Functional Requirements for the Modeling and Simulation of Advanced (Non-LWR) Reactor Mechanistic Source Term

Nuclear Science and Engineering Division

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

DOCUMENT AVAILABILITY

Online Access: U.S. Department of Energy (DOE) reports produced after 1991 and a growing number of pre-1991 documents are available free at OSTI.GOV (<http://www.osti.gov/>), a service of the US Dept. of Energy's Office of Scientific and Technical Information.

Reports not in digital format may be purchased by the public from the National Technical Information Service (NTIS):

U.S. Department of Commerce
National Technical Information Service
5301 Shawnee Rd
Alexandria, VA 22312
www.ntis.gov
Phone: (800) 553-NTIS (6847) or (703) 605-6000
Fax: (703) 605-6900
Email: orders@ntis.gov

Reports not in digital format are available to DOE and DOE contractors from the Office of Scientific and Technical Information (OSTI):

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
www.osti.gov
Phone: (865) 576-8401
Fax: (865) 576-5728
Email: reports@osti.gov

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor UChicago Argonne, LLC, nor any of their employees or officers, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of document authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, Argonne National Laboratory, or UChicago Argonne, LLC.

Functional Requirements for the Modeling and Simulation of Advanced (Non-LWR) Reactor Mechanistic Source Term

Prepared by
Shayan Shahbazi, David Grabaskas
Nuclear Science and Engineering Division, Argonne National Laboratory

June 2020

Acknowledgements

This report is part of a larger collaboration between Argonne National Laboratory (Argonne) and Sandia National Laboratories (SNL) to assist in the development of modern mechanistic source term (MST) modeling and simulation tools, as part of the U.S. Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. The authors would like to thank Nathan Andrews (SNL), Andrew Clark (SNL), David Luxat (SNL), Jennifer Leute (SNL), Jim Jerden (Argonne), Matthew Bucknor (Argonne), and Bo Feng (Argonne) for their support in this effort.

Table of Contents

1	INTRODUCTION.....	1
1.1	PROJECT OBJECTIVES.....	1
1.2	REGULATORY CONSIDERATIONS	2
1.3	ASME/ANS NON-LWR PROBABILISTIC RISK ASSESSMENT STANDARD	3
2	HIGH TEMPERATURE GAS REACTORS.....	4
3	MOLTEN SALT REACTORS.....	13
3.1	SALT-FUELED MOLTEN SALT REACTORS	14
3.1.1	SALT RELEASE.....	14
3.1.2	FUEL PROCESSING SYSTEM RELEASE	24
3.1.3	OFF-GAS SYSTEM RELEASE	27
3.2	SALT-COOLED MOLTEN SALT REACTORS	28
4	SODIUM-COOLED FAST REACTORS	39
4.1	LEAD-COOLED FAST REACTOR CONSIDERATIONS.....	47
	BIBLIOGRAPHY	48

1 Introduction

A number of industry vendors are leading a resurgence in advanced reactor development. As the assurance of public safety from the accidental release of radionuclides to the environment is central to regulatory licensing and the reactor design process, the development of a mechanistic source term (MST) assessment has been identified by the advanced reactor industry as a foremost priority [1]. In contrast to historic light-water reactor (LWR) source term analysis efforts, an MST attempts to realistically model the release and transport of radionuclides from the source to the environment for specific scenarios, while accounting for retention or transmutation phenomena and associated uncertainties. This work seeks to aid in the development of MST modeling and simulation tools by performing a preliminary identification of functional requirements for the major advanced reactor types.

1.1 Project Objectives

The current work is part of a larger collaboration between Argonne National Laboratory (Argonne) and Sandia National Laboratories (SNL) to assist in the development of modern MST modeling and simulation tools, as part of the U.S. Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. To understand the necessary capabilities of the modeling and simulation tools, one must first specify the objective and form basic functional requirements. With the objective to realistically estimate the potential radiological material release from the reactor systems to the environment, the MST development pathway outlined in Figure 1-1 is used for general development guidance. The pathway begins with the identification of radionuclide sources within the reactor-type of interest. This is followed by the identification of potential release, transport, and retention phenomena. Once identified, modeling and simulation tools can be developed to account for those phenomena of high importance, before being utilized as part of regulatory applications.

Figure 1-1: MST Development Pathway

The current work seeks to perform a preliminary assessment of the first two steps of the MST development pathway by identifying sources of radionuclides within advanced reactor designs and outlining potentially influential transport and retention phenomena. The phenomena are then utilized to develop modeling functional requirements, which will be used by a follow-on study

that surveys the capabilities of currently available modeling and simulation tools to identify potential gaps. This report is not meant to be a final assessment of the MST for the reactor concepts reviewed, but rather a starting point for continued modeling and simulation tool development.

For each advanced reactor type, a review of the chemical and physical phenomena related to modeling the source term release was conducted. Corresponding functional requirements for the modeling and simulation of these reactors are outlined. The data and models needed to fulfill those functional requirements are then introduced. The characteristics are outlined in a series of tables for each reactor type. The tables are meant to be a comprehensive overview of possible phenomena that might influence the production and transport of radionuclides in that reactor type, yet they are not meant as a ranking or declaration of relative importance of the phenomena. Additionally, phenomena that may be useful in modeling reactor performance or transient scenarios but which have no impact on the production, release or transport of the source term were not included in the tables.

In each table, the functional requirements needed are divided into different analysis phases. These analysis phases are different for each reactor, but generally include one phase of analysis that involves modeling the inventory of radionuclides produced in the reactor system, one phase of analysis that involves predicting the transport of radionuclides during normal reactor operation, and one or more analysis phases that involve predicting and modeling the transport of radionuclides during various potential accident sequences in different locations of the reactor system. The segregation of functional requirements into these distinct categories was conducted in a manner similar to the historic capabilities of various analysis codes.

The remainder of this section provides insight into current MST regulatory considerations and requirements specified by the ASME/ANS Non-LWR probabilistic risk assessment (PRA) standard. This is followed by separate sections for each of the major advanced reactor types: high temperature gas reactors (HTGRs), molten salt reactors (MSRs), and sodium-cooled fast reactors (SFRs), with insights for lead-cooled fast reactors (LFRs).

1.2 Regulatory Considerations

The U.S. Nuclear Regulatory Commission (NRC) has repeatedly indicated an expectation that future advanced reactor license applications will include a mechanistic assessment of potential radionuclide releases [2-4]. This is further amplified by the NRC commission's recent approval [5] of the risk-informed performance-based licensing framework provided by the Licensing Modernization Project (LMP) [6]. The LMP process is largely dependent on the results of a detailed PRA that includes a mechanistic assessment of offsite dose consequences. In preparation for the submittal of advanced reactor license applications, the NRC has outlined a vision and strategy for computer code development, including source term modeling [7].

1.3 ASME/ANS Non-LWR Probabilistic Risk Assessment Standard

The ASME/ANS PRA Standard for Advanced Non-LWR Nuclear Power Plants [8], which was released for trial use¹ in 2013, sets the requirements for PRAs used in support of risk-informed decisions and provides insight into the necessary characteristics of an MST. The standard defines an MST as:

A source term that is calculated using models and supporting scientific data that simulate the physical and chemical processes that describe the radionuclide inventories and the time-dependent radionuclide transport mechanisms that are necessary and sufficient to predict the source term.

Within the standard, the Mechanistic Source Term element outlines the objectives of the analysis:

- (a) Characterize the sources and inventories of radioactive material to be included in the mechanistic source term analysis within the selected PRA scope. The technical requirements in this trial-use version of the standard are limited to sources of radioactive material within the reactor coolant system pressure boundary (RCPB). Technical requirements for other sources of radioactive material such as the spent fuel system are deferred to future editions of this standard;*
- (b) Identify and address the relevant phenomena that may impact the transport of radioactive material from the radionuclide source to the environment for each modeled event sequence;*
- (c) Define the reactor-specific radionuclide release categories for use in defining the end states of the event sequences and for grouping event sequences in the Event Sequence Analysis (ES);*
- (d) Determine the parameters that define the mechanistic source terms for each radionuclide release category, e.g., quantities of radionuclides released for each species, timing of release, thermal energy of release, physical and chemical form of each species, etc., as needed to support the Radiological Consequence Analysis (RC) and Risk Integration (RI) analysis per the requirements in Secs. 4.5.17 and 4.5.18, respectively;*
- (e) Identify and evaluate the uncertainties in the source term development;*
- (f) Provide documentation for the mechanistic source term analysis*

¹ A revised version of the standard, based on trial use feedback, is expected to be released in late 2020.

2 High Temperature Gas Reactors

Table 2-1 below outlines the MST phenomena in HTGRs. The phenomena reviewed would be applicable to transients such as a primary piping break or core overheating event. The table is divided into four columns: Analysis Phase, Phenomena, Functional Requirements, and Necessary Data or Models. The analysis phases broadly represent the different steps taken in a source term analysis, grouping similar chemical or physical phenomena together. The functional requirements and corresponding data or models needed to accomplish those requirements are listed alongside each phenomenon.

The first two analysis phases include the production and release of radionuclides from the TRISO fuel particles, and the production of radionuclides outside of the fuel (e.g. tritium production via neutron activation of ^{10}B). While fissions inside the fuel kernel are the main production of radionuclides, care must be taken to estimate radionuclides produced via neutron activation of the core materials. These core materials are including but not limited to a graphite fuel matrix, graphite reflectors, carbon bricks, helium coolant, and control rods and absorber balls containing boron and lithium [9]. Recent studies have been performed focusing on the tritium source in HTGR designs [10], including source term analyses specific to irradiated graphite [11]. Additional materials used in a specific core design must be accounted for in a neutron activation analysis with the corresponding thermal neutron fluence. Additionally, there is possibility for heavy metal or uranium contamination outside of the fuel particle coating layers, where source term from fission or absorption reactions are possible [12]. The initial transport pathways of these radionuclides from their source are also outlined in these analysis phases. For fission products inside the fuel kernel, potential pathways of release include diffusion through intact coatings, failed particle coating layers due to normal operation or extreme burn-up, and defective coating layers from manufacturing. Studies have been performed on the irradiation of TRISO particles with fission product analysis, as well as comparing this data to multiple computational codes for the predictive modeling of TRISO particle failure fractions at different burnups and temperatures [13]. While there is considerable data on the chemical behavior and release of cesium, strontium and iodine from TRISO particles [14], there is still a need for further research in these fission products, as well as other relevant fission products such as $^{110\text{m}}\text{Ag}$ [13]. Also important are reactions of the fuel kernel with air or steam during air ingress or water ingress accidents, as defective coatings provide pathways for these reactions to occur [15].

The third analysis phase considered here is the interaction of the radionuclides with core materials, or generally speaking, the transport of the radionuclides between the exterior of the fuel particle and the flow of coolant in the primary coolant system (PCS). An important discovery from early test reactors was the formation of dust particles caused by the wear of graphite by fuel reshuffling or other movements in the core [16]. Dust particles represent sorptive vehicles for radionuclides in the core and can be mobilized to the PCS. Similarly, the sorption of radionuclides by graphite in the core represents an important phenomena in reducing source term release and retaining some radioisotopes in the core. Previous studies have been completed to elucidate mathematical formulations and estimate diffusion and adsorption coefficients for some radioisotopes in graphite or TRISO particle layers, although an updated analysis is still warranted [17, 18].

The fourth analysis phase considered here is the interactions of the radionuclides in the PCS, starting with understanding the chemical speciation of the radionuclide in the coolant, as well as its transport and interactions throughout the primary circuit. Studies have been performed on fission product transport in the primary circuit, focusing on dust behavior throughout the PCS, yet the amount of necessary data concerning this important source term issue has still been deemed insufficient [19]. Similarly, while there are reports on the consequences of water ingress on the primary circuit [20], additional focus is needed on the impact these hypothetical accident sequences might have on the radionuclide source term. Finally, the table is careful to differentiate phenomena that may seem similar in outcome, but are caused by fundamentally different processes. For example, fission products may be desorbed from a PCS surface by “liftoff” during a depressurization incident [12], by an air ingress accident, or simply by a change in the thermodynamics of adsorption due to fluctuations in temperature during normal flow. These distinctions are important to the development of code for modeling source term analysis.

