ANL/NSE-23/79

Progress on Long-Term Objectives for the Development of Advanced Reactor Mechanistic Source Term Modeling and Simulation Tools

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Nuclear Science and Engineering Division

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1 Introduction

To assist both the advanced reactor industry and U.S. Nuclear Regulatory Commission (NRC) in the pursuit of reactor design and licensing, the U.S. Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program has established a mechanistic source term (MST) research project under the auspices of the Multiphysics Applications technical area. MST analysis is central to the reactor licensing process and a focus of the NRC regarding their mission to provide reasonable assurance of adequate protection of public health and safety and environment. The MST research project is a collaboration between Argonne National Laboratory (Argonne) and Sandia National Laboratories (SNL) with a high-level objective to coordinate the development of comprehensive advanced reactor MST mod/sim capabilities to support risk-informed design and licensing decisions.

As part of this effort, an MST mod/sim development pathway was developed in FY21 [1], which outlines the high-level objectives and near-term tasks necessary to achieve the project objectives. Since publication of the development pathway, Argonne and SNL have focused initial efforts on addressing the "near-term tasks" outlined in the report, and the current work provides a status update of the progress achieved in FY22. This report is a progress update on 6 tasks supporting molten salt reactor (MSR) and sodium fast reactor (SFR) MST analyses which were initiated in FY22 as recommended high priority near-term tasks. These tasks were continued into FY23 as they also support previously recommended long-term development objectives to advance MST analyses for these advanced reactors.

Due to budgetary constraints in FY23, this project did not perform any tasks explicitly supporting high-temperature gas reactor (HTGR) MST. Despite this, NEAMS continues to support HTGR technology development through code and model development activities that are crosscutting with those previously recommended to advance HTGR MST analyses. This includes advancing modeling of core neutronics and fuel depletion with *Griffin*, thermal hydraulics with *Pronghorn* and *SAM*, high-fidelity multiphysics with *Cardinal*, and TRISO fuel performance with *BISON*, including RN releases from TRISO particles. Additionally, the DOE-NE ART Gas-Cooled Reactor (GCR) campaign supports modeling studies as well as experimental efforts, many of which are improving the understanding of RN transport in HTGR systems.

2 MSR

The long-term objectives for MSR and fluoride high-temperature reactor (FHR) MST mod/sim are briefly described here and are in accordance with the important phenomena, relevant computational capabilities, and the resulting information gaps which have been outlined in previous reports [2, 3] and expand on recent efforts in MSR MST model development [4, 5]. The long-term development objectives are summarized below and are described in more detail in [1].

Because MSRs are strongly multiphysics in nature, many of the development objectives are cross-cutting with other MSR-related research programs. Computational challenges in reactor physics for MSRs, which are being addressed, will also provide for improvements in radionuclide inventories. Salt chemistry plays a pivotal role in the design, operation, safety, and decommissioning of MSRs, and therefore this development objective overlaps with ongoing research which supports all these facets. Solubility, precipitation, vaporization, bubble transport, and aerosol formation all affect reactivity of the nuclear reactor in some way, and thus this supports other research considering operational concerns. The production of tritium in large amounts by some MSR designs warrants further investigation on the radiological consequence of such releases due to its ability to permeate alloy structural components. Additionally, off-gas system modeling, as well as other auxiliary systems such as on-line fuel processing, affects the operation, safety, and radionuclide/material accountancy of MSRs. And finally, accident scenario modeling will also motivate design choices while potentially optimizing the economics of MSRs.

In FY23, four tasks were performed to support the aforementioned long-term development objectives, continuing work initiated in FY22 [6]. One task that is ongoing into FY24 plans to be submitted as a conference paper. The other three tasks are documented separately in Argonne technical reports (cited below) because they are partial efforts supporting larger collaborations with other DOE-funded research projects. Two of these Argonne reports have been completed and are awaiting release to OSTI, while the third report will be completed in FY24.

S. Thomas and J. Jackson, "Integrated Process Testing of MSR Salt Spill Accidents," Argonne National Laboratory, ANL/CFCT-23/25, September 2023.

Y. Cao, J. Fang, Y. Jeong, S. Shahbazi, and T. Fei, "Multiphysics Simulation and Benchmark of MSRE," Argonne National Laboratory, ANL/NSE-23/58, September 2023.

R. Gong and S. Shahbazi, "Demonstrating PyCalphad, ESPEI, and MSTDB-TC for MSR Applications," Argonne National Laboratory, ANL/NSE-24/64, September 2024.

2.1 Salt Chemistry

Long-Term Development Objectives: [1]

- *Continue development of molten salt thermodynamic database and solver which improves confidence in chemical and phase equilibrium calculations for radionuclide speciation and phase changes in molten salt systems*
- *Develop radionuclide species-tracking tool which couples aforementioned thermodynamic modeling tool with various relevant mass transport phenomena, including those listed in the following sections*
- *Develop and validate models on radiolytic effects in molten salts and its impact on radionuclide transport during normal operation as well as accident scenarios*

2.1.1 Task Update: Salt thermochemical modeling and UQSA with MSTDB-TC

An Argonne technical report will be completed in FY24 [7] providing the details of this task which was initiated in FY22 and continued in FY23. An update summary is provided here. The *Molten Salt Thermal Properties Database – Thermochemical (MSTDB-TC)* [8, 9] is a NEAMS-developed thermodynamic database for molten salt thermochemical equilibrium calculations completed with Gibbs energy minimization (GEM) solvers such as *Thermochimica* [10, 11], *FactSage* [12], and *PyCalphad* [13, 14]. The database depends on a base library of pure compound data and relies on the modified quasichemical model in the quadruplet approximation (MQMQA) solution model to realistically model interactions between components dissolved in molten salts. The solution model parameters are typically optimized from experimental data for lower-order systems such as pseudo-binary solid-liquid phase diagram data. As such, a study was recommended to perform uncertainty quantification and sensitivity analyses (UQSA) and assess the quality of the database for calculations relevant to source term such as vapor-liquid equilibria which may not always be accurately accounted for by solid-liquid phase diagram data.

In order to assess the quality of *MSTDB-TC*, experimental data on vapor-liquid equilibria of molten salt systems is necessary. Data from the ORNL Molten Salt Reactor Program (MSRP) represents one of the only sources of experimental vapor pressure data of multicomponent molten salt mixtures. Specifically, a series of low-pressure distillation experiments allowed approximate measurement of values of relative volatility with respect to LiF across over a dozen different solutes. A total of over 50 unique mixtures were distilled in these experiments, with systems containing between 2 (pseudobinary) and 5 (pseudoquinary) total endmembers.

Much of the work associated with this task was completed in FY22 and FY23 prior to the release of *MSTDB-TC* v3.0 [15], thus the bulk of assessment work reported here involves the use of *MSTDB-TC* v2.0 [9]. Nonetheless, the recently released *MSTDB-TC* v3.0 includes addition of new salt-soluble fission product elements not previously included. And so, the same calculations completed with v2.0 were run with v3.0 and preliminarily verified towards the end of FY23. In addition to this, two lanthanides for which there exist experimental ORNL Molten Salt Reactor Program (MSRP) data (Sm, Eu) have not yet been optimized and added into v3.0. Therefore, *Extensible Self-optimizing Phase Equilibria Infrastructure (ESPEI)* was used to optimize the model parameters for those endmembers not yet added. This served as both a method of closing information gaps as well as an investigation into the utility of *ESPEI* for automatic, highthroughput optimization of MQMQA solution model parameters from experimental phase diagram data, which could be used to improve efficiencies in *MSTDB-TC* development.

