacket [6] for chemical equilibrium calculations, Pronghorn [7] or SAM [8] for thermal-hydraulics, and Mole [6] or the Chemical Reactions module for mass transfer/reaction kinetic – most built on the MOOSE framework – are ideal for such analysis [4].

BATEMAN EQUATIONS WITH ISOTOPIC EXTRACTION

The explicit methodology for solving the Bateman equations with concentration-dependent feed/removal rates over multiple regions is shown in [9] and briefly reiterated here. The specifics of the detailed terms in the Bateman equations are not explicitly listed here and instead the discussion will pick up with the matrix form of the problem. The reader is directed to [9] for a more detailed treatise of this problem.

The Bateman equations, a system of ordinary differential equations, for all nuclides in the reactor system and in the off-gas system can be formulated in a standard depletion matrix solve as follows in Equation 1.

$$\frac{d\boldsymbol{n_{PL}}}{dt} = \boldsymbol{A_{PL}}(t)\boldsymbol{n_{PL}}(t)$$

$$\frac{d\boldsymbol{n_{OG}}}{dt} = \boldsymbol{A_{OG}}(t)\boldsymbol{n_{OG}}(t) \quad (1)$$

Here $\boldsymbol{n_{PL}}(t)$ is the time dependent isotope concentration vector for species in the primary loop (PL) which includes the reactor core, pump, heat exchanger, etc., $\boldsymbol{n_{OG}}(t)$ is the time dependent isotope concentration vector for species in the off-gas (OG), and $\boldsymbol{A_{PL}}(t)$ and $\boldsymbol{A_{OG}}(t)$ are the time dependent transition matrixes for the two regions respectively. Current MSR capabilities are limited to a 2-region problem where there is only removal from the reactor primary loop (PL) region to the off-gas (OG) region without allowing for species to re-enter the primary loop. Griffin builds the complete depletion matrix from the two-region depletion matrices from Equation 1, as seen in Equation 2, and solves the method explicitly with the Chebyshev Rational Approximation Method (CRAM) [10].

$$\boldsymbol{A_2} = \begin{bmatrix} \boldsymbol{A_{OG}} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{A_{PL}} \end{bmatrix}$$

$$\boldsymbol{n_2} = \begin{bmatrix} \boldsymbol{n_{OG}} \\ \boldsymbol{n_{PL}} \end{bmatrix} \tag{2}$$

In Equation 2, A_{OG} is the depletion matrix for off-gas region and A_{PL} is the depletion matrix for the primary loop region. Now that both regions can be resolved, the loss matrix $\boldsymbol{L}$ is introduced to couple the two regions within the CRAM solver where the L_n terms on the diagonal are the removal terms for each isotope in the primary loop region. Here the L_n terms are reduced order terms that capture the combined physics of chemical volatilization and the mass transfer speed of isotopes from the primary fuel-salt loop to the off-gas system without allowing for the possibility of species to return to the reactor primary loop region. The loss matrix reads as follows in Equation 3.

$$\boldsymbol{L} = \begin{bmatrix} L_1 & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & L_n \end{bmatrix} \tag{3}$$

Incorporating Equation 3 back into Equation 2 yields Equation 4: the two-region depletion matrix with one way removal which is then solved in Griffin. Since the removal is only one-directional without allowing for species in the off-gas system to reenter the primary loop – there is only one loss matrix $\boldsymbol{L}$ in the upper right-hand corner.

$$\boldsymbol{A_2} = \begin{bmatrix} \boldsymbol{A_{OC}} & \boldsymbol{L} \\ \boldsymbol{0} & \boldsymbol{A_{IC}} \end{bmatrix} \tag{4}$$

It is important to note that CRAM was developed with the expectation of a specific sparsity pattern in the depletion matrix, with the depletion matrix containing significant off-diagonal terms in either the upper or lower triangular portion of the matrix. In Griffin, the depletion matrix is designed to be upper triangular as nuclides are ordered such that light nuclides are at the top of the matrix and heavy, fissionable nuclides are at the bottom. This ordering results in the fission product production terms clustering in the rightmost columns of the depletion matrix, which makes the depletion matrix in Griffin largely upper triangular with lower triangular off-diagonal terms clustered near the diagonal.