The fifth and sixth analysis phases considered here have to do with the interactions of radionuclides in the reactor building or containment shell of the reactor system, and the transport of the radionuclides from the leak location to the environment. While these can be very similar to those phenomena for both LWRs and other advanced non-LWR reactors, an attempt to summarize and point out key differences has been made.

Although this report does not discuss the development of codes for MST analyses, the outline of the table can motivate potential structure of the code. For example, previous efforts have concluded that the source term analysis of a pebble bed HTGR depends on: reactor analysis (which determines the radionuclide inventory), fuel performance analysis (which determines fission product release), neutron activation analysis of the coolant, and dust composition and circulation [21]. This report suggests similar compartmentalization of phenomena yet notes the importance of inter-phenomena dependencies. Other researchers have noted this importance by combining individual nuclear codes to better estimate source term inventory [22], as well as accident analysis [23].

The Next Generation Nuclear Plant Project (NGNP) is one of the largest recent national efforts in developing an advanced reactor technology, and was focused on a generic design of a HTGR. The outcome of this project was many published reports detailing research and development in the technology, including a white paper on MST development [24]. The authors note the five barriers to radionuclide release include: the fuel particle kernel, the silicon carbide and pyrocarbon coatings of the fuel particle, the fuel matrix and fuel element graphite, the helium pressure boundary, and the reactor building. An MST assessment will require a deeper understanding of the role of each of these barriers in radionuclide retention. A summary diagram of these barriers and the associated phenomena was published in the NGNP white paper and is provided in Figure 2-1. This section hopes to expand on the technology gaps highlighted in the white paper as a supplement to the outlined technology development roadmap. While much research has already been performed in supporting source term analysis of HTGR, identified needs for further research include, but are not limited to, the effects of air and water ingress during accidents, high burnup effects of fuel, radiation damage to core materials, and thermal effects on graphite [25].

Figure 2-1: Summary Diagram for HTGR Radionuclide Retention Phenomena [24]

Table 2-1: Functional Requirements for Modeling Source Term Release in HTGRs

Analysis Phase	Phenomena	Functional Requirements	Necessary Data or Models
Production and Release of Radionuclides from the Fuel Matrix	Chemical forms of fission products in TRISO fuel particle	Ability to predict fission products created, and chemical form of the fission product in the TRISO fuel particle.	Depletion analysis; fission products in TRISO fuel particle; temperature.
	Decay of radionuclides into radionuclide daughters	Ability to account for the decay daughters of all radionuclides, especially noting the half-life and if a change in chemical group occurs.	Specific identification of all relevant radioisotopes of interest with decay chains; thermochemistry data.
	Diffusion of radionuclides in particle layers and coating	Ability to predict release due to diffusion of fission products through each TRISO layer, assuming intact particle coating.	Diffusion coefficients of the chemical form of each fission product in each layer of a TRISO fuel particle
	Particle coating failure due to manufacturing, and associated radionuclide release	Ability to predict fraction of TRISO particles with defective or failed particle coating layer(s); represents a baseline fission product release.	Dependent on manufacturing process and stochastically determined.
	Particle coating failure due to operation at high temperatures	Ability to predict extent and severity of particle coating failure at extreme temperatures, which has direct effect on iodine release, and other FPs.	Experimental data on TRISO particles
	Steam oxidizes UO_2 in particles with defective coating, allowing easier release of volatiles, during steam ingress accident	Ability to predict phase change of UO_2 as it is oxidized in presence of steam which enters due to defective coating. This change in lattice allows more volatile FPs to escape.	Oxidation potential of UO_2
	Steam hydrolyzes UC_x in particles with defective coatings, accompanied by release of some fission products, during steam ingress accident	Assuming a phase of uranium carbide exists in a TRISO particle of UO_2 fuel (or in a TRISO particle where UC_x is expected), ability to predict the reaction of UC_x with steam that enters through a defective coating. (Unexpected UC_x may be present due to failed manufacturing of coatings)	Oxidation potential of UC_x
	Reaction with air causing particle failure or disintegration, during air ingress accident	Ability to predict failure extent and rate of failure for TRISO particle coating layer reaction with air at extreme temperatures.	Irradiation experiments of TRISO fuel in air coupled with fission product release in fuel kernel at very high temperatures.

	Effect of reactivity transients and high burnup on failure of TRISO particle	Ability to predict the extent and rate of failure for TRISO particles during reactivity transients and/or after high burnup, and specifically the impact on FP release.	Experimental data on TRISO particles
	Delayed neutrons and delayed neutron precursors	Ability to track delayed neutrons and delayed neutron precursors.	Coupled neutronics and thermal hydraulic codes, DNP fractions.
	Tritium production due to ternary fission	Ability to predict the amount of tritium produced from ternary fission in the fuel kernel.	Neutronics and depletion analysis; burn-up, fissile nuclides, energy released per fission, thermal and fast neutron fluxes.
	Tritium release from TRISO particle	Ability to predict the release rate of tritium in TRISO fuel.	Fraction of defective particle coatings; diffusion characteristics of tritium in materials in the TRISO particle; temperature; experimental data.
Production of Radionuclides Outside of the Fuel	Production of radionuclides other than tritium via neutron activation of fuel materials, core materials, gas coolant or structural materials	Ability to outline all the materials in the flux of neutrons in the reactor system, including fuel particle coatings, core materials, gas coolant, and structures. Ability to predict the neutron activation of those materials.	Outline all materials in the neutron flux of the core, including fuel, core, coolant and structural, and specifically which isotopes; neutron activation analysis of these materials. Helium may have impurities also.
	Tritium production due to activation of ^3He , ^6Li , ^7Li , ^{10}B	Ability to predict the amount of tritium produced from activation of He, Li, and B nuclei found in the reactor core, typically ^3He in the coolant and Li and B in the graphite, absorber balls, control rods and carbon bricks.	Cross section data, neutronics, concentration and location of activated nuclei, design dependent
	Tritium speciation in coolant or core materials	Ability to predict the chemical form of newly produced tritium in the helium coolant or nearby core materials: HT(g) , $\text{H}_2\text{(g)}$, $\text{T}^+\text{(ion)}$ or other.	Chemical potential and thermochemistry of activation reaction producing tritium in helium coolant or core materials; temperature; hydrogen and helium cover gas partial pressures.
Interaction of Radionuclides with Core Materials	Redox state and chemical form of fission product in core environment (graphite fuel element)	Ability to predict redox state and chemical form of the fission product in the core environment. (Assumed to be very reducing)	Redox chemistry of fission products in graphite at given temperatures.

	Chemisorption of fission products on graphite	Ability to predict chemisorption of Cs, Sr, iodine, etc on graphite; the effect of cosorption, or competing sorption from other fission products, on sorption rates is also important.	Equilibrium vapor pressures of fission products sorbed on graphite; formation enthalpies; diffusion coefficients in graphite; gas flow velocity or residence time over graphite surface; also dependent on accident temperature and fast neutron fluence, but must account for annealing in graphite that occurs at the high accident temperatures.
	Chemical reaction of fission products plated on graphite with H ₂ or CO which are formed when steam attacks graphite	Ability to predict chemical reactions of FPs sorbed onto or into graphite with hydrogen gas and/or CO, which are preferentially formed when steam attacks graphite. The diffusion of fission products further into graphite might occur when steam attacks graphite (gasification of graphite).	Thermochemistry of reactions; sorption status of FPs; partial pressures of gas reactants; temperature.
	Volume expansion due to steam attacking graphite	Ability to predict level of volume expansion due to steam reacting with graphite and producing gaseous products CO and H ₂ .	Thermodynamics; equation of state for gas volume.
	Chemical reaction of fission products with other volatile fission products	Ability to predict chemical reactions that might occur (e.g. CsI formation).	Thermodynamics of reaction; equilibrium vapor pressures over graphite; temperature.
	Effect of irradiation of graphite on sorption capacity	Ability to predict the changing sorption capacity on irradiated graphite over time for each fission product.	Irradiation experiments coupled with sorption experiments for graphite.
	Dust formation and interaction with radionuclides	Ability to predict how dust is formed, how it mobilizes and transports in the core and primary circuit, and how it interacts with fission products.	Identification of the chemical form and source of dust from the fuel matrix and graphite; sorption studies of dust with radionuclides; transport studies of dust in helium flow; largely experimental data.
	Interaction of tritium with graphite in reactor core	Ability to predict the sorption of tritium by graphite in the core, as well as diffusion through graphite.	Experimental data, temperature dependent.

	Interaction of tritium with B ₄ C in the reactor core	Ability to predict the sorption of tritium by B ₄ C after it is produced in control rods.	Experimental data, temperature dependent.
Interaction of Radionuclides in the Primary Coolant System (PCS)	Redox state and chemical form of fission product in primary coolant	Ability to predict redox state and chemical form of the fission product in the helium coolant flow.	Redox chemistry of fission products in helium at given temperatures.
	Adsorption capacity of PCS materials for fission products	Ability to predict the chemisorption and physisorption characteristics of Cs, Sr, iodine in the submonolayer regime on metals with and without an oxide layer.	Material characteristics such as composition and existence of an oxide layer; surface roughness; porosity; type of fission product and chemical form; temperature.
	Plate-out of fission products in heat exchanger or other PCS surface	Ability to predict the plate-out of fission products on heat exchanger or other PCS surfaces.	Thermodynamics of sorption to surface or solid phase formation.
	Equilibrium partition between fission products sorbed in the metals and that in the gas phase	Ability to predict the fission product's equilibrium concentration in the sorbed state on the metal and in the gas phase; enthalpy values associated with returning to solid or sorbed state.	Vapor pressure of fission product in sorbed state; coolant gas phase composition; sorption enthalpy.
	Stability of fission products in volatile state in high-temperature helium carrier	Ability to predict the fission product's stability in the gas phase.	Vapor pressure equilibrium of FP in various chemical forms in He in temperature range in presence of other FPs over graphite or metals.
	Desorption of sorbed fission products due to depressurization in PCS	Ability to predict volatilization or desorption of fission products adsorbed on metals or graphite in primary circuit specifically due to a depressurization of helium.	"'Liftoff' fraction is a function of shear ratio, wall shear stress, blowdown duration, temperature, humidity, and surface oxidation state." [Petti 2013]
	Desorption of fission products from surfaces due to flow of air or steam, during air and/or water ingress accident	Ability to model air flow transport during an air ingress accident, similar to helium depressurization. Also includes mobilization due to simple mass transfer and kinetics, without chemical reaction. Must also factor in transport due to evaporation of water.	CFD analysis of PCS dependent on geometry and leak type and air ingress is important; model shear forces along surfaces mobilizing the FPs; predict chemical reactions of FPs with air.

	Mobilization of fission products plated on metals by reaction with steam from water ingress or concrete decomposition, or reaction with air from air ingress.	Ability to predict the mobilization of fission products from chemisorbed state due to reaction with water and formation of hydroxide or other hydrogen containing compound. Also including chemical reactions with air as well as shear forces along surfaces.	Chemical status and sorption status of FPs; thermodynamics of reaction; equilibrium vapor pressures over graphite; temperature (some experimental data exists for Cs).
	Chemical reaction of fission products plated on metals by H ₂ or CO which are formed when steam attacks graphite	Ability to predict chemical reactions of FPs sorbed onto or into metals with hydrogen gas and/or CO, which are preferentially formed when steam attacks graphite.	Thermochemistry of reactions; sorption status of FPs; partial pressures of gas reactants; temperature.
	Dust mobilization behavior in PCS	Ability to predict how dust mobilizes and transports in the PCS, how plateout occurs, and if it interacts further with fission products. Ability to predict its mobilization specifically due to depressurizations or air ingress.	CFD analysis of depressurization dependent on geometry and leak type is important to give good flow pattern for dust behavior; model shear forces along surfaces mobilizing the dust.
	Permeation of tritium through steel into secondary loop	Tritium can permeate stainless steel tubes of the steam generator at high temperature, therefore it is important to be able to predict this permeation rate.	Experimental data or diffusion modeling.
	Removal of tritium from helium coolant by using Helium Purification System (HPS)	Ability to predict amount of tritium removed from helium flow in a helium purification system (if applicable). HPS can also remove hydrogen, CO, CO ₂ , H ₂ O, O ₂ , N ₂ , CH ₄ , radioactive dust and volatile FPs.	Design dependent.
Chemical Interactions of Radionuclides in the Reactor Building	Oxidation of fission product metals by air	Ability to predict speciation of Cs, Sr, etc as hydroxide, oxide, carbonate, etc after reacting with air: water, oxygen, carbon dioxide.	Thermodynamics of chemical speciation; temperature; partial pressures of air.