For all work involving *PyCalphad*, the conversion of *MSTDB-TC* from .dat format to .xml format was necessary. This conversion was verified by comparing the phase diagrams of multiple important systems that are calculated using the .xml format with *PyCalphad* against those diagrams submitted with the database documentation. *PyCalphad* and *FactSage* have also been verified against the NEAMS tool *Thermochimica* previously [14]. Once verified, *PyCalphad* and *ESPEI* were used in this demonstration because of the unique UQSA capabilities that *ESPEI* offers which utilize Bayesian statistics. The utility of *ESPEI* in the model selection process is demonstrated by using Bayesian inference to inform the optimized model parameters. The optimization process relies on finding the higher probability of likelihood that a set of parameters represents the experimental data. Newly optimized parameters are discovered for lower order systems not previously known. The *ESPEI*-optimized parameters are used to produce phase diagrams that are shown to be in good agreement with literature.

A modified version of *MSTDB-TC* was created (referred here as v3-mod) to be able to include noble gas species in calculations which allows a predominantly noble gas vapor phase above the salt, just as in a real MSR. Additionally, other modified versions were made in certain exercises when comparing *ESPEI*-optimized values against the parameters in *MSTDB-TC*. Therefore, a collection of different databases including multiple versions of *MSTDB-TC* as well as baseline ideal solution models were used to calculate the relative volatilities of every solute in every salt system for which experimental data was provided by the ORNL MSRP distillation experiments. The quality and utility of *MSTDB-TC* is assessed and demonstrated by comparing calculations to experiment, taking into account the experimental errors estimated by the original ORNL experimentalists. The improved performance and accuracy of the MQMQA solution model within databases such as *MSTDB-TC* over simpler formulations is demonstrated.

Finally, we used *PyCalphad* to generate Ellingham diagrams which are useful in understanding redox reactions of various elements of interest. This was created not only for those elements accounted for in *MSTDB-TC* v3.0, but also additional elements for which pure compound data exists in the *FactSage* thermochemical database *FactPS* or other available databases. The caveat here is that the nonideal effects of molten salt solutions are not accounted for when only considering pure compound thermodynamic data, but for most elements, especially noble metals, this approximation is sufficient and still serves to show trends in speciation and thus behavior in MSR primary loops or RN transport in MSR systems.

2.2 Salt Aerosols and Bubble Transport

Long-Term Development Objectives: [1]

- *Develop models for molten salt aerosol formation, transport, and removal mechanisms and complete ongoing salt spill experimental studies for validation.*
- *In support of model development and validation, complete studies on bubble transport in molten salts, investigating volatile fission product solubility, radionuclide entrainment in flowing bubbles, noble metal behavior as surfactants, and the impact of bubble bursting at surfaces on aerosol formation*

2.2.1 Task Update: Developing mechanistic models for RN transport in salt aerosols

In FY22, the NEAMS MST work package completed literature review of the state of knowledge on molten salt aerosols as it relates to MSR MST analysis [16]. At the same time, experimental work in characterizing the behavior of spilled molten salts was funded in FY21 and FY22 by the ART MSR Campaign [17, 18]. And during FY23, the ART MSR Campaign funded integrated process tests on simulated MSR salt spill accidents that were conducted at a laboratory scale with the goal of characterizing surrogate fission product behavior [19]. The NEAMS MST work package supported the development of a mechanistic model that can describe RN transport behavior for this accident sequence investigated in ref [19], utilizing NEAMS tools where possible. The developed RN transport model can inform and/or be implemented into severe accident analysis codes such as *MELCOR* or NEAMS tools such as *MOOSE/CR*, *Mole*, and/or *SAM* [20]. Details of both the experimental and modeling effort are documented in the FY23 milestone deliverable report supporting the MSR Campaign [19], and high-level conclusions from the modeling work are provided here.

The laboratory tests were conducted using FLiNaK doped with surrogate fission products and simultaneously quantified the heat transfer behavior of the spilled salt, compositional changes to the bulk salt, and the release of surrogate fission products from the spilled salt as aerosol particles [19]. The results from these tests showed that the masses of particulate cesium and iodine measured on aerosol collection filters increased with the initial temperature of the spilled salt. These observations suggested that the main cesium- and iodine-bearing aerosol formation mechanism was vaporization from the spilled molten salt pool followed by the condensation of these species as they moved to cooler regions of the atmosphere. An attempt was made to quantitatively model the formation of Cs- and I-bearing aerosols above a spilled pool of molten salt based on this proposed aerosol formation process. The mass of vapor species released to the atmosphere from the spilled molten salt was approximated with a time-dependent mass rate correlation that is a function of the saturation concentration gradient of the species. The temperature-dependent vapor pressures of each cesium- and iodine-bearing species of interest above FLiNaK were calculated using *MSTDB-TC* v2.0 [9]. The calculated vapor pressures of species of interest for one tested salt composition are shown as a function of temperature in Figure 1. The mass transfer coefficients of the correlation were also varied to investigate their sensitivity on the predicted amounts of cesium and iodine released to the aerosol collection filter. It was assumed that vapor release from the salt pool ceased when the salt surface froze.

When comparing the mass transport model to experimental measurements of the total collected mass of cesium and iodine aerosol particles under various test conditions, it was found that the model predicts such values to within one to two orders of magnitude. The predicted amount of iodine released was typically greater than that predicted for cesium, which was driven by the larger vapor pressure values of KI compared to CsI and CsF. But the mass of particulate cesium collected on the filters was typically three to five times larger than that of particulate iodine. Further development of the mechanistic model as well as experimental measurements are recommended to validate the vapor pressure curves for these species above FLiNaK.

In addition to uncertainties associated with the calculated vapor pressures, possible explanations for discrepancies between the modeled and measured values include experimental measurement uncertainties (e.g., unknown aerosol collection efficiencies), the use of mass transfer coefficients that are constant with temperature, and other physical assumptions inherent to the model chosen (e.g., assuming all released vapors condense to form aerosol particles). Although experimental results from the specific test system support the hypothesis that aerosol formation was correlated to the vaporization and condensation mechanism, aerosol formation is likely a complicated combination of different mechanisms in varying magnitudes. It is recommended that the mass transport model is expanded in the future to account for additional formation mechanism phenomena when more experimental data becomes available. Without high-quality experimental data and corresponding mechanistic model development, information gaps in modeling aerosol behavior for MST analyses will continue to exist. As such, engineering-scale design concepts have been proposed and recommended recently to aid in the closing of these information gaps and support MSR licensing [21]. This work represents one of the first attempts to characterize RN transport by aerosol formation from spilled surrogate MSR fuel salt through joint studies in experiment and modeling. Other recent efforts have demonstrated similar modeling approaches but have not had experimental data to validate results against [22-24]. This work also demonstrates the importance of continued development of thermochemical modeling capabilities such as *MSTDB-TC* and *Thermochimica* to MSR design and safety analyses.

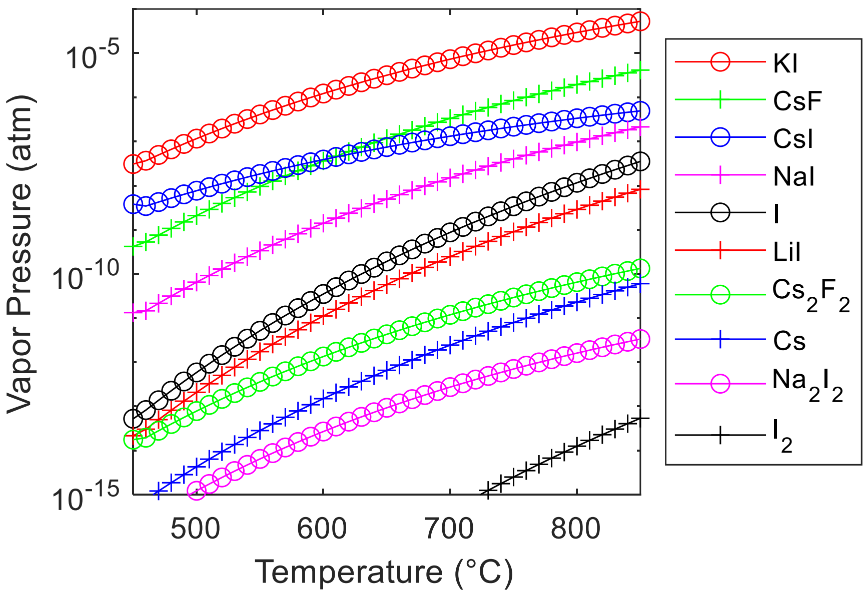

Figure 1: The 10 cesium- and iodine-bearing species with the highest vapor pressures above the surrogate FP-containing FLiNaK (“low burn-up” mixture), as calculated by MSTDB-TC v2.0.