By only considering flow from one region to another and no return flow, it is possible to make the 2-region depletion matrix comply with the expected sparsity pattern of CRAM. This yielded the performant solves for the one-direction removal system. However, it should be noted that in the event of two-way flow, the sparsity pattern expected by CRAM would be violated, potentially necessitating the use of another matrix exponential solver, which is not the scope of this study.

PROBLEM DESCRIPTION FOR CONFIRMATORY STUDIES

To ensure that this new depletion capability was correctly implemented into Griffin, the following test case was devised as a benchmark problem to test Griffin against SERPENT. For simplicity, a cube-geometry, infinite, homogenous medium model was chosen to be representative of an MSR core.

The specifics of the simplified MSR design used in the problem are listed in Table I. The MSR design model is a fast spectrum molten chloride salt reactor using High-Assay Low-Enriched Uranium (HALEU) UCl_3 fuel. In these confirmatory studies, the model is used for depletion with and without removal and is also the starting point for analyzing what type of insoluble material may be removed into the off gas during burnup.

TABLE I. Simple MSR Test Case Design Specifications

Parameter	Value
Salt Composition	$0.33UCl_3$-0.67NaCl
Salt Density	3.13 g/cm^3
Enrichment	19.75%
Volume	1.00E+6 cm^3
Temperature	700ºC
Power Density – Power	200W/g – 321 MW
1G Flux	6.72E+15 neutrons/(cm^2-s)
Fission Rate	9.89E+12 fissions/(cm^3-s)
K_{eff}	1.23 ~ 1.21
Spectrum	Fast

DEPLETION RESULTS & ANALYSIS

The following section lists the various isotopic depletion studies that were conducted to verify Griffin against SERPENT for isotope removal to an off-gas system. It is important to note that isotopic depletion has been previously implemented in Griffin and verified against both ORIGEN and DRAGON-5 [11]. The following studies verify the newly added isotope removal capability against SERPENT for a given depletion period. The process for each study starts with

a depletion calculation in SERPENT. The spectrum weighted microscopic one-group cross sections and the resulting one group flux are taken from SERPENT and are then exported and converted to ISOXML format to be used in Griffin. Griffin does not perform a neutronics solve, but rather uses this information from SERPENT to only perform a depletion solve. Currently the microscopic cross sections generated by SERPENT are not tabulated by burnup, but instead one set from mid-life burnup are chosen and used in Griffin for the entire burnup calculation.

Isotopic Depletion Without Removal

The first study conducted was the simple case of depletion with no removal. This was done to ensure that Griffin can replicate SERPENT results for a normal depletion problem within expected error bounds.

TABLE II. Isotopic Depletion Without Removal

	SERPENT	Griffin	Abs Diff
Fission Reaction Rate [fission/(cm^3-s)]	9.89E+12	9.94E+12	0.5%
I^{127} In Primary Loop [atom/(b-cm)]	1.04E-7	1.05E-7	0.6%
I^{129} In Primary Loop [atom/(b-cm)]	4.08E-7	4.09E-7	0.2%

Table II shows the fission reaction rate density calculated by SERPENT and Griffin for midlife burnup are in good agreement despite Griffin using a constant flux to perform the burnup. The atomic densities of two iodine isotopes are compared at 70 days burnup and show good agreement. Discrepancies in results are attributed to the fact that Griffin is using a one-group flux and one-group cross sections collapsed from the continuous energy cross sections generated by SERPENT for midlife burnup. Using burnup tabulated fluxes and microscopic cross sections should yield even closer results.