	Volatile fission product behavior	Aerosol formation and condensation is not likely due to lower overall releases, therefore the ability to predict the behavior of volatile fission products in elemental or oxidized form is important. Focus on interaction with materials in reactor building, and iodine behavior modeling.	Mobilization by transport processes of air; diffusion through containment; other pathways for escape.
	Transport of radionuclides from the reactor system to the reactor building during depressurization	Ability to predict the initial release of radionuclides into the reactor building consisting of the circulating activity and "liftoff during blowdown".	Combination of the source term circulating in the primary coolant system and the source term that is the fraction "lifted off" during blowdown, dependent on several phenomena described above.
	Transport of radionuclides from the reactor system to the reactor building after depressurization	Ability to predict the delayed release of radionuclides after depressurization due to core heat-up over span of 100+ h. The driving force is the slow heat-up of the core following blowdown.	Design dependent as the gas behavior is mostly a function of natural convection and exchange flow phenomena.
Transport from Leak Location to Environment	Transport of radionuclide aerosols and gases/vapors from leak compartment to environment, or retention within compartment	Ability to predict transport from the leak location to the environment. Possible phenomena include aerosol deposition, vapor/gas condensation, chemical reactions of airborne radionuclides, and plugging of leaks by radionuclide compounds.	Dependent on reactor design, number of containment layers for reactor systems, and specific accident sequence, but likely utilizes temperature/pressure conditions in the compartment and a characterization of the radionuclide.
	Off-site dispersion of radionuclides	Ability to predict off-site dispersion and transport of the radionuclides released from the reactor building into the environment. Transport rates through soil and other surrounding materials.	Physical and chemical form of released radionuclides; Interactions of radionuclide with the environment; Site data and characteristics; Applicable dose conversion factors.

3 Molten Salt Reactors

While there is significant diversity in MSR concepts, from a source term development perspective, the factor of greatest importance is the choice of either a solid fuel or molten fuel salt. Although both salt-fueled (liquid-fueled) and salt-cooled designs share many traits related to coolant chemistry, the presence of a significant radionuclide inventory constantly circulating throughout the core and primary coolant system requires additional considerations. In contrast, salt-cooled MSRs use a fuel specifically designed with radionuclide retention properties, with additional radionuclide barriers between the fuel and coolant. For these reasons, separate analyses were performed for salt-cooled and salt-fueled MSRs. Lastly, for both types of MSRs, the production of tritium may be an additional concern depending on the salt form used, and requires additional mitigation and capture strategies [26]. A brief review of literature relevant to source term analysis for salt-fueled MSRs is provided.

Several efforts have provided overviews of the safety characteristics of MSRs and begun safety analyses using techniques such as HAZOP studies, Functional Failure Mode and Effects Analysis (FFMEA), and Master Logic Diagram (MLD) [27-29]. Other efforts have also completed safety assessments using methodologies published from IAEA's International Project on Innovative Nuclear Power Reactor and Fuel Cycles (INPRO) [30]. Published safety analyses of MSRs using an assortment of nuclear codes, often coupled with thermal hydraulic codes, are available in refs [31-39]. Some of these efforts have shown that source term functionality can be added to these codes by tracking specific fission product isotopes with the flow [40-43]. Additionally, Diamond et al. have tabulated phenomena important to neutronic and thermal hydraulic modeling in molten salt reactors [44].

McFarlane et al. have performed considerable work in characterizing the chemistry of fission products, especially for volatile species transported to the off-gas system [45, 46]. Greenwood et al. have developed a code utilizing a flexible library of models that can be used for tracking source term as trace substances in MSR, and they note future work may include incorporating chemistry processes into the models [47]. Hu et al. have been developing the SAM code for MSR transient analyses, adding capabilities of a Point-Kinetics model with delayed neutron drift, as well as modeling decay heat generation and reactivity feedback in the core [48-51]. They also benchmarked the newly developed code against MSRE transient experimental data which showed good agreement, but noted slight differences may be due to the absence of a core neutronics model in the code [52, 53]. There are ongoing efforts in developing MELCOR for modeling of various accident scenarios for advanced reactors, including MSRs [7]. Objectives for MELCOR development for non-LWRs include providing “a tool capable of performing severe accident progression modeling and source term characterization while allowing the capability for performing uncertainty analyses.” Recently, Betzler et al. completed a high-level assessment on the functional needs for modeling and simulation for MSR licensing, noting interdependencies between the functional needs and prioritizing them by importance [54]. They reported that the functional needs with the highest priority and impact on other functional needs were: fuel depletion, mass transport, heat transport, steady-state core neutronics, salt thermochemistry, source term accountability, and salt thermophysical properties. Similarly, they reported that the highest priority inputs to the functional needs were: salt thermophysical properties, salt constituent volatilities, salt constituent solubilities, mass inventories and corresponding distributions, and chemical interactions. Finally, Jerden et al. have published on ongoing efforts in

providing a basis for developing a MSR source term model, recommending a pathway that is both rigorous and technology-neutral [55].

3.1 Salt-Fueled Molten Salt Reactors

One of the main advantages of salt-fueled MSRs is that having a mobile fuel form greatly simplifies decay heat removal during accidents, allowing developers flexibility in the design of passive decay heat removal systems. Additionally, part of the appeal of salt-fueled MSRs is their versatility and diversity in design options. By having a liquid fuel, the developer is left with many design choices in terms of fuel type, fuel salt, coolant salts, moderator materials, core design, and even the neutron energy spectrum. These choices allow developers to design reactors to accomplish different objectives whether related to economics, safety, security, grid optimization, refueling, operations and maintenance, mass production, or producing energy forms other than electricity. Conversely, this diversity also makes it difficult to construct generic analyses. With the radionuclide inventory of the reactor distributed through the primary circuit and associated support systems, multiple potential release pathways were reviewed for the current assessment. A summary diagram of the physical and chemical phenomena for radionuclide transport in salt-fueled MSRs has been provided in Figure 3-1 [56].

Recently, ORNL hosted a workshop with the goal of identifying initiating events (IE) for radioactive releases as precursors to licensing basis events (LBE) for salt-fueled MSRs [57]. They noted 140 different IEs that warranted further investigation. Some of these high-level events selected for extended discussion include: primary heat exchanger failures, primary boundary breaches, primary fuel salt composition changes, primary fuel salt void fraction changes, drain tank heat removal failure, drain tank breaches, and off-gas system breaches/failures. Similar events were considered while assessing the modeling functional requirements for MSRs in this report. First, a salt release event was assessed, which includes scenarios such as leaks or breaches from the primary circuit or drain tank in the reactor building, as well as a heat exchanger rupture or failure. Second, potential release avenues from an on-line fuel processing system were also reviewed. Lastly, although not documented in a separate table, possible releases from an off-gas system were also examined and are briefly discussed.

3.1.1 Salt Release

Table 3-1 below outlines the MST phenomena for a salt release transient in a salt-fueled MSR. This includes a potential salt spill from the reactor vessel, primary circuit piping, or heat exchangers. The first two analysis phases include the production and behavior of radionuclides, respectively. While the production of radionuclides for most reactors is concentrated in one location, radionuclides can be produced anywhere in the core or coolant circuit for MSRs. Although this makes mechanistic source term analysis more challenging, the approach taken here is to group similar phenomena together and analyze each phase sequentially. Although fissions will be the largest source of produced radioactivity, the fuel salt may also contain isotopes of elements prone to neutron activation. It is not only important to account for this production in the fuel salt, but also for the primary coolant, which comes in close proximity to the neutron flux of the fuel salt in the heat exchanger, as well as for a blanket breeder salt if used. After the radionuclide source term is estimated, speciation of each radionuclide in the corresponding salt can be modeled, for example, by using thermochemical models [58]. Additional phenomena

related to the behavior of radionuclides inside the MSR system include corrosion [59-62], transport of gaseous species through the salt [46], phase stability of species initially soluble in the salt [63-65], and other interactions. All of these phenomena in the first two analysis phases set up the initial conditions of the source term in the reactor.

The third analysis phase deals with the behavior of tritium in the reactor system. Starting with establishing the speciation in the coolant salt, tritium can be modeled as it interacts with other chemical species in the core and coolant, or transports as elemental or ionic tritium through metallic structures. Tritium behavior phenomena are separated into their own analysis phase because it may be beneficial to model tritium transport, mitigation and capture separately from other radionuclides due to an already complex source term circulating in MSRs. Additionally, tritium does not behave like any of the other radionuclides as it is not as easily contained by defense barriers and poses additional challenges in modeling. Simulating tritium source term transport is probably best accomplished in dedicated codes, as has been completed for the FHR [66].

The fourth and fifth analysis phases here are specific accident scenarios that are grouped separately because they may be best modeled independently, just as with tritium behavior. The first of these accident sequences is a generic salt spill or leaking of molten salt from somewhere in the reactor system into the reactor building. Much of this analysis phase is design dependent, but as much detail as possible is provided. An additional phenomenon important here is not only the leaking of radioactivity to the exterior of the system, but the ingress of air to the interior of the reactor, which may cause additional safety concerns. The second accident sequence is a generic rupture in the primary heat exchanger, where the primary fuel salt will likely mix with the primary coolant salt. Phenomena of note here include understanding the pressure gradient between the two salt flows as well as considering any chemical reactions that may occur between the salts and components.

The sixth analysis phase is the transport from the leak location to the environment. This may be similar as in other reactor types, except with special focus on tritium transport, and the behavior of halide salts in atmosphere, as well as hydroxides and oxides.

Figure 3-1: Summary Diagram for Salt-fueled MSR Radionuclide Transport Phenomena [56]

Table 3-1: Functional Requirements for Modeling Source Term Release in MSRs

Analysis Phase	Phenomena	Functional Requirements	Necessary Data or Models
Production of Radionuclides	Fuel depletion and fission product yields	Ability to predict fission product yields based on fissile element and core average energy spectrum.	Fissile element nuclear data, neutron energy spectrum, delayed neutrons and delayed neutron precursors, fissile element addition/breeding, time, temperature.
	Decay of radionuclides into radionuclide daughters	Ability to account for the decay daughters of all radionuclides, especially noting the half-life and if a change in chemical class occurs, affecting the speciation and transport.	Specific identification of all relevant radioisotopes of interest with Bateman decay chains; thermochemistry data.
	Neutron activation products of fuel salt, moderator, core materials, and structural materials	Ability to predict neutron absorption reactions for all salt components including actinides, moderator materials such as control rods, core structural materials, and reflector and shielding materials for fast spectrum.	Cross section data for neutron absorption reactions for all isotopes of all elements of materials, neutron energy spectrum and flux, delayed neutrons and DNPs, time, temperature.
	Neutron activation products of primary coolant salt and blanket salt	Ability to predict neutron absorption reactions for all components in the primary coolant salt and blanket salt circuit (if applicable) including actinides and other salt components, structural materials, and potential impurities.	Cross section data for neutron absorption reactions for all isotopes of all elements of materials; neutron energy spectrum and flux; delayed neutrons and DNPs are especially important; flow characteristics; temperature.
	Delayed neutrons and delayed neutron precursors	Ability to tracking of delayed neutrons and delayed neutron precursors.	Coupled neutronics and thermal hydraulic codes; DNP fractions.
	Tritium production due to ternary fission	Ability to predict the amount of tritium produced from ternary fission in the fuel salt based on burn-up, fissile nuclide, energy released per fission, thermal power of reactor.	Neutronics and depletion analysis.
	Tritium production due to activation of fuel salt, coolant and core materials	Ability to predict the amount of tritium produced from activation of Li, Be, B, F nuclei found in the reactor core and fuel salt, typically F/Li/Be in the fuel salt, Li/Be/B in the graphite, absorber balls, control rods and carbon bricks.	Cross section data; neutronics; concentration and location of activated nuclei (design dependent).