2.3 Tritium Modeling

<u>Long-Term Development Objectives: [1]</u>

- *If deemed necessary due to radiological impact, implement tritium-specific transport models within severe accident analysis or system-level analysis codes to aid in the simulation of tritium releases during accident or transient scenarios*
- *Complete experimental studies to better understand tritium releases from graphite or other trapping materials during temperature transients*

2.3.1 Task Update: Developing a SAM tritium transport model of the MSRE

Tritium is considered to be a potential radiological hazard if released to the environment as a unique source term that can diffuse through metallic components and escape the reactor system [25]. Therefore, it is important to track inventory and monitor pathways of tritium to assess the risks associated with releases from MSRs. It necessitates a comprehensive model to simulate tritium behavior within MSR systems, which is multiphysics problem that involves both chemical and physical processes governing the transport and retention of the tritium. Experiences gained from MSRE are the only source of experimental data representing behavior of tritium in the operation of salt fueled MSRs to verify/validate available modeling and simulation tools. Tritium transport modeling capabilities within the *System Analysis Module (SAM)* [26] code developed by Argonne can be leveraged, which have been implemented as a result of collaborations between Argonne and Kairos Power for capturing key phenomena of FHRs [27].

A *SAM* tritium transport model of the MSRE has been prepared based on a *SAM* reactivity insertion test model for MSRE previously developed [28]. It consists of the primary loop, which includes a representative core channel, a representative graphite moderator, a heat exchanger, a pump, and piping connecting major components. Tritium (T_2) is treated as passively transported by the bulk fluid flow. Permeation of tritium (T) within a metal such as the heat exchanger tube is modeled as a diffusion process according to Fick’s law. Permeation and retention of the tritium (T) within a graphite is modeled by calculating the diffused and trapped tritium concentrations via a single bulk-diffusion process. The transport of tritium from the fuel salt into any solid region and vice-versa was modeled as convective mass transfer coupling the diffusion and advection processes occurring in each domain [27]. Generation of tritium via fission was modeled by a species source term in the core as a function of power density. Removal of tritium by the off-gas system at the pump bowl was modeled as a volumetric scalar sink term. The baseline condition of the MSRE tritium transport analysis was set as nominal operation conditions for 8 MWth power with 95 % tritium removal by the off-gas system with tritium allowed to escape through the primary heat exchanger. Temperature-independent tritium transport properties for the fuel salt, graphite, and steel were considered initially for the baseline calculations. Details of the model and the baseline simulation results of the MSRE transport analysis are available in a separate NEAMS milestone report supporting the MSR Multiphysics Applications work package [29].

The results show that the MSRE tritium transport model reflects and captures expected phenomena in the primary system. For instance, more than half of the tritium inventory was calculated to be located outside the core region in the ex-core region of the primary fuel salt loop. In addition, the moderator surface had a steep gradient of the tritium concentration. These align with experimental observations during MSRE operation. For example, tritium was measured in the cell atmosphere which was believed to be caused by permeation from the reactor structures and piping, providing evidence of tritium’s ability to permeate alloys. And the tritium concentration as a function of depth was measured in graphite specimens [30-33]. Although the baseline conditions used in the calculation might be different from the true operating conditions and history of the MSRE, results appear to agree with experiment. Improvements are still needed in off-gas system modeling as well as a better strategy to reach the equilibrium state, which may take varying timescales depending on the phenomena included.

Indeed, the calculated tritium distribution of the MSRE should vary depending on the input parameters used in the model, such as tritium transport- or thermal-fluids-specific properties (e.g., thermophysical properties), assumptions in the chemical form of the tritium, simplifications of the off-gas system modeling, etc. The impact of parameter changes and deviations from baseline conditions can be further explored to fill the gap of understanding on the MSRE tritium distribution and its pathways. For instance, leveraging advanced software for sensitivity analysis, such as the *MOOSE Stochastic Tools* module [34] can be utilized in performing sensitivity analyses on a large number of the parameters related to coupled physics phenomena with reduced computational expenses. The sensitivity study is expected to provide useful information in analyzing the radiological consequence of tritium releases from MSRs to assess the risks associated with MSRs that produce larger tritium inventories than other reactors. Such a study is planned as future work pending funding.

2.4 Auxiliary System Modeling

Long-Term Development Objectives: [1]

- *Develop modeling tools for the auxiliary systems proposed alongside the primary loop reactor system as details become available*
- *Integrate chemistry knowledge specific to the auxiliary systems into the modeling tools*
- *Coordinate experiments aimed at investigating noble metal-specific behavior in off-gas systems such as gas phase transport and deposition*

2.4.1 Task Update: Modeling RN transport in MSR off-gas systems

This task was continued in FY23 in collaboration with INL researchers supported by the NEAMS MSR Applications work package and the DOE-NE ART MSR Campaign. The work includes the development and demonstration of NEAMS tools to provide RN inventories in hypothetical auxiliary systems of MSRs. Specifically, this ongoing work is being documented in an upcoming conference paper or journal article that will be released during FY24. Demonstrated is the use of NEAMS tools for calculating RN inventories in an MSR off-gas system (OGS) with benchmarking against non-NEAMS tools. Additionally, reduced order models based on Bateman equations that were previously reported [35] are used to provide new insights and investigate radiochemical transport phenomena to influence and inform NEAMS tool development. A brief review of this ongoing work is provided with some preliminary results.

Recent development efforts funded by the NEAMS program have enhanced code capabilities in modeling MSR systems and have demonstrated multiphysics code coupling for problems like chemical species transport through cross-cutting funded efforts under the ART MSR Campaign. The resulting state-of-the-art multiphysics framework for chemical species transport analyses is shown in Figure 2, consisting of coupled NEAMS tools *Griffin*, *Pronghorn*, and *Thermochimica*, enabling depletion-driven spatially resolved thermochemical modeling of MSR systems [36]. This framework can be directly leveraged to perform source term modeling of both MSR systems and auxiliary systems such as the OGS.