Isotopic Depletion with Removal to Off-Gas System

The second study conducted involved the removal of iodine from the core region to the off-gas system in the MSR model. Table III shows good agreement between the atomic densities of the same two iodine isotopes that have been removed to the off-gas system over a 70-day burnup period. The removal rate for iodine in the system is set at $r = 3.85 * 10^{-3}\frac{1}{s}$, which is very fast when compared with the generation from fission, transmutation, and decay terms. As a result, nearly all the iodine generated in the primary loop is quickly extracted to the off-gas system, and a slightly greater amount is allowed to build up there compared with Table II since the amounts in the off-gas system do not experience any transmutation loss from the flux in the core.

TABLE III. Isotopic Depletion with Removal to Off-Gas System

	SERPENT	Griffin	Abs Diff
I^{127} In Off-Gas [atom/(b-cm)]	1.06E-7	1.07E-7	0.5%
I^{129} In Off-Gas [atom/(b-cm)]	4.10E-7	4.11E-7	0.3%

Effect of Iodine Solubility on Iodine Removal to Off-Gas System using Griffin

Lastly, it is important to look at how chemical solubility will affect the isotopic removal rate used in such a depletion analysis. Like the previous studies, these results are reported over a 70-day burnup.

TABLE IV. Effect of Iodine Solubility on Iodine Removal to Off-Gas System

	Decreasing Solubility →			
Removal rate (1/s)	3.85e-9	3.85e-8	3.85e-7	3.85e-6
Percentage I^{129} in Off-Gas	1.13%	10.5%	60.1%	95.4%

Table IV summarizes how the removal rates for chemical species to the off-gas system are largely a function of the solubility of the species in the fuel-salt and why it is critical to determine these rates accurately for predicting amounts of material accumulating in the off-gas system.

It is important to note that performing depletion and predicting species removal to the off-gas system in such a way is considered a reduced order approach. Most notably the spatial dependence of isotope concentrations is not included here which will be significant for isotopes that have a decay half-life in the order of the circulation time of the fuel in the reactor loop. Also, chemical solubility, mass transfer/reaction kinetics, and flow effects from the velocity and temperature field are all wrapped into this one removal rate term within the Bateman equations. This approach may be suitable for certain situations, but the removal rate term must be correctly informed by higher fidelity models operating on smaller spatial/time domains.

DISCUSSION & CONCLUSION

This work details the implementation of isotope removal capabilities for multi-region depletion in the NEAMS

neutronics code Griffin for MSR off-gas analysis. Furthermore, it showcases the importance of accurately determining the removal rates of various insoluble fission products in an MSR system – a rate better informed through multiphysics modeling of such coupled systems. Accurate estimation of nuclear isotopes inventory in the off-gas system is key for the source term estimation in MSRs.

Future work currently underway at Idaho National Laboratory is extending capabilities in determining the isotope removal rates via multiscale and multiphysics modeling of MSR systems using the NEAMS codes built upon the MOOSE framework. The incorporation of thermochemical properties from the Molten Salt Thermodynamic Database (MSTDB) [12] in a Gibbs Energy Minimizer such as Thermochimica or Yellowjacket can give solubility information for various chemical species. Next the spatial component of species transport can be resolved through the coupling of mass transfer/reaction kinetics code Mole and thermal-hydraulics code Pronghorn.

Utilizing the NEAMS code suite allows researchers to obtain a best estimate of the multiphysics occurring in the system and further facilitates the determination of key operational requirements of various reactor components in the experimental reactor system. This enables requirements for the off-gas system to be determined analytically by identifying the variety of possible insoluble species and quantifying the amount of such insoluble material accumulating in this region for source term and safety analysis. A similar approach could be leveraged to determine the requirements of a heat exchanger given the chemical potential and flow induced corrosion that could arise due to changes in the salt composition throughout burnup.

Finally, this approach can identify key data that may be missing from current databases which can inform the priority of future experiments. Moving forward the mutual support of modeling and simulation with active experiments will ensure the most important data is gathered so that the first experimental advanced reactors will be built and ultimately commercialized for the clean energy needs of this century.

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