Behavior of Radionuclides	Chemical form of radionuclides in the salt	Ability to predict the chemical form of each isotope involved in the molten salt mixture including chemical speciation.	Speciation of isotope in salt; salt redox potential; species chemical potentials; thermodynamic and kinetic data; temperature; pressure.
	Chemical reactions between fission products or other radionuclides	Ability to predict likely reactions occurring between common fission products or other activation products, such as cesium iodide.	Gibbs free energy of formation; thermodynamic and kinetic data; temperature; pressure.
	Physical form of radionuclides in the salt	Ability to predict the initial phase of the isotope in its chemical form and understand likely phase changes. Ability to predict solubility of the isotope in the salt mixture.	Phase diagrams; vapor pressure equilibrium data; salt solubility data; thermodynamic and kinetic data; salt temperature and pressure.
	Vaporization behavior and phase stability of chemical species in the salt	Ability to predict if chemical species initially stable in the salt can be easily vaporized out of the salt at varying compositions and temperatures.	Species chemical potential; species solubility and vaporization behavior; thermodynamic and kinetic data; temperature; pressure.
	Fluorination of metals in the salt	Ability to predict the production of free fluorine gas or other gaseous fluorides which can volatilize initially solid or liquid metals or other radionuclides. Ability to predict the transport of those volatile metal fluorides.	Identifying volatile metal fluorides that may be present such as UF_6 , MoF_6 , etc; fluorination experimental data for various gaseous fluorides; effect of temperature and redox chemistry of salt on the production of fluorine gas or other free fluorine ions; temperature fluctuations.
	Corrosion	Ability to predict the exchange of stable metal atoms from structural materials into salt solution with metals or other ions out of salt solution.	Electrochemistry and redox potentials; thermodynamic and kinetic data; chemical potentials; temperature; identification of materials in contact with metal ions.
	Isotope exchange of chemical species with core materials	Ability to predict the isotope exchange rates of key radioisotopes with stable isotopes of structural or core materials due to corrosion or similar redox reactions.	Isotope exchange rates; salt redox potential; corrosion reactions; thermodynamic and kinetic data; temperature.
	Precipitation or plating of chemical species	Ability to predict precipitation of chemical species on surfaces of vessel, piping, heat exchanger and other core materials.	Rate of deposition (kinetics, thermodynamics); sorption thermodynamics.

	Volatile chemical species transport	Ability to predict transport of volatile chemical species in salt mixture and bubble transport through salt to the off-gas system. Important to note some fission products have noble gas precursors such as ^{137}Cs and ^{90}Sr , so while the FP may not be assumed to be in the off-gas system initially, it may end up there after decay from a volatile precursor.	Thermodynamics; phase diagrams; modeling bubble formation with transport equations; fission product yields of volatile species, while tracking decay of those isotopes with interactions in the salt and in the off-gas system. Potentially beneficial to separate volatile species in different classes of volatility and half-life.
	Mechanical agitation of salt caused by normal pump operation	Ability to predict the effect that the pump has on the mechanical agitation of the salt and its loosely bound components. This agitation may have a sort of "ultrasonic effect" on the salt, easing transport or dissolution.	Sorption thermodynamics of volatile or insoluble species in salt; specific experiments focused on agitation of molten salt mixtures; momentum and mass transport phenomena.
	Salt-insoluble chemical species transported via sweep gas	Ability to predict radionuclides formed that are insoluble in the salt mixture but may be transported via sweep gas to the off-gas system and/or plate-out along the way as particulates. Ability to model particulate size and transport of components such as Nb, Mo, Tc, Ru.	Thermodynamics of formation and chemical speciation; particulate formation; transport via gaseous flow or volatilization; plating on pipe or vessel walls dependent on temperature and flow characteristics; movement in off-gas system is design dependent.
	Aerosolized salt transported via sweep gas	Ability to predict the formation of fuel salt mist in the sweep gas and transported to the off-gas system or elsewhere. Ability to model fuel salt phase change via aerosolization. Ability to model changes in gas phase parameters of sweep gas as it moves through salt, resulting in possible condensation of aerosol back into salt.	Phase diagrams of molten salts; salt aerosol formation and kinetics; gas phase composition; temperature; pressure; experimental data.

	Off-gas system	Ability to predict chemical speciation in each stage of the off-gas system for radionuclide accountability during normal operation. Ability to model escape routes for radionuclides in each stage. Common stages include a hydroxide scrubber, silver mordenite aerogel trap, tritium trap, and other traps or delay beds.	Thermodynamic and kinetic data; design dependent.
Behavior of Tritium	Tritium speciation in coolant salt	Ability to predict the chemical form of newly produced tritium in the coolant salt or nearby core materials: TF, HT(g), H ₂ (g) or T ⁺ (ion). Ability to predict chemical reaction with other radionuclides, such as iodine.	Chemical potential and thermochemistry of activation reaction producing tritium in salt; salt composition; temperature; cover gas or sweep gas partial pressures of hydrogen or other gases.
	Interaction of tritium with graphite in reactor core	Ability to predict the sorption of tritium by graphite in the core, as well as diffusion through graphite.	Experimental data; temperature dependence.
	Permeation of tritium through metals in core	Ability to predict the transport of tritium (typically HT/T ₂) through metallic structures and components.	Experimental data on tritium gas transport in metals; diffusion modeling.
	Interaction of tritium with B ₄ C in the reactor core	Ability to predict the sorption of tritium by B ₄ C after it is produced in the control rods.	Experimental data; temperature dependence.
	Interaction of tritium with coolant salt	Ability to predict the sorption of tritium by the coolant salt, diffusion and transport through the salt, and potential escape into the cover gas. TF and T ⁺ may stay dissolved in the salt or attack metallic surfaces, while most HT/T ₂ will diffuse through the salt and escape into the cover gas unless oxidized.	Dependent on chemical speciation of tritium: thermochemistry, salt composition, temperature, hydrogen and helium cover gas partial pressure.
	Permeation of tritium through metals in heat exchanger into the secondary loop	Tritium can permeate metal surfaces of the heat exchanger as well as stainless steel tubes of the steam generator at high temperature, therefore it is	Experimental data; diffusion modeling.

		important to be able to predict this permeation rate.	
	Removal of tritium from salt coolant	Ability to predict amount of tritium removed from salt coolant using a salt purification system, sacrificial carbon anode, or a sweep gas.	Design dependent.
Accident: Salt Spill in the Reactor Building	Volatilization of chemical species from the salt spill	Ability to predict the volatilization of species in the salt that spills into the building which holds the reactor systems. Ability to account for changes in pressure of containment (i.e. containment pressure changing with time affects volatilization rate).	Vapor pressure determination of all chemical species in the salt mixture within a range of temperatures; dissociation kinetics and thermodynamics of various salt systems in air at ambient temperatures and pressures.
	Aerosolization of salt from the salt spill	Ability to model the splash radius of the salt and aerosol creation as it leaks into the environment surrounding the pipe/MSR component, as well as aerosolization due to falling liquid.	Conditions associated with salt spill, such as fall height, temperature, etc. Will be design/accident dependent.
	Fluorination of metals in the salt as it cools	Ability to predict the production of free fluorine gas or other gaseous fluorides which can volatilize initially solid or liquid metals or other radionuclides as the spilled salt cools. Ability to predict the transport of those volatile metal fluorides in containment.	Identifying volatile metal fluorides that may be present such as UF_6 , MoF_6 , etc; fluorination experimental data for various gaseous fluorides; effect of temperature and redox chemistry of salt on the production of fluorine gas or other free fluorine ions; temperature during cooling.
	Passive heat removal	Ability to predict the rate at which decay heat and contained heat is removed and the salt freezes in the catch pan. Ability to predict how much transport occurs until that point, whether to drain tank (if connected to one) or catch pan.	Heat contained in salt can be estimated by salt temperature at breach and rate of heat generation dependent on radioisotope composition; rate of heat removal dependent on temperature of salt and methods of heat transport to surroundings: radiative to environment, conductive to catch pan surface and steel below, convective to natural or forced circulation of air.

	Degradation and failure of catch pan due to molten salt	Ability to predict failure rate of catch pan due to conductive and convective heat transfer from molten salt.	Material properties of catch pan; conductive and convective heat transport phenomena modeling; temperature of molten salt exiting leak.
	Deformation of structural material below catch pan due to molten salt	Ability to model heat transfer from molten salt to structural material and corresponding deformation of that material (possibly steel or concrete).	Material properties of the structural material; conductive and convective heat transport phenomena modeling; temperature of molten salt exiting leak from deformed catch pan.
	Chemical reaction of molten salt with structural components, air, or water during salt spill and severe accident	Ability to predict potential reactions of molten salt components with structural components (possible steel or concrete), air, and/or water during salt spill and air or water ingress.	Reaction thermodynamics, molten salt composition, salt temperature, alloy or concrete composition, air composition.
	Delayed leaking of volatile species from breached reactor flow path	Ability to calculate and predict volatile species produced after breach has already occurred.	Reactivity model to predict delayed FP production, thermodynamics, kinetics of desorption of volatiles from salt and vaporization.
	Reaction of air with core components and salt during air ingress	Ability to predict potential reactions of core components or molten salt with air inside the reactor. Ability to predict volatilization of UF ₆ and associated criticality considerations.	Salt redox potential, reaction thermodynamics, molten salt composition, state of graphite in core, salt temperature, air composition, UF ₆ volatilization after air interaction.
Accident: Heat Exchanger Rupture	Volatilization of chemical species from fluid of higher pressure to lower pressure	Ability to predict volatilization of chemical species in each fluid with transfer into the other, if the pressure gradient favors that	Salt composition, coolant composition, vapor pressure data, temperature, pressure.
	Mixing of molten fuel salt into primary coolant	Ability to predict miscibility of fuel salt with coolant salt; ability to predict partitioning and speciation of chemical species in each fluid	Salt composition, coolant composition, thermodynamics, temperature, pressure.
	Pressurization due to chemical reaction between fuel salt and primary coolant	Ability to predict chemical reactions occurring between fuel salt and primary coolant, producing volatile species, leading to increased pressure.	Salt composition, coolant composition, thermodynamics, temperature, pressure.

	Transport of transferred chemical species or immiscible fuel salt particles through primary coolant loop due to lack of pump shutdown	Ability to model transport of dissolved chemical species or immiscible fuel salt particles in the primary coolant loop due to either forced flow from a pump or natural circulation.	Thermal hydraulics, temperature, pressure
	Leaking of chemical species from primary coolant to steam coolant	Ability to predict volatilization or transfer of radionuclides in coolant salt to steam side	Coolant composition; steam generator design; radionuclide species vapor pressure data; temperature; pressure; diffusion.
	Leaking of chemical species from steam coolant to environment outside MSR system	Ability to predict transport of chemical species from steam to environment outside MSR system.	Steam generator design; radionuclide species vapor pressure data; temperature; pressure; diffusion.
	Reaction of air with core components and salt during air ingress	Ability to predict potential reactions of core components or molten salt with air inside the reactor.	Reaction thermodynamics, molten salt composition, state of graphite in core, salt temperature, air composition.
Transport from Leak Location to Environment	Transport of radionuclide aerosols and gases/vapors from leak compartment to environment, or retention within compartment	Ability to predict transport from the leak location to the environment. Possible phenomena include aerosol deposition, vapor/gas condensation, chemical reactions of airborne radionuclides, and plugging of leaks by radionuclide compounds.	Dependent on reactor design, number of containment layers for reactor systems, and specific accident sequence, but likely utilizes temperature/pressure conditions in the compartment and a characterization of the radionuclide.
	Off-site dispersion of radionuclides	Ability to predict off-site dispersion and transport of the radionuclides released from the reactor building into the environment. Transport rates through soil and other surrounding materials.	Similar to that for other nuclear reactor designs (see HTGR table) with additional focus on tritium and fluoride salts.