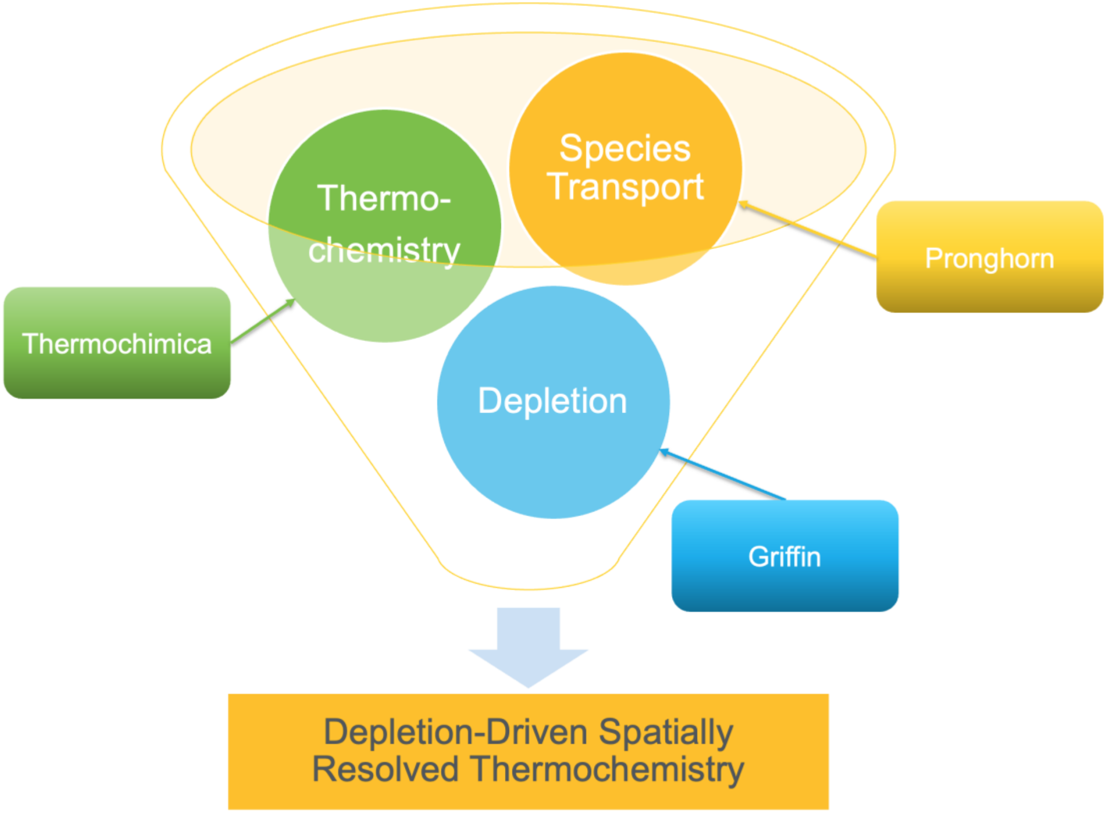

Figure 2: Multiphysics and multiscale framework for MSR species transport and source term analysis being developed by the NEAMS program.

As seen in Figure 2, three different codes are used to model this multiphysics problem at the correct scale and fidelity to capture the evolving behavior of radionuclide transport in MSR systems. Starting first with species transport, *Pronghorn* solves certain spatially resolved partial differential equations of key radionuclide chains in the reactor system undergoing depletion with the explicit tracking of mass transport of insoluble radionuclides out of the MSR system to interfaces and the OGS [37, 38]. *Griffin* solves the Bateman equations (i.e., zero-dimensional ordinary differential equations) for multi region depletion to track the total nuclide inventory in the MSR system and the auxiliary OGS [39, 40]. *Thermochimica* then solves the thermochemical equilibrium of the system given the existing *MSTDB-TC* database based on the nuclide inventory from *Griffin* and the spatial distribution of chemical species from *Pronghorn*. This determines key chemical equilibrium information like the redox potential of the fuel salt and speciation fractions of radionuclides in the ideal gas phase [36, 41]. This information is then re-iterated in the multiphysics solve to form a self-consistent solution that determines the extraction rate of radionuclides to an OGS for source term analysis.

Prior to demonstration with NEAMS tools, it was necessary to complete a scoping study on nuclides-of-interest for modeling MSR off-gas systems through use of a tool that has been benchmarked against MSRE experimental data [42]. This depletion model of the actual MSRE power history quickly provides a first approximation at the highest contributors to (1) total effective dose equivalent (TEDE) and (2) decay heat. Preliminary results are shown for the highest fission products in Table 1 below. Each value is normalized as a percentage of the total salt's TEDE dose or decay heat at that depletion step. The RN inventory is calculated at the inlet of the core region during normal operation, therefore it accounts for considerable decay of the shortest-lived nuclides ($t_{1/2}$ < 5 seconds) throughout the MSRE primary loop (ex-core residence time ~ 20 seconds). The TEDE values are calculated from dose conversion factors (DCF) as tabulated in FGR 11 and 12 [43, 44].

Table 1: Preliminary results of a MSRE scoping study to identify fission products that are the highest contributors to TEDE (left) and decay heat (right)

Fission Product	TEDE [% of total]	Fission Product	Decay Heat [% of total]
Ce144	26.655%	Cs138	2.650%
Y91	5.896%	La142	2.557%
I131	3.638%	I134	2.520%
Ru106	2.859%	Rb91	2.493%
Zr95	1.975%	Rb89	2.422%
Sr90	1.903%	Rb90	2.359%
Te132	1.653%	Sr93	2.352%
I133	1.357%	La144	2.269%
Ce141	1.259%	Y94	2.163%
Pr143	1.227%	Kr89	2.099%
La140	1.006%	Cs140	2.002%
Zr97	0.824%	Y95	1.961%
Sr89	0.789%	Rb88	1.797%
Ce143	0.765%	I132	1.708%
Mo99	0.742%	La140	1.621%
Pm147	0.713%	Ba141	1.474%
Ba140	0.663%	Kr88	1.437%
Nb95	0.631%	Cs139	1.428%
Y93	0.552%	Xe137	1.419%
La142	0.417%	Sr94	1.410%

There are several caveats to these results and should be treated as preliminary only. First, the fission product yields will differ between the fuel sources and these results are indicative of a fission product inventory produced from roughly 1.5 years of cumulative burn-up of ^{235}U operation followed by 1 year of cumulative burn-up of ^{233}U operation. Second, these results do not account for cleaning of the salt as well as flowing fuel effects such as in-loop chemistry, and only consist of activities measured on-line. As a result, they give more weight to the shortest-lived nuclides based on the higher activities. The true impact on source term releases can only be estimated after accounting for cooling time that represents material hold-up and transport throughout the reactor system. Therefore, these results do not constitute an MST analysis, but rather a scoping study on depletion results that may be perceived as the first step in an MST analysis. Third, although the dose conversion factors (DCFs) utilized are from the NRC-recommended FGR 11 and 12 reports, published in 1988 and 1993, respectively, it may be advisable to assess the appropriateness of using these DCFs for liquid-fueled reactors. Similarly, updated values for some DCFs have been reported since the publication of these documents, therefore it is recommended that future work revisits the validity of these coefficients.

Based on the above results, ^{144}Ce represents one of the most important RNs in terms of dose consequence due to it being one of the highest activity FPs as well as having a high DCF for committed effective dose equivalent (CEDE), a major portion of TEDE. But given the chemistry of its precursors, this nuclide will likely stay in the salt to a large degree and may not be as much of a concern for off-gas systems. Nuclides that could represent large dose contributors that also have precursors that exhibit varying chemical behaviors include ^{91}Y and ^{131}I. Additionally, as mentioned, results will vary with cooling time, eventually giving weight to longer-lived FPs. As such, ^{137}Cs becomes one of the top 20 FPs in terms of dose consequence after just 2 days of cooling time. Similarly, ^{140}La becomes the largest contributor to decay heat after just 30 minutes of cooling time. Therefore, using these nuclides as examples, radiochemical transport analysis is briefly demonstrated below as a way to investigate dose consequence and decay heat sensitivities of MSR off-gas systems.

A brief demonstration is provided on the Molten Salt Fast Reactor (MSFR) benchmark design [45]. Specifically, results are provided from (1) NEAMS tools to demonstrate multiphysics-based species transport, and (2) reduced order models based on Bateman equations which are useful for quick parametric sensitivity studies. These Bateman models can be used to efficiently investigate the impact of species transport on inventory calculations as well as analyze transient behaviors.

For example, ^{91}Y was discussed as one of the most important FPs in this context, but the A=91 fission product isobars are actually predominantly produced as either ^{91}Rb, ^{91}Kr, and ^{91}Sr, each beta-minus-decaying into their daughter, ultimately into 58.51-day ^{91}Y. Therefore, unlike in non-liquid-fueled reactors, the ^{91}Y source term will actually be distributed considerably between the fuel and the OGS due to some function of loop residence time, and precursor chemistry and half-life. Figure 3 (top) shows how removing noble gases in the ex-core region of MSFR can impact the predicted inventory of daughter FPs, reducing ^{91}Y by roughly 10%. Similarly, Figure 3 (bottom) shows how removing both noble gases as well as alkali metals (i.e., Rb) can further reduce the predicted inventory of daughters, reducing ^{91}Y by roughly 34%.