3.1.2 *Fuel Processing System Release*

Salt-fueled MSRs have the unique option of having on-line fuel processing systems, if the developers so choose to utilize it. This capability can be used for fissile material control and fission product removal for benefits in either neutronics, thermal hydraulics, isotope production, or waste optimization and disposal. It is also an essential component of molten salt breeder reactor concepts. While an on-line processing system can be useful by reducing the circulating source term inside the reactor core, the radionuclides still exist in a different area of the reactor system. The system nonetheless requires an MST assessment to understand potential pathways for radionuclide release during transient scenarios. Another important use of a source term model for the fuel processing system would be to better understand feedback effects of the processing system on the reactor neutronics and thermal hydraulics. Potential failures in the fuel processing system's ability to remove certain isotopes could have serious effects on the performance of the salt-fueled reactor. Therefore, the development of a model for analysis of the fuel processing system has multiple motivations.

While little public information is available from MSR vendors regarding the design of chemical fuel processing system for a salt-fueled MSR, past design efforts are available. The processing system for the fuel salt of the single-fluid Molten Salt Breeder Reactor (MSBR) is used here as an example of how one might characterize each chemical processing step in terms of functional requirements and necessary data for modeling [67-69]. The process generally uses alternating stages of fluorination (to remove volatile fluorides) and reductive liquid-liquid extraction (to remove or transfer specific metals), with molten salt and liquid bismuth as the two immiscible phases. Table 3-2 below contains only one analysis phase, which represents the different stages of separations in the fuel processing system for MSBR.

Table 3-2: Functional Requirements for Modeling Source Term in the Fuel Processing System of MSBR

Analysis Phase	Phenomena	Functional Requirements	Necessary Data or Models
Fuel Processing Stages of Separations	1A: Fluorination of UF_4 in unprocessed molten salt to UF_6	Ability to predict reaction yields in volatilizing ^{233}U from molten salt as well as estimate yields of other radioisotopes removed in this fluorination step when using F_2	Salt composition at inlet, temperature, pressure, thermodynamic data
	1B: Purification of UF_6 from volatile fission products	Ability to predict adsorption of fission products onto NaF trap; verify passage of UF_6 through trap dependent on temperature.	Salt composition at inlet, temperature, pressure, adsorption data, thermodynamic data
	1C: Addition of UF_6 to processed salt (leaving 2B) or removal for storage	Ability to predict adsorption of UF_6 into processed molten salt, and reduction to UF_4 with H_2 gas	Gas composition at inlet, processed salt composition at inlet, temperature, pressure, adsorption data, thermodynamic data
	2A: Exchange of Pa/U into liquid Bi/Li/Th from molten fluoride salt containing FPs	Ability to predict distribution coefficients for Pa/U in reductive extraction between molten salt and liquid bismuth phases	Salt composition at inlet, temperature, pressure, activity or equilibrium constants, or distribution coefficients for reductive extraction reaction
	3A: Hydrofluorination of Pa/U into secondary molten fluoride salt from liquid Bi/Li/Th	Ability to predict reaction yields in oxidizing Pa/U from Bi stream to secondary fluoride salt stream; assumption is complete oxidation	Liquid Bi composition at inlet, temperature, pressure, thermodynamic data, gas-liquid reaction with equilibrium constants
	3B: Fluorination of UF_4 in secondary fluoride salt (containing U/Pa/others) to UF_6	Ability to predict reaction yields in volatilizing ^{233}U from molten salt as well as estimate yields of other radioisotopes removed in this fluorination step when using F_2	Salt composition at inlet, temperature, pressure, thermodynamic data
	3C: ^{233}Pa decay to ^{233}U while contained in secondary fluoride salt	^{233}Pa decays to ^{233}U in a storage tank, other radioisotopes will decay also: simple decay and buildup of isotopes	Salt composition at inlet, Bateman equations
	3D: Fluorination of newly produced UF_4 in secondary fluoride salt (containing U/less Pa/others) to UF_6	Ability to predict reaction yields in volatilizing ^{233}U from molten salt as well as estimate yields of other radioisotopes removed in this fluorination step when using F_2	Salt composition at inlet, temperature, pressure, thermodynamic data

	3E: Remaining salt waste stored in tank	Ability to predict chemical form and composition of fluoride salt waste stream leaving stage 3D and decay of FPs	Salt composition at outlet of 3D, temperature, pressure, thermodynamic data
	2B: Exchange of REE/Th into liquid Bi/Li stream from molten salt leaving Stage 2A	Ability to predict distribution coefficients for REE/Th in reductive extraction between molten salt and liquid bismuth phases	Salt composition at inlet, temperature, pressure, activity or equilibrium constants, or distribution coefficients for reductive extraction reaction
	2C: Exchange of REE into molten LiCl salt from liquid Bi/Th	Ability to predict distribution coefficients for REE and other elements present in reductive extraction between molten LiCl salt and liquid bismuth phases	Liquid Bi composition at inlet, temperature, pressure, activity or equilibrium constants, or distribution coefficients for reductive extraction reaction
	2D: Exchange of trivalent REE into liquid Bi/0.05 mol% Li from molten LiCl salt	Ability to predict distribution coefficients for REE and other elements present in reductive extraction between molten LiCl salt and liquid bismuth phases	Salt composition at inlet, temperature, pressure, activity or equilibrium constants, or distribution coefficients for reductive extraction reaction
	2E: Exchange of divalent REE into liquid Bi/0.5 mol% Li from molten LiCl salt	Ability to predict distribution coefficients for REE and other elements present in reductive extraction between molten LiCl salt and liquid bismuth phases	Salt composition at inlet, temperature, pressure, activity or equilibrium constants, or distribution coefficients for reductive extraction reaction
	2F: Remaining salt waste stored in tank	Ability to predict chemical form and composition of fluoride salt waste stream leaving stage 2E and decay of FPs	Salt composition at outlet of 2E, temperature, pressure, thermodynamic data
	4A: Storage of trivalent REE in liquid Bi waste stream	Ability to predict chemical form and composition of liquid bismuth waste stream leaving stage 2D and decay of FPs	Liquid Bi composition at outlet, temperature, pressure, thermodynamic data
	4B: Storage of divalent REE in liquid Bi waste stream	Ability to predict chemical form and composition of liquid bismuth waste stream leaving stage 2E and decay of FPs	Liquid Bi composition at outlet, temperature, pressure, thermodynamic data

3.1.3 *Off-Gas System Release*

As mentioned previously, a description of a prototypical off-gas system for MSRs was given by McFarlane et al. along with an introductory discussion on the associated source term [45, 46]. A summary of that off-gas scrubber system is given here, followed by a brief discussion of a source term analysis approach.

The first stage of the off-gas system is being designed as a molten hydroxide scrubber, utilizing a eutectic mixture of NaOH-KOH, which will effectively remove aerosols and acidic gases. It should also dissolve alkaline species, noble metals and halogens. The second stage of the off-gas system might be a trap for long-lived halides by using silver mordenite or silver-impregnated aerogels. The third stage suggested is a tritium trap, as some tritium that enters the hydroxide scrubber will react with hydroxide ion and form water, eventually evaporating and transporting out of this first stage. As mentioned elsewhere in this report, tritium is a major issue for many reactors designs, and a dedicated tritium source term analysis may be warranted as it can be very design dependent. The next stage suggested is an oxygen gas trap, which is used for impurity control of the off-gas system's carrier gas of helium. The final stages of the off-gas system are for purifying the helium from the radionuclide noble gases. Initially, activated charcoal beds can be used to allow as many short lived isotopes to decay. Beyond this, longer-lived noble gases can be separated from the helium carrier using more costly industrial techniques such as pressure swing adsorption or cryogenic distillation.

In every stage of the off-gas system, the most important phenomenon will be the chemical speciation and stability of the radionuclide in that type of trap or separations process. The ability to predict what chemical form that radionuclide forms will influence how stable it is in that phase at the given temperature and pressure, as well as any chemical interactions it may have with other radionuclides. The ability to model the off-gas system's effectiveness is instrumental to modeling the source term. Possible outcomes for a radionuclide include being trapped in the system for an extended period of time, being removed from the system via a waste stream after trapping, escaping the off-gas system through unexpected pathways, decaying to a stable isotope, or returning to the reactor with the helium carrier. Understanding the likelihood of all of these pathways will be important for all radionuclides that enter the off-gas system inlet stream. At every stage of the off-gas system, an individual mechanistic source term analysis will need to occur, as every stage utilizes a different separations technique based on different chemistry. Potential releases during core accident scenarios (or otherwise) are consequently very design dependent.

3.2 Salt-Cooled Molten Salt Reactors

One of the most widely reported salt-cooled MSR designs is the fluoride salt-cooled high temperature reactor (FHR) concept that was predominantly developed by researchers at University of California, Berkeley, MIT, University of Wisconsin, Madison, and Oak Ridge National Laboratory, along with support from other universities, national laboratories and industry [70]. A series of four white papers were released in 2013, providing a comprehensive review of the technology developed thus far for the FHR concept [71-74]. Additionally, considerable work has recently been completed in identifying important phenomena and knowledge gaps for neutronics, thermal hydraulics, and materials design for the modeling and simulation of a FHR [75-79], although the development of a mechanistic source term assessment has yet to be published. A summary diagram for radionuclide release paths in a generic salt-cooled molten salt reactor is provided in Figure 3-2 [7].

Table 3-3 below outlines the phenomena important for modeling source term release in fluoride salt-cooled high temperature reactors (FHR). Similar to that for HTGR, the first two analysis phases include the production and release of radionuclides from the TRISO fuel particles, and the production of radionuclides outside of the fuel (e.g. tritium production via neutron activation of ^6Li). While fissions inside the fuel kernel are the main production of radionuclides, care must be taken to estimate radionuclides produced via neutron activation of the core materials and coolant. By using a fluoride salt mixture as coolant, additional care must be taken in a source term analysis compared to that for HTGR. There are more pathways to tritium production than compared to an equivalent HTGR design, which uses helium as a coolant [80, 81]. Additionally, many fission products are chemically reactive with fluoride ion and soluble in fluoride salts, thereby potentially increasing the circulating activity of a FHR relative to a HTGR. Recent studies on the chemistry of fluoride salts in FHR have highlighted additional gaps in knowledge such as the impact of impurities on reactor chemistry and corrosion [82], and the compatibility of fluoride salts with silicon carbide in TRISO fuel [83]. Although corrosion does not always directly impact the source term, the addition of new compounds into the molten salt coolant changes the chemistry, which could have a considerable effect on source term analysis.

The third analysis phase considered here is the interaction of the radionuclide source term with core materials, or generally speaking, the transport of the source term between the exterior of the fuel particle and the flow of coolant in the primary coolant system (PCS). Similar to HTGR, the formation of dust from graphite wear in the core, potentially due to fuel reshuffling, can be an issue with FHR [84, 85]. As the addition of graphite dust to the salt can have neutron-moderating qualities, special care must be taken in exploring this phenomenon further, not just for source term analysis. It is also worth noting that graphite, ceramic and structural materials are likely exposed to a higher neutron flux in FHR than compared to HTGR [71, 86], and therefore the effect of irradiation on the radionuclide sorption characteristics of these materials is an important phenomena to investigate further.

The fourth analysis phase considered here is the interactions of the radionuclide source term in the PCS, starting with understanding the chemical speciation of the radionuclide in the salt coolant, as well as its transport and interactions throughout the primary circuit. The relevant phenomena in this phase are more closely aligned with those of the liquid-fueled molten salt reactor (MSR), as the generic designs of each primary circuit outside of the core should be near

identical. The chemical and physical interactions of radionuclides in the fluoride salt coolant flow with the metal surfaces of the PCS should be similar for both reactors, other than the fact that the salt chemistry for MSR is changing more frequently, with a more varied composition. A major difference between the FHR and HTGR in this analysis phase might be the inclusion of a sweep gas to clean the salt of volatile components, maintain tritium control, or serve another function [71]. Sparging some gas component or mixture can be beneficial to the redox control, corrosion control, or impurity removal, but adds additional levels of complexity to a source term analysis with more phenomena to model in the PCS, and more escape pathways for the source term.

The fifth and sixth analysis phases considered here have to do with the interactions of radionuclides in the reactor building or containment shell of the reactor system, and the transport of the radionuclides from the leak location to the environment. While these can be very similar to those phenomena for both light water reactors (LWR) and other advanced non-LWR reactors, an attempt to summarize and point out key differences has been made. Similar to MSR, additional focus is needed on an accident scenario that involves the potential leaking of coolant salt from the reactor system inside the reactor building, although the salt is holding considerably less radioactivity than its liquid-fueled counterpart.

Computational fluid dynamics (CFD) analyses have been performed for a FHR [87], but similarly for salt-fueled MSR, any source term analysis of the primary circuit should involve modeling the flow of salt in conjunction with modeling the relevant phenomena that may be occurring in the PCS. Researchers have utilized existing codes for modeling of a FHR demonstration reactor [88], but future work in source term analysis will need to combine the utility of multiple codes with focused analysis of the chemistry of radionuclides in FHR. A comprehensive model for estimating the circulating source term can be useful for more than just source term analyses, as studies have shown the impact even small amounts of fission product impurities in the coolant salt have on reactivity and achievable burnup [89].

Salt-cooled reactors**Environment**

Figure 3-2: Summary Diagram for Salt-cooled MSR Radionuclide Retention Phenomena [7]

Table 3-3: Functional Requirements for Modeling Source Term Release in FHRs

Analysis Phase	Phenomena	Functional Requirements	Necessary Data or Models
Production and Release of Radionuclides from the Fuel Matrix	Chemical forms of fission products in TRISO fuel particle	Ability to predict fission products created, and chemical form of the fission product in the TRISO fuel particle.	Depletion analysis; fission products in TRISO fuel particle; temperature.
	Decay of radionuclides into radionuclide daughters	Ability to account for the decay daughters of all radionuclides, especially noting the half-life and if a change in chemical group occurs.	Specific identification of all relevant radioisotopes of interest with decay chains; thermochemistry data.
	Diffusion of radionuclides in particle layers and coating	Ability to predict release due to diffusion of fission products or actinides through each TRISO layer, assuming intact particle coating.	Diffusion coefficients of the chemical form of each radionuclide in each layer of a TRISO fuel particle.
	Particle coating failure due to manufacturing, and associated radionuclide release	Ability to predict fraction of TRISO particles with defective or failed particle coating layer(s); represents a baseline radionuclide release.	Dependent on manufacturing process and stochastically determined.
	Particle coating failure due to operation at high temperatures	Ability to predict extent and severity of particle coating failure at extreme temperatures, which has direct effect on iodine release, and other FPs.	Experimental data on TRISO particles
	Steam oxidizes UO_2 in particles with defective coating, allowing easier release of volatiles, during steam ingress accident	Ability to predict phase change of UO_2 as it is oxidized in presence of steam which enters due to defective coating. This change in lattice allows more volatile FPs to escape.	Oxidation potential of UO_2
	Steam hydrolyzes UC_x in particles with defective coatings, accompanied by release of some fission products, during steam ingress accident	Assuming a phase of uranium carbide exists in a TRISO particle of UO_2 fuel (or in a TRISO particle where UC_x is expected), ability to predict the reaction of UC_x with steam that enters through a defective coating. (Unexpected UC_x may be present due to failed manufacturing of coatings)	Oxidation potential of UC_x
	Reaction with air causing particle failure or disintegration, during air ingress accident	Ability to predict failure extent and rate of failure for TRISO particle coating layer reaction with air at extreme temperatures.	Irradiation experiments of TRISO fuel in air coupled with fission product release in fuel kernel at very high temperatures.

	Effect of reactivity transients and high burnup on failure of TRISO particle	Ability to predict the extent and rate of failure for TRISO particles during reactivity transients and/or after high burnup, and specifically the impact on FP release.	Experimental data on TRISO particles
	Delayed neutrons and delayed neutron precursors	Ability to track delayed neutrons and delayed neutron precursors.	Coupled neutronics and thermal hydraulic codes, DNP fractions.
	Tritium production due to ternary fission	Ability to predict the amount of tritium produced from ternary fission in the fuel kernel.	Neutronics and depletion analysis; burn-up, fissile nuclides, energy released per fission, thermal and fast neutron fluxes.
	Tritium release from TRISO particle	Ability to predict the release rate of tritium in TRISO fuel.	Fraction of defective particle coatings; diffusion characteristics of tritium in materials in the TRISO particle; temperature; experimental data.
Production of Radionuclides Outside of the Fuel	Production of radionuclides other than tritium via neutron activation of fuel materials, core materials, coolant salt or corrosion products	Ability to outline all the materials in the flux of neutrons in the reactor system, including fuel particle coatings, core materials, coolant salt, and structures. Ability to predict the neutron activation of those materials.	Outline all materials in the neutron flux of the core, including fuel, core, coolant and structural, and specifically which isotopes; neutron activation analysis of these materials.
	Neutron activation of impurities in salt coolant, either from initial conditions or built up during normal operation	Ability to predict the impurities present in the salt coolant due to the initial salt source, salt loading, normal operation, and diffusion/permeation from external sources or the fuel. Ability to predict the neutron activation of those impurities.	Dependent on source of coolant salt, and specific experimental data on the salt after loading and after various experiments; identifying other sources of impurities; neutron activation analysis of impurities.
	Tritium production due to activation of ${}^6\text{Li}$, ${}^7\text{Li}$, ${}^9\text{Be}$, ${}^{10}\text{B}$, ${}^{19}\text{F}$ outside of the fuel matrix	Ability to predict the amount of tritium produced from activation of Li, Be, and B nuclei found in the reactor core, typically Li/Be in the coolant, Li/Be/B in the graphite, absorber balls, control rods and carbon bricks.	Cross section data; neutronics; concentration and location of activated nuclei; design dependent.
	Tritium speciation in coolant salt	Ability to predict the chemical form of newly produced tritium in the coolant salt or nearby core materials: TF, HT(g), H ₂ (g) or T ⁺ (ion).	Chemical potential and thermochemistry of activation reaction producing tritium in salt; salt composition; temperature; hydrogen and helium cover gas or sparge gas partial pressures.

	Chemical reaction of radionuclides with silicon fluorides formed from corrosion of fuel particle	Ability to predict chemical speciation of metal radionuclides with silicon hexafluoride or volatile silicon tetrafluoride formed as a result of fluoride salt interactions with the SiC TRISO fuel particle. Ability to predict amount of silicon fluorides formed.	Thermochemistry of possible reactions between radionuclides and silicon fluorides, including metals and SiF_6 or radiohalides and SiF_4 .
	Formation of metallic carbides from corrosion of fuel particle	Ability to predict the chemical reaction or exchange of metal radionuclides with carbon in the SiC layer of TRISO fuel particles. Ability to predict speciation and phase, which is necessary to understand its transport through the salt.	Thermochemistry of possible reactions between metal radionuclides and SiC, forming various metal carbides.
Interaction of Radionuclides with Core Materials	Redox state and chemical form of fission product in core environment (graphite fuel element)	Ability to predict redox state and chemical form of the fission product in the core environment is very important. (Assumed to be very reducing)	Redox chemistry of fission products in graphite at given temperatures
	Chemisorption of fission products on graphite	Ability to predict chemisorption of Cs, Sr, iodine, etc on graphite; the effect of cosorption, or competing sorption from other fission products, on sorption rates is also important.	Equilibrium vapor pressures of fission products sorbed on graphite; formation enthalpies; diffusion coefficients in graphite; gas flow velocity or residence time over graphite surface; accident temperature; fast neutron fluence, but must account for annealing in graphite that occurs at the high accident temperatures.
	Chemical reaction of fission products plated on graphite with H_2 or CO which are formed when steam attacks graphite	Ability to predict chemical reactions of FPs sorbed on or in graphite with H_2 and/or CO , which are preferentially formed when steam attacks graphite. The diffusion of fission products further into graphite might occur when steam attacks graphite (gasification of graphite).	Thermochemistry of reactions, sorption status of FPs, partial pressures of gas reactants, temperature
	Gas volume expansion due to steam attacking graphite	Ability to predict level of gas volume expansion due to steam reacting with graphite and producing gaseous products CO and H_2 .	Thermodynamics; equation of state for gas volume.
	Chemical reaction of fission products with other volatile fission products	Ability to predict chemical reactions that might occur (e.g. CsI formation).	Thermodynamics of reaction; equilibrium vapor pressures over graphite; temperature.

	Stability of fission products in volatile state in high-temperature cover gas	Ability to predict the fission product's stability in the cover gas phase.	Vapor pressure equilibrium of FP in various chemical forms in cover gas in temperature range in presence of other FPs over graphite or metals.
	Effect of irradiation of graphite on sorption capacity	Ability to predict the changing sorption capacity on irradiated graphite over time for each fission product; note more pronounced effect of graphite irradiation than HTGRs.	Irradiation experiments coupled with sorption experiments for graphite
	Dust formation and interaction with radionuclides	Ability to predict how dust is formed, how it mobilizes and transports in the core and primary circuit, and how it interacts with fission products or other radionuclides such as tritium.	Identification of the chemical form and source of dust from the fuel matrix and graphite; sorption studies of dust with radionuclides; transport studies of dust in fluoride salt flow; largely experimental data.
	Interaction of tritium with graphite in reactor core	Ability to predict the sorption of tritium by graphite in the core, as well as diffusion through graphite.	Experimental data, temperature dependent.
	Permeation of tritium through metals in core	Ability to predict the transport of tritium (typically HT/T ₂) through metallic structures and components.	Experimental data on tritium gas transport in metals, or diffusion modeling.
	Interaction of tritium with B ₄ C in the reactor core	Ability to predict the sorption of tritium by B ₄ C after it is produced in the control rods.	Experimental data, temperature dependent.
Interactions of Radionuclides in the Primary Coolant System (PCS)	Adsorption capacity of PCS materials for fission products	Ability to predict the chemisorption and physisorption characteristics of Cs, Sr, iodine in the submonolayer regime on metals with(out) an oxide layer.	Material characteristics such as composition and existence of an oxide layer; surface roughness; porosity; type of fission product and chemical form; temperature.
	Plate-out and equilibrium partition of fission and activation products on HX or other PCS surface	Ability to predict the plate-out of fission, corrosion or activation products on heat exchanger surface or other PCS surfaces. Also need to calculate equilibrium partition between the two states.	Gibbs free energy of formation between elemental and salt forms of fission products and alloy metals, temperature.
	Desorption of fission products from surfaces due to flow of air or steam, during air and/or water ingress accident	Ability to model air flow transport during an air ingress accident. Also includes mobilization due to simple mass transfer and kinetics, without chemical reaction. Must also factor in	CFD analysis of PCS dependent on geometry and leak type and air ingress is important; model shear forces along surfaces mobilizing the FPs; predict chemical reactions of FPs with air.