Multiple iodine isotopes are among the largest contributors in terms of either dose consequence and/or decay heat. Precursor elements include tellurium, antimony, and tin, each with varying levels of solubility in molten salts. Tellurium has also been found to attack intergranular cracks in nickel-based alloys [46] and it is unclear if this corrosive behavior will also apply to stainless steels. Because Te is often one of the highest yield precursors for these important iodine isotopes, its deposition on components and removal from the salt will have an influence on the transport behavior of iodine in the primary loop and into the OGS. As can be seen in Figure 4, the removal rate of Te precursors in the ex-core region of the MSFR can considerably reduce the circulating inventory of iodine daughters, ultimately reducing the iodine inventory that transfers to an OGS, or in a spilled salt as a result of an accident scenario. The reduction in these four iodine isotopes ranges from roughly 70% to almost 2 orders of magnitude just through removal of tellurium from the salt. This insight motivates future experimental investigations into tellurium behavior in salt and alloy systems of interest. Ultimately, NEAMS tools such as *MSTDB-TC* and/or Mole will need to be able to account for Te transport in engineering scale simulations, possibly informed by or investigated with tools such as the *MOOSE Phase Field* module or the electrochemical fluid transport code *MOSCATO*.

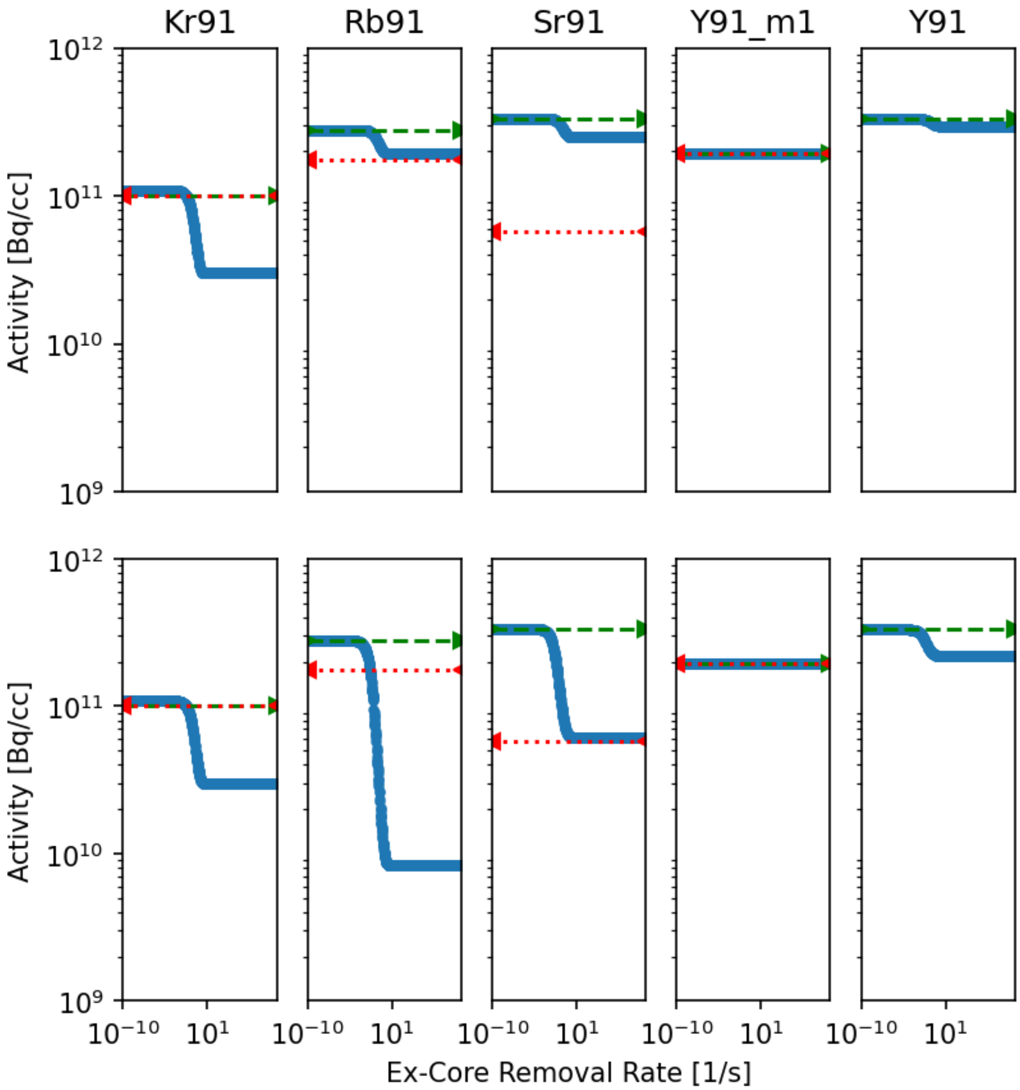

Figure 3: Activities of A=91 fission product isobars as a function of noble gas (top) and noble gas + alkali metal (bottom) chemical removal rates between 10^{-10} and 10^{10} s^{-1} removed in the MSFR ex-core. Red dotted lines pointing left and green dashed lines pointing right represent the independent and cumulative fission yields, respectively.

Using the same tool, it can be shown that continuously removing 13.6-sec ^{140}Xe from the circulating salt will reduce the ^{140}La inventory by roughly 22%, which is approximately the fraction of the ^{140}La cumulative yield that corresponds to ^{140}Xe. It can be concluded that designing an MSR that ensures cleaning of noble gases from the salt within seconds of leaving the center of the core will help maximize the removal of this important precursor. As previously discussed, ^{140}La was the largest decay heat contributor in MSRE after the salt was allowed to cool for 30 minutes and stays the largest until sufficient time has passed to allow decay of its longer-lived precursor ^{140}Ba. An example of such a decay, which would represent a transient shutdown of the MSFR after operating for one year, is shown in Figure 5 below.

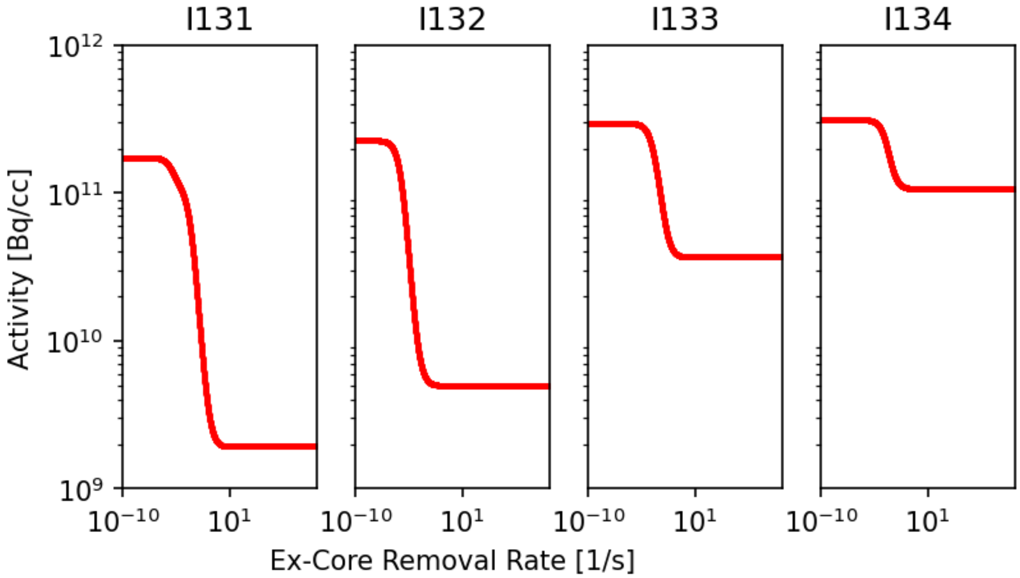

Figure 4: Activities of fission product iodine isotopes as a function of tellurium chemical removal rates between 10^{-10} and 10^{10} s^{-1} removed in the MSFR ex-core.