		transport due to evaporation of water.	
	Mobilization of fission products plated on metals by reaction with steam from water ingress or reaction with air from air ingress	Mobilization of fission products from chemisorbed state due to reaction with water and formation of hydroxide or other hydrogen containing compound. Also including chemical reactions with air as well as shear forces along surfaces.	Chemical status and sorption status of FPs, thermodynamics of reaction, equilibrium vapor pressures over graphite, temperature (some experimental data exists for Cs).
	Chemical reaction of fission products plated on metals by H ₂ or CO which are formed when steam attacks graphite	Ability to predict chemical reactions of FPs sorbed onto or into metals with hydrogen gas and/or CO, which are preferentially formed when steam attacks graphite.	Thermochemistry of reactions; sorption status of FPs; partial pressures of gas reactants; temperature.
	Volatilization of chemical species	Ability to predict volatilization of chemical species in salt mixture and bubble transport through salt to the off-gas system. Important to note some fission products have noble gas precursors such as ¹³⁷ Cs and ⁹⁰ Sr; these FP may end up in the off-gas after decay from a volatile precursor. Potentially beneficial to separate volatile species in different classes of volatility.	Thermodynamics; phase diagrams; modeling bubble formation with transport equations; fission product yields of all volatile species regardless of half-life; decay of those daughters with interaction in off-gas system.
	Mechanical agitation of salt caused by normal pump operation	Ability to predict the effect that the pump has on the mechanical agitation of the salt and its loosely bound components. This agitation may have a sort of "ultrasonic effect" on the salt, easing transport of volatile or insoluble components.	Sorption thermodynamics of volatile or insoluble species in salt; specific experiments focused on agitation of molten salt mixtures; momentum and mass transport phenomena.
	Salt-insoluble chemical species transported via sweep gas	Fission products or other activation products formed that are insoluble in the salt mixture may be transported via sweep gas and plated out in HX or elsewhere.	Thermodynamics of formation and chemical speciation, particulate formation, transport via gaseous flow or volatilization, plating on pipe or vessel walls dependent on temperature and flow characteristics.
	Aerosolized salt transported via sweep gas	Ability to predict the formation of fuel salt mist in the sweep gas and transported to the off-gas system or elsewhere (if applicable). Modeling of salt	Phase and stability diagrams of molten salts; aerosol formation and kinetics of molten salts; gas phase composition, temperature and pressure.

		phase change via aerosolization.	
	Chemical species stable in salt mixture	Ability to predict stability diagrams of salt mixtures with varying compositions at varying temperatures.	Thermodynamics, solubility, kinetics of chemistry, temperature, pressure.
	Dust mobilization behavior in PCS	Ability to predict how dust mobilizes and transports in the PCS, how plateout occurs, and if it interacts further with radionuclides. Ability to predict its mobilization in cover gas or due to air ingress. Ability to predict dust particle sizes and interactions.	Mass transport phenomena in salt flow; chemical interaction of graphite dust with salt of changing composition; CFD analysis of cover gas phase containing dust during normal operation or during air ingress; model shear forces along surfaces mobilizing the dust.
	Interaction of tritium with coolant salt	Ability to predict the sorption of tritium by the coolant salt, diffusion and transport through the salt, and potential escape into the cover gas. TF and T ⁺ may stay dissolved in the salt or attack metallic surfaces, while most HT/T ₂ will diffuse through the salt and escape into the cover gas unless oxidized.	Dependent on chemical speciation of tritium: thermochemistry, salt composition, temperature, hydrogen and helium cover gas partial pressure.
	Permeation of tritium through metals in HX into the secondary loop	Tritium can permeate metal surfaces of the heat exchanger as well as stainless steel tubes of the steam generator at high temperature, therefore it is important to be able to predict this permeation rate.	Experimental data or diffusion modeling.
	Removal of tritium from salt coolant	Ability to predict amount of tritium removed from salt coolant using a purification system of the salt, sacrificial carbon anode, or a sparge gas.	Design dependent
Chemical Interactions of Radionuclides in the Reactor Building	Oxidation of fission product metals by air	Ability to predict speciation of Cs, Sr, etc as hydroxide, oxide, carbonate, etc after reacting with air: water, oxygen, carbon dioxide.	Thermodynamics of chemical speciation; temperature; partial pressures of air.
	Volatile fission product behavior	Due to lower fission product releases in FHR, aerosol formation and condensation is not likely, therefore the ability to predict the behavior of volatile fission products in elemental or oxidized form is important. Focus on	Mobilization by transport processes of air; diffusion through containment; other pathways for escape.

		interaction with materials in reactor building.	
	Reaction of air with core components and salt during structure failure or air ingress accident	Ability to predict potential reactions of core components or molten salt with air inside the reactor.	Reaction thermodynamics, molten salt composition, state of graphite in core, salt temperature, air composition.
	Delayed transport of radionuclides from the reactor system to the reactor building after structure failure or air ingress accident	Delayed release of radionuclides after breach in system from core heat-up over span of 100+ h if coolant fails. The driving force is the slow heat-up of the core following the breach.	Design dependent as the gas behavior is mostly a function of natural convection and exchange flow phenomena.
	Volatilization of chemical species in environment outside MSR system during salt spill	Ability to predict the volatilization of species in the salt exposed by the leak. Ability to account for changes in pressure of containment: increase or decrease in pressure of the environment where the leak is located.	Vapor pressure determination of all chemical species in the salt mixture; dissociation kinetics and thermodynamics of various salt systems in air at ambient temperatures and pressures; temperature.
	Aerosolization of salt in reactor building during salt spill	Ability to model the splash radius of the salt and aerosol creation as it leaks into the environment surrounding the pipe/MSR component, as well as aerosolization due to falling liquid.	Conditions associated with salt spill, such as fall height, temperature, etc. Will be design/accident dependent.
	Passive heat removal during salt spill	Ability to predict the rate at which heat is removed and the salt freezes in the catch pan. Ability to predict how much transport occurs until that point, whether to drain tank (if connected to one), through catch pan material to the concrete below, or otherwise.	Heat contained in salt can be estimated by salt temperature at breach; rate of heat removal dependent on temperature of salt and methods of heat transport to surroundings: radiative to environment, conductive to catch pan surface and concrete below, convective to natural or forced circulation of air.
	Degradation and failure of catch pan due to molten salt during salt spill	Ability to predict failure rate of catch pan due to conductive and convective heat transfer from molten salt.	Material properties of catch pan; conductive and convective heat transport phenomena modeling; temperature of molten salt exiting leak.
	Deformation of concrete below catch pan due to molten salt during salt spill	Ability to model heat transfer from molten salt to concrete and corresponding deformation of concrete.	Material properties of concrete; conductive and convective heat transport phenomena modeling; temperature of molten salt exiting leak from deformed catch pan.

	Chemical reaction of molten salt with structural components, air, or water during salt spill and severe accident	Ability to predict potential reactions of molten salt components with structural components (possible steel or concrete), air, and/or water during salt spill and air or water ingress.	Reaction thermodynamics, molten salt composition, salt temperature, alloy or concrete composition, air composition.
Transport from Leak Location to Environment	Transport of radionuclide aerosols and gases/vapors from leak compartment to environment, or retention within compartment	Ability to predict transport from the leak location to the environment. Possible phenomena include aerosol deposition, vapor/gas condensation, chemical reactions of airborne radionuclides, and plugging of leaks by radionuclide compounds.	Dependent on reactor design, number of containment layers for reactor systems, and specific accident sequence, but likely utilizes temperature/pressure conditions in the compartment and a characterization of the radionuclide.
	Off-site dispersion of radionuclides	Ability to predict off-site dispersion and transport of the radionuclides released from the reactor building into the environment. Transport rates through soil and other surrounding materials.	Similar to that for other nuclear reactor designs see HTGR table) with additional focus on tritium and fluoride salts.

4 Sodium-Cooled Fast Reactors

Significant progress has been made on the development of an MST for sodium-cooled fast reactor (SFRs) due to recent DOE- and industry-sponsored efforts [90-95]. These efforts provide an overview of source term development for a generic metal fuel, pool-type SFR, an assessment of radionuclide release fractions from metal fuel, a trial MST calculation, and initial code development efforts. The major outcomes of the trial calculation were the identification of knowledge gaps in models and data, as well as the ranking of phenomena that contribute most to uncertainty in the assessment by sensitivity calculations. These outcomes can be used to prioritize future research regarding the development of a mechanistic source term for the SFR. The current report is meant only as an overview of the phenomena relevant to modeling source term in an SFR, as well as the functional requirements needed, with additional information available in the reports referenced above. A summary diagram of the SFR radionuclide transport pathways was published in those reports and is provided below in Figure 4-1.

Table 4-1 below outlines the phenomena important for MST modeling in an SFR, with the main focus being a radionuclide release from core fuel and primary sodium. The first two analysis phases include the production and release of radionuclides from the metal fuel pin, and the production of radionuclides outside of the fuel (e.g. ^{22}Na production via neutron activation of ^{23}Na). While fissions inside the fuel are the main production of radionuclides, care must be taken to estimate radionuclides produced via neutron activation of the core materials. Additional sources of radionuclides include ^{41}Ar which can be created from either potassium impurities in the sodium pool or argon which may be used as a cover gas [90]. Radionuclide behavior in metal fuel, which is typical of U.S. SFR designs, is complex, with high retention of certain radionuclides and high mobility of others. Therefore, it is of paramount importance to understand the phenomena associated with radionuclide interactions in the fuel pin, fuel cladding, and the bond sodium between the two, as well as the fission gas plenum above. These interactions not only include the chemical or physical interactions of the isotope in the material, but also the interactions between these different materials at varying temperatures, likely changing compositions, and increasing gas pressures in the fuel subassembly (if not vented). All of these factors can impact radionuclide release.

The third analysis phase considered here is the interactions of radionuclides in the core and primary coolant system (PCS). The core interactions in this analysis phase are taken to mean those that occur outside the fuel cladding, or as the radionuclide has already escaped the fuel pin. After release from the fuel, the radionuclides will interact with the significant sodium inventory within the reactor vessel. Also design dependent are auxiliary systems that may be used in the reactor, such as a Cover Gas Cleanup System (CGCS) or a Primary Sodium Purification System (PSPS). These could be important to the source term as they remove (sometimes in large quantities) radionuclides as they travel through the core, coolant circuit or cover gas. However, it is possible that such systems may not be utilized during transient events. These potentially complex separations systems are left in the table as a single phenomenon for brevity, therefore the reader is once again referred to the RTDP reports for more information [90].

The fourth and fifth analysis phases considered here have to do with the interactions of radionuclides in the reactor building or containment of the reactor system, and the transport of the radionuclides from the leak location to the environment. While these can be very similar to those

phenomena for both light water reactors (LWR) and other advanced non-LWR reactors, an attempt to summarize and highlight key differences has been made. An important phenomenon is the leakage of aerosols from the cover gas in the reactor system to the reactor building, as well as the leakage from the reactor building. Especially unique is the possibility of the leakage path becoming plugged with oxidized sodium caused by sodium reacting with surrounding materials and air to precipitate as a solid [90]. This obviously has a sort of self-healing effect on source term analysis, but must be investigated further. This can be a difficult phenomenon to model properly and is mainly dependent on the specific leak type, location, and size, which may not be known.

Figure 4-1: Summary Diagram for SFR Radionuclide Retention Phenomena [90]

Table 4-1: Functional Requirements for Modeling Source Term Release in SFRs

Analysis Phase	Phenomena	Functional Requirements	Necessary Data or Models
Production and Behavior of Radionuclides in the Fuel Pin	Chemical forms of fission products in metal fuel pin	Ability to predict fission products created, and chemical form of the fission product in the metal fuel.	Depletion analysis; fission products in metal fuel; temperature.
	Decay of radionuclides into radionuclide daughters	Ability to account for the decay daughters of all radionuclides, especially noting the half-life and if a change in chemical class occurs, affecting the speciation and transport.	Specific identification of all relevant radioisotopes of interest with decay chains; thermochemistry data.
	Diffusion of fission products in metal fuel	Ability to predict transport of fission products due to diffusion through the metal fuel pin, especially volatile fission products.	Diffusion coefficients of the chemical form of each fission product in the metal fuel pin, as well as the geometry of the fuel pin.
	Diffusion of fission products in bond sodium and cladding	Ability to predict transport of fission products due to diffusion through the bond sodium and cladding exterior to the fuel pin.	Diffusion coefficients of the chemical form of each fission product in both the bond sodium and cladding, as well as the geometry of the fuel cladding.
	Migration to fission gas plenum, pressure build-up in fission gas plenum in fuel cladding, if not vented	Ability to predict the amount of gaseous fission products that escape the metal fuel but are kept within the fuel cladding. Ability to calculate the partial pressures of each fission product and other gas components. If vented fuel pin design is used, ability to predict transport of vented gas through reactor core to cover gas.	Component in gas phase in plenum; volume of gas space; temperature.
	Fuel pin failure due to manufacturing, and associated fission product release	Ability to predict fraction of fuel pins with defective or failed cladding, or lattice imperfections in metal alloy structure; represents a baseline fission product release.	Dependent on manufacturing process and stochastically determined.
	Cladding failure due to high burnup and operation at extreme temperatures	Ability to predict extent and severity of cladding failure at extreme temperatures, likely due to creep rupture from fuel swelling and gas pressurization.	Experimental data on specific fuel and cladding design; materials testing; phase diagrams and eutectic point of fuel and cladding.