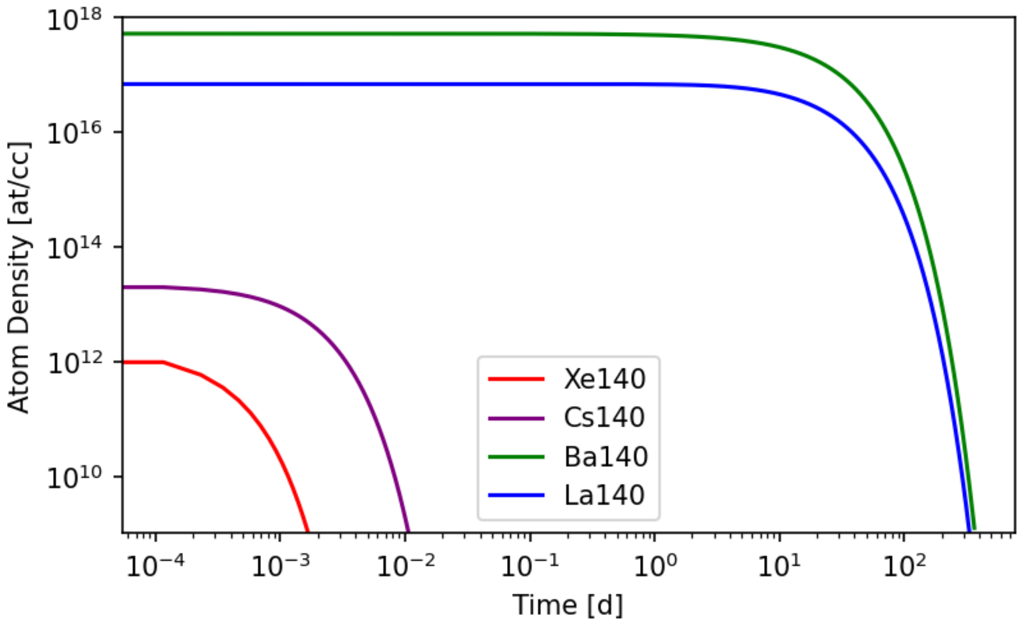

Figure 5: Atom densities of A=140 fission product isobars as a function of the cooling time after MSFR was operated for 1 year.

With an improved understanding of radionuclide transport behavior phenomena, the NEAMS multiphysics framework for MSR analysis can be demonstrated. The framework is used to model the same MSFR design as discussed to illustrate the fidelity of the approach in determining the amounts of ^{137}Cs and ^{131}I that will accumulate in the OGS.

There are several caveats to these results and should be treated as preliminary only and rely heavily on the engineering design under consideration. Starting first with the MSFR design, here a quiescent liquid-gas interface is imposed at the top of the upper plenum as seen in the red boundary in Figure 6. This interface will serve as the extraction point where volatile chemical species not soluble in the molten salt will be advected and diffuse through the interface to a sweeping cover gas. Here we assume that the noble gases and sparse volatile radionuclides will transport to this interface independently of one another and do not agglomerate into larger gas bubbles driven by buoyancy forces.

To model the mass transfer of volatile radionuclides to this OGS interface, the mass transfer equivalent of the Dittus-Boelter turbulent heat-transfer coefficient is used: [47]

$$K = 0.023\frac{D}{d}\left(\frac{\rho d v}{\mu}\right)^{0.8}\left(\frac{\mu}{\rho D}\right)^{0.4} \quad 0.6 \le Sc \le 160; \quad Re_d > 10{,}000$$

where: D is the diffusion coefficient of the species in the fluid, d is the hydraulic diameter of the pipe, v is the velocity, μ is the viscosity, and ρ is the density of the fluid. The mass transfer coefficient is then used in a mass flux boundary condition:

$$m'' = K \cdot (c_{i,interface} - c_{i,bulk})$$

where m'' is the mass flux in moles/(m^2-s), $c_{i,interface}$ is the molar concentration of the species at the liquid-gas interface, and $c_{i,bulk}$ is the molar concentration of the species in the bulk region of the liquid flow [36].

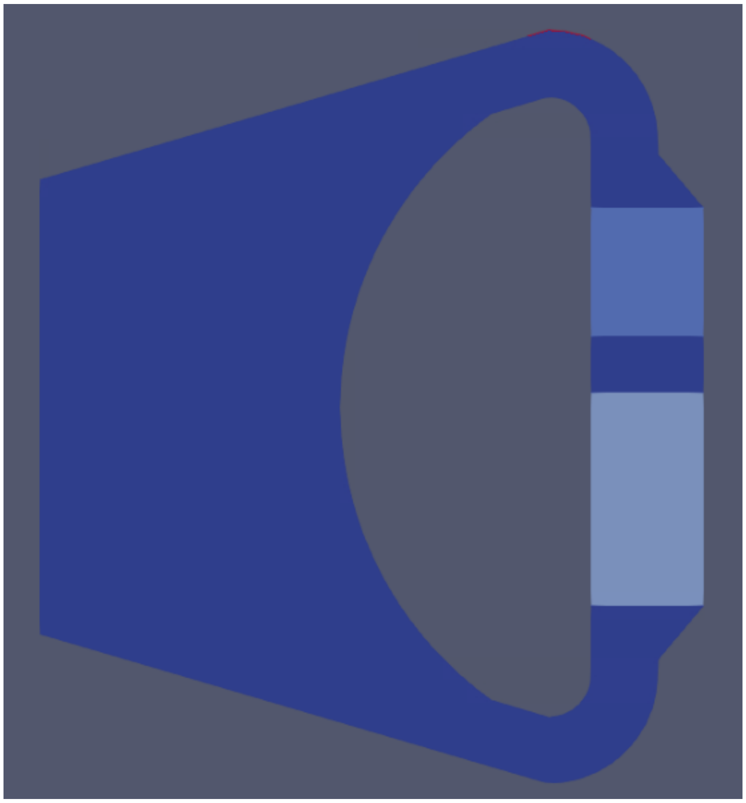

Figure 6: MSFR mesh showing extraction boundary to OGS marked in red at top right of upper plenum.

When analyzing the results of the ^{137}Cs and ^{131}I radionuclide behavior in the MSFR, there are two dominant processes that will drive these species into the OGS. The first is the insoluble mass transport of noble gas precursors to the quiescent liquid-gas interface as described by the mass transfer equation and correlation listed above. This is a dominant factor for ^{137}Cs since its immediate precursor is ^{137}Xe which will diffuse out of the fuel salt. Therefore, the specific engineering design of the OGS and the liquid-gas interface should explicitly consider this to determine how much ^{137}Xe is being extracted. Using the mass transfer modeling identified above, the results for ^{137}Xe in the MSFR are shown in Figure 7 where the extraction of ^{137}Xe can be visualized at the interface and the effect this has on the steady-state ^{137}Xe concentration in the reactor and the buildup of ^{137}Cs in the off-gas is shown in Table 2.

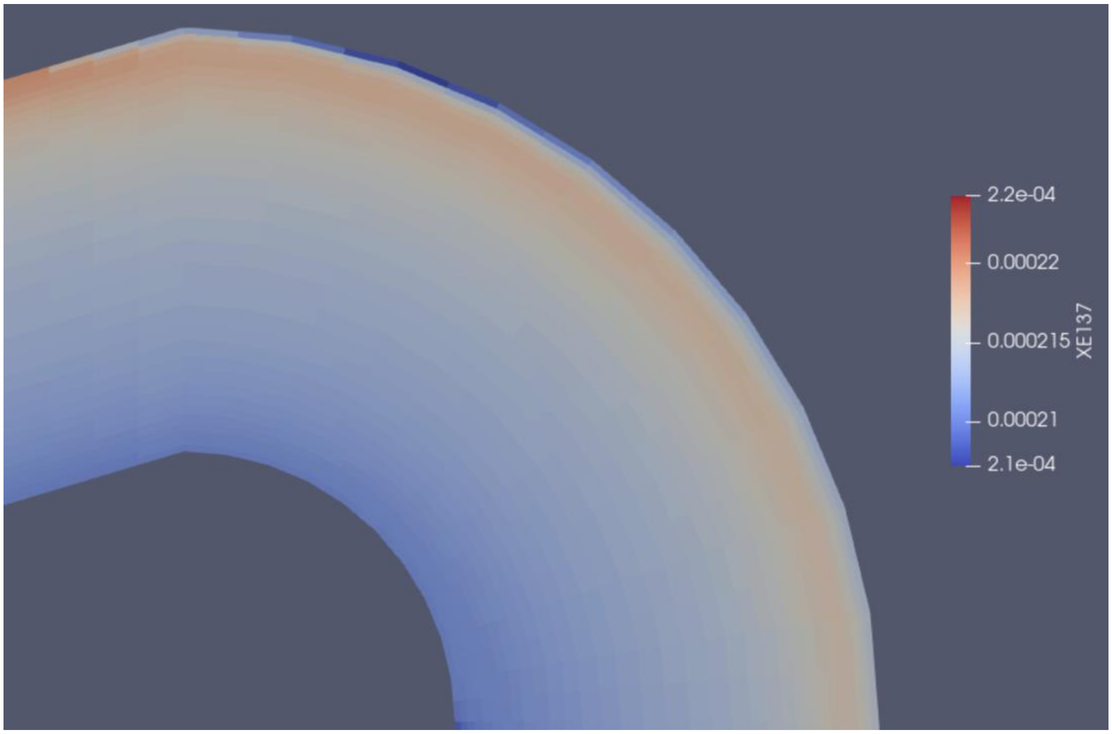

Figure 7: Extraction of ^{137}Xe at the OGS liquid-gas interface.

Table 2: Radionuclides in OGS of MSFR after 2.07 MWd/Kg-U burnup.

Radionuclide	Concentration in Core [atoms/b-cm]	Concentration in OG System [atoms/b-cm]	Percentage in OG System
^{137}Xe	9.94e-11	5.26e-12	5.02%
^{137}Cs	8.87e-07	4.14e-08	4.46%

The second factor that should be considered for both ^{137}Cs and especially ^{131}I is the thermochemical stability in the fuel salt as a function of depletion. Figure 8 shows the fluorine potential and stable amount of iodine in the ideal gas phase at the beginning of life with reduced fuel salt conditions. Here the amount of iodine in the ideal gas phase is extremely small due to the redox potential of the fuel salt being controlled. This is contrasted with Figure 9 which shows the fluorine potential and stable amount of iodine in the ideal gas phase at 2.07 MWd/Kg-U. Here the fluorine potential has greatly increased due to the oxidizing effect of depletion and the lack of redox potential control, and the volatilizing effect this has on iodine can be seen in the increase of stable iodine gas.

These results can be quantitatively examined in Table 3. Here for the scenario where the redox potential is controlled, there is a negligible amount of ^{131}I that has accumulated into the OGS. However, this modestly increases to 0.03% of the total ^{131}I that is extracted to the OGS when the redox potential is not controlled thereby allowing for an increase in iodine volatilization. This is still a very small fraction, showing just how soluble iodine and cesium are in molten fluoride salts. However, given scenarios where the fluorine potential is more oxidizing or the temperatures are higher, such as during an accident scenario, more iodine will volatilize and this must be adequately controlled and predicted for safe MSR operation.

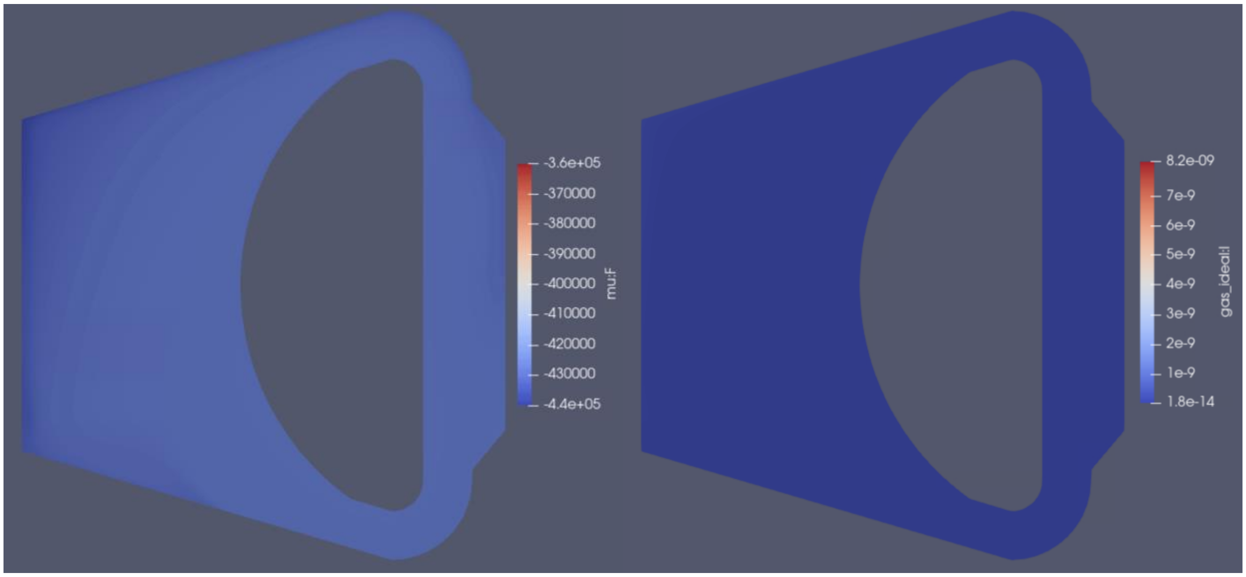

Figure 8: Fluoride (F-) potential (left) [J/mol] and corresponding iodine in stable ideal gas phase (right) [mol] at beginning of life with reduced fuel salt.

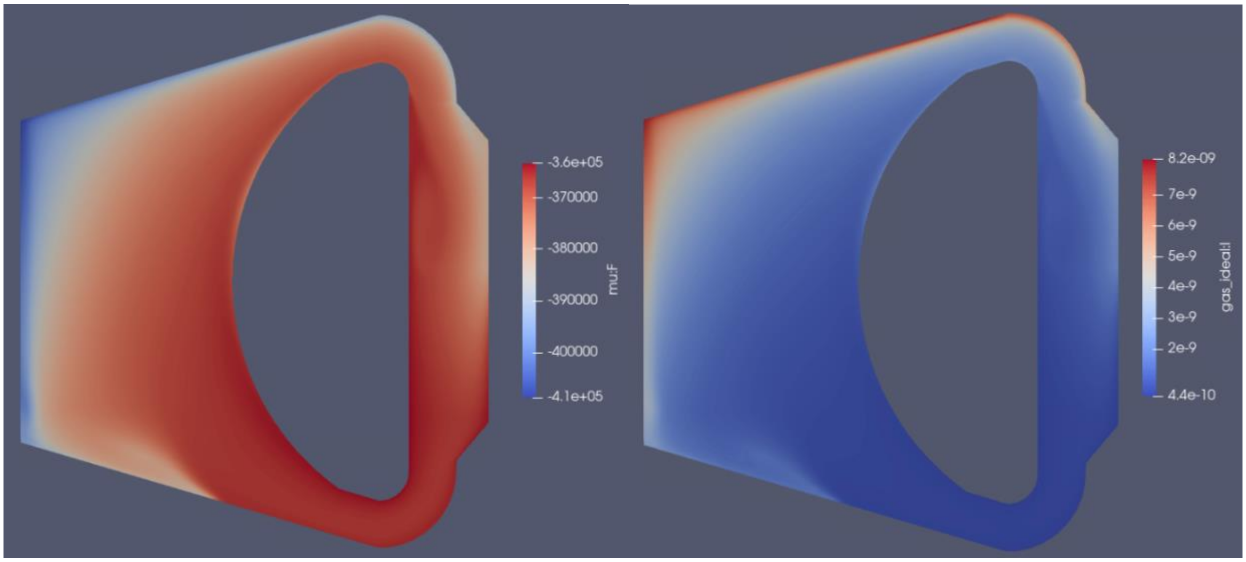

Figure 9: Fluoride (F-) potential (left) [J/mol] and corresponding iodine in stable ideal gas phase (right) [mol] at 2.07 MWd/Kg-U burnup with no redox potential control.

Table 3: ^{131}I in OGS of MSFR after 2.07 MWd/Kg-U burnup with and without redox potential control.

Radionuclide	Concentration in Core [atoms/b-cm]	Concentration in OG System [atoms/b-cm]	Percentage in OG System
^{131}I – redox control	1.75e-07	~0.0	~0.00%
^{131}I – no control	1.74e-07	5.46e-11	0.03%

3 SFR

In general contrast to other advanced reactor types, significant work has been performed in support of SFR MST analysis, due in large part to both historical and recent DOE efforts [48-50]. The results of these projects provide vital insights to remaining gap areas. Based on this information and the assessments performed as part of the current project, the long-term objectives for SFR MST development include those discussed in [1].

Due to past model development and data collection efforts, the long-term objectives for SFR MST focus on completing or enhancing the development and validation of mechanistic models for multiple phenomena. These tasks include detailed modeling of radionuclide migration and release from metal fuel pins within the core and improvements to existing modeling approaches for assessing radionuclide transport within the sodium pool. Lastly, the project also seeks to improve the understanding and modeling of sodium aerosols and vapors within the cover gas region and containment/confinement, including the possibility of leakage pathway plugging due to sodium oxide creation.

In FY23, two tasks were performed to support the aforementioned long-term development objectives, continuing work initiated in FY22 [6]. This includes completion of two topical reports on modeling radionuclide transport for SFR MST via aerosols and vapor-liquid equilibria:

T. Starkus, D. Grabaskas, and S. Shahbazi, "Review and Assessment of Available Data Regarding the Behavior of Sodium Aerosols," Argonne National Laboratory, ANL/NSE-22/68, 2023.

S. Shahbazi, S. Thomas, T. Starkus, and D. Grabaskas, "Modeling Radionuclide Vaporization from Sodium Pools for SFR Mechanistic Source Term Analysis," Argonne National Laboratory, ANL/NSE-22/69, 2023.

3.1 Radionuclide Behavior in the Sodium Pool

Long-Term Development Objectives: [1]

- *Validation of existing mechanistic models for radionuclide bubble transport in liquid sodium*
- *Validation and potential improvement of mechanistic models for the vaporization of radionuclides from the sodium pool*

3.1.1 Task Update: Modeling RN vaporization from sodium pools

This task was summarized in the FY22 milestone report [6] for this work package and is reproduced here. Since FY22, the updated assessment of the sodium pool vaporization thermodynamic database has been completed, and details are provided in a separate Argonne technical report that has been publicly released [51]. The database includes thermodynamic data for all possible species (consisting of the considered elements) that are available in the pure substance database of the *FactSage* commercial chemistry software [12]. The database is in the format of a *FactSage* Compound Database and activity coefficients are utilized in the *FactSage Equilib* module. The *FactSage* Compound Database can be easily converted to *ChemSage* format (.dat file) for use with the NEAMS Gibbs Energy Minimization (GEM) solver *Thermochimica* [10] which can be utilized with *MOOSE*-based codes as well as *MELCOR*. But because of the way in which activity coefficients are translated from *FactSag*e to the *ChemSage* format in this workflow, the non-ideal behavior represented by these activity coefficients are not currently accounted for in *Thermochimica* GEM solves. As such, allowing for the non-ideal behavior of these specific solutes (e.g., Cs) by *Thermochimica* requires the creation of not only the FactSage Compound Database, but also a *FactSage* Solution Model Database based on the relevant solution model which represents liquid sodium. Such a task can be completed in the future if needed.

Details of the development methodology for the sodium pool thermodynamic database are reported separately but it builds on and verifies the work that was reported previously [50]. The elements considered are listed in Table 3-1 along with the species considered to be in solution (ideal and non-ideal mixing) with the Na pool. Assumptions are made regarding how to treat the mixing thermodynamics of various species. Certain liquid phase elemental species were assumed in ideal solution with the Na pool. Based on solubility measurements, the following sodium compounds were also assumed in ideal solution with the Na pool: NaBr, NaI, Na_2O, Na_2Te. Due to a lack of solubility data of oxides in liquid sodium and past SFR operational experiences [52], all oxides and other compounds were not assumed in ideal solution with the Na pool to treat them as conservatively as possible in their vaporization fractions to the cover gas. The activity coefficient functions for the following species were also re-assessed and implemented to represent their non-ideal solution behavior seen experimentally: Cs, Rb, Sr, Ba, NaBr, NaI, Te.

Table 3-1: List of considered elements (left) and species modeled in solution together (right)

Elements considered			Species in ideal solution together*		
Am	La	Rh	Am	Mo	Sb
Ar	Mn	Ru	Ba*	Na	Se
Ba	Mo	Sb	NaBr*	Nb	Sm
Br	Na	Se	Ce	Nd	Sr*
Ce	Nb	Sm	Co	Ni	Tc
Co	Nd	Sr	Cr	Np	Te*
Cr	Ni	Tc	Cs*	Na_2O	Na_2Te
Cs	Np	Te	Eu	Pd	U
Eu	O	U	Fe	Pu	Y
Fe	Pd	Xe	NaI*	Rb*	Zr
I	Pu	Y	La	Rh	
Kr	Rb	Zr	Mn	Ru	

*These species are modeled as exhibiting non-ideal behavior in the Na solution which is represented by temperature-dependent activity coefficient functions. All other species listed are assumed to be in ideal solution with Na.

3.2 Radionuclide Aerosol and Vapor Behavior

Long-Term Development Objectives: [1]

- *Validation and potential improvements to existing mechanistic models for vapor and aerosol behavior within the cover gas region and containment/confinement*
- *Development of mechanistic models for assessment of vapor and aerosol leakage pathway plugging*

3.2.1 Task Update: Modeling sodium aerosols

For SFR MST analyses, the behavior of radionuclide aerosols is a key factor, as noted in recent studies [50]. Due to the use of sodium as the primary coolant, many potential transient scenarios involve the release of radionuclide aerosols in conjunction with sodium aerosols. For such scenarios, sodium may be the dominant aerosol species, compared to that of released radionuclides. Therefore, understanding the behavior of sodium aerosols is particularly important to an accurate assessment of aerosol transport. Although many approaches to aerosol modeling exist, largely drawing from experience with aqueous fluids, it is vital to establish these capabilities in evaluating sodium aerosol behavior for SFR analyses.

In coordination with the DOE-NE Fast Reactor (FR) program, an evaluation was performed of the existing experimental database regarding sodium aerosols. The assessment had two objectives:

1. Assess the available database of sodium aerosol experiments for the evaluation and validation of aerosol models
2. Assess the validity of the Henry correlation for the assessment of sodium aerosol behavior, including limitations and best practices

To achieve the first objective, a survey of available experimentation regarding sodium aerosol behavior was performed, with a specific focus on data concerning aerosol concentration over time. Such information is vital for the assessment of aerosol deposition models. The second objective focused on the validation of the Henry correlation for aerosol modeling. The Henry correlation is a useful, simplified method for the calculation of aerosol concentration over time and is used in several source term analysis codes that are currently being utilized by the SFR industry.

In total, over 100 individual experiments were analyzed from 15 experimental programs. The experimental database was found to span a wide range of aerosol concentrations, with varying compartment characteristics. In addition, the database includes tests that involve both elemental sodium and sodium oxide aerosols. The analysis also found that the Henry correlation provides an accurate assessment of sodium aerosol behavior across the range of conditions covered by the available experimental database. Best practices regarding the utilization of the Henry correlation for sodium aerosol modeling were also identified and documented. The task report is currently pending publication clearance and will be published to OSTI by the end of FY23 [53].

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