	Chemical reactions between fission products and fuel pin	Ability to predict the possible chemical reactions occurring between fission products and the fuel components, such as uranium forming $U\text{I}_3$, or eutectic formation.	Thermochemistry of chemical reactions; concentrations of fission products and activity coefficients; temperature.
	Chemical reactions between fission products and bond sodium	Ability to predict the possible chemical reactions occurring between fission products escaping the fuel and the bond sodium, such as CsI forming NaI and Cs ion.	Thermochemistry of chemical reactions; concentrations of fission products and activity coefficients; temperature.
	Chemical interactions between fuel and cladding	Ability to predict the possible metal mixtures created from interdiffusion of elements between the fuel alloy and the cladding at elevated temperatures; likely formation of eutectic mixture.	Metal composition in both fuel and cladding and diffusion characteristics of each at elevated temperatures; experimental data; phase diagrams of possible mixtures.
	Fuel melting	Ability to predict the melting temperature of the fuel pin and the rate and likelihood of volatilization of individual components of the fuel pin after burnup. Important at high temperatures for the potential release of iodine in $U\text{I}_3$.	Phase diagrams of possible fuel pin mixtures after burnup; vapor pressure equilibrium of fission product compounds over melted fuel pin composition; temperature.
	Delayed neutrons and delayed neutron precursors	Ability to track delayed neutrons and delayed neutron precursors.	Coupled neutronics and thermal hydraulic codes, DNP fractions.
Production of Radionuclides Outside of the Fuel	Neutron activation of primary sodium coolant	Ability to predict the amount of radionuclides produced via neutron activation: $^{23}\text{Na}(n,2n)^{22}\text{Na}$ and $^{23}\text{Na}(n,p)^{23}\text{Ne}$ and $^{23}\text{Na}(n,g)^{24}\text{Na}$.	Nuclear data (neutron capture cross sections) for relevant isotopes of Na and Ne; neutron flux and energy spectrum.
	Neutron activation of corrosion products of core components	Ability to predict the amount of radionuclides produced via neutron activation: ^{51}Cr , ^{54}Mn , ^{59}Fe , ^{58}Co , ^{60}Co , ^{182}Ta .	Outline all materials in the neutron flux of the core, including core and structural, and specifically which isotopes; neutron activation analysis of these materials.
	Neutron activation of cover gas	Ability to predict the amount of radionuclides produced via neutron activation of the cover gas, especially if argon is used: $^{40}\text{Ar}(n,g)^{41}\text{Ar}$ (in cover gas or in sodium coolant pool)	Nuclear data (neutron capture cross sections) for relevant isotopes of Ar, K, or other cover gas; neutron flux and energy spectrum.

		and $^{41}\text{K}(\text{n,p})^{41}\text{Ar}$ (impurity in sodium coolant pool).	
Interactions of Radionuclides in the Core and Primary Coolant System (PCS)	Chemical form of radionuclides in core environment outside of fuel subassembly	Ability to predict chemical form of the fission product in the core environment after it has escaped the fuel matrix and cladding.	Design dependent; fission products at given temperatures.
	Transport of fission products from fuel subassembly through core and into sodium coolant	Ability to predict the rate at which volatile fission products transport through the fuel subassembly and core and into the sodium coolant.	Dependent of fuel matrix design; diffusion data; experimental data.
	Interaction of primary sodium with metal fuel and breached cladding	Ability to predict the chemical reactions or phase changes that may occur if there is a breach in cladding and primary sodium interacts with the metal fuel and fission gas plenum.	Thermochemistry of reactions between liquid sodium and metal fuel, fission gases and cladding; phase diagrams of possible metal mixtures; temperature.
	Chemical reaction of fission products with other volatile fission products	Ability to predict chemical reactions that might occur (e.g. CsI formation).	Thermodynamics of reaction; equilibrium vapor pressures over graphite; temperature.
	Chemical speciation of fission products in primary sodium coolant	Ability to predict the speciation of Cs, Sr, iodine and other released fission products from failed fuel pins in the primary sodium coolant pool and PCS.	Thermochemistry of fission products in liquid sodium pool with other fission, corrosion and activation products; temperature.
	Interaction of radionuclide vapors and bubbles with sodium coolant	Ability to predict the fate of fission products transporting through the liquid sodium pool, typically via bubble transport. Volatile noble gases may not dissolve into sodium, some vapors may condense to a liquid phase, and other vapors may directly dissolve into the sodium due to high solubility.	Solubility of the radionuclide in liquid sodium; vapor pressure of radionuclide over liquid sodium; bubble transport formation and mechanics in liquid sodium; thermochemistry of radionuclide with liquid sodium.
	Chemical species insoluble in liquid sodium transported via fission product vapors	Fission products or other particulates formed that are insoluble in the liquid sodium may be transported via bubble transport by volatile or gaseous fission products, and plated out in HX or elsewhere. These may be released into the sodium pool if the bubble collapses.	Thermodynamics of formation and chemical speciation, particulate formation, transport via gaseous flow or volatilization, plating on pipe or vessel walls dependent on temperature and flow characteristics.

	Chemical species soluble in liquid sodium	Ability to predict stability diagrams of mixtures of the radionuclide chemical species with liquid sodium at varying compositions and varying temperatures.	Thermodynamics, solubility, kinetics of chemistry, temperature, pressure.
	Transport of noble gas fission products through coolant to cover gas system	Ability to predict the rate of transport of noble gas radionuclides through liquid sodium to the cover gas. Volatile radionuclides include gaseous fission products from the fuel as well as noble gases produced from sodium coolant activation.	Henry's Law; kinetics of gas-liquid mass transport/diffusion.
	Stability of fission products in volatile state in cover gas	Ability to predict the fission product's stability in the cover gas phase.	Vapor pressure equilibrium of FP in various chemical forms in cover gas in temperature range in presence of other FPs over liquid sodium.
	Evaporation of gaseous species or bubbles from sodium coolant pool	Ability to predict the evaporation rate of the gaseous species dissolved in the sodium pool, largely dependent on the vapor pressures of the species, temperature and mixing.	Thermochemistry and chemical potential; chemical state of radionuclide in sodium; volatility and vapor pressure over liquid sodium; chemical state of radionuclide upon release to cover-gas.
	Aerosol behavior in cover gas region	Ability to predict the formation and behavior of aerosolized vapors of sodium or other fission products from the sodium pool.	Aerosol formation experimental data; vapor pressure equilibrium data.
	Condensation of radionuclide vapors on PCS surfaces	Ability to predict the condensation rate on PCS surfaces of vapors carrying radionuclides in the cover-gas region.	Vapor pressure equilibrium of radionuclide vapor in cover gas in temperature range over liquid sodium.
	Plate-out of radionuclide particulates on PCS surfaces	Ability to predict the plate-out or mechanical deposition of fission, corrosion or activation products on heat exchanger surface or other PCS surfaces. Ability to calculate equilibrium partition between the two states.	Gibbs free energy of adsorption between elemental or other relevant forms of fission products and the PCS surfaces; temperature.

	Desorption of radionuclide from PCS surface due to mechanical agitation of vapor flow	Ability to model the mobilization of particulates sorbed on metal surfaces in the cover-gas region due to simple mass transfer and kinetics, without chemical reaction. Includes the resuspension or revaporization of the radionuclide in the sodium pool or cover gas.	CFD analysis of PCS dependent on geometry and vapor flow parameters; model shear forces along surfaces mobilizing the FPs.
	Resuspension of fission product metals decayed from volatile fission products in cover-gas	Ability to predict the resuspension of chemical species in the sodium pool from the cover-gas region. Some fission products have noble gas precursors such as ^{137}Cs and ^{90}Sr , so while the FP may be in the cover-gas initially, it may end up in the sodium pool after decaying to a nonvolatile daughter.	Thermodynamics, phase diagrams, modeling bubble formation with transport equations, fission product yields of all volatile species regardless of half-life, and decay of those daughters with interaction in off-gas system. Potentially beneficial to separate volatile species in different classes of volatility.
	Mechanical agitation of sodium caused by normal pump operation	Ability to predict the effect that the pump has on the mechanical agitation of the sodium and its loosely bound components. This agitation may have a sort of "ultrasonic effect" on the sodium, easing transport of volatile or insoluble components.	Sorption thermodynamics of volatile or insoluble species in sodium; specific experiments focused on agitation of liquid sodium mixtures; momentum and mass transport phenomena.
	Leakage from the reactor vessel and plugging of leakage pathways by aerosols	Ability to predict the leakage rate caused by a specific leak in the reactor vessel, and the potential plugging of leakage pathways by oxidation of aerosols on both sides of the leak, hindering further leaking.	Leak specific information such as size and location; aerosol characteristics such as components and temperature of condensation; oxidation potential of aerosolized sodium.
	Removal of radionuclides from cover gas by Cover Gas Cleanup System (CGCS)	Ability to predict amount of volatile fission products and other gaseous radionuclides removed from the cover gas using a cleanup system utilizing cryogenic distillation.	Design dependent; common designs include cryogenic distillation and a cold trap that removes Xe and Kr from Ar.
	Removal of radionuclides from sodium coolant by Primary Sodium Purification System (PSPS)	Ability to predict amount of tritium, cesium and other radionuclides removed from sodium coolant using a purification system utilizing a cold trap.	Design dependent; common designs include a cold trap that exploits the saturation temperature of liquid sodium for these radionuclides.

Chemical Interactions of Radionuclides in the Reactor Building	Chemical reaction of fission products and sodium vapor with air	Ability to predict speciation of Na, Cs, Sr, etc as hydroxide, oxide, carbonate, etc after reacting with air: water, oxygen, carbon dioxide. Also includes conversion of iodine as metal iodide to elemental iodine after the metal forms an oxide: NaI.	Thermodynamics of chemical speciation, temperature, partial pressures of air.
	Volatile fission product behavior	Ability to predict the behavior of volatile fission products is important. Focus on interaction with materials in reactor building such as oxygen, water, or surfaces.	Transport processes and diffusion through air; dependent on characteristics of the reactor building and air composition, temperature and pressure.
	Aerosolization of sodium vapor and fission products	Ability to predict aerosol formation and behavior of sodium vapor outside of the primary circuit. Condensation of aerosol particles in the reactor building and the leakage rate from the reactor are both very important.	Behavior of vaporized sodium and fission products in air at ambient conditions.
	Plate-out of radionuclide particulates on reactor building surface	Ability to predict the plate-out or mechanical deposition of leaked radionuclides on the much cooler surfaces of the reactor building. Also need to calculate equilibrium partition between the two states.	Gibbs free energy of adsorption between elemental or other relevant forms of fission products and the building surfaces; temperature.
	Delayed transport of radionuclides from the reactor system to the reactor building after a leak	Delayed release of radionuclides after breach in system from core heat-up over span of 100+ h if coolant fails. The driving force is the slow heat-up of the core following the breach.	Design dependent as the gas behavior is mostly a function of natural convection and exchange flow phenomena.
	Leakage from the reactor building and plugging of leakage pathways by aerosols	Ability to predict the leakage rate caused by a specific leak in the reactor building, and the potential plugging of leakage pathways by oxidation of aerosols on both sides of the leak, hindering further leaking.	Leak specific information such as size and location; aerosol characteristics such as components and temperature of condensation; oxidation potential of aerosolized sodium.

Transport from Leak Location to Environment	Transport of radionuclide aerosols and gases/vapors from leak compartment to environment, or retention within compartment	Ability to predict transport from the leak location to the environment. Possible phenomena include aerosol deposition, vapor/gas condensation, chemical reactions of airborne radionuclides, and plugging of leaks by radionuclide compounds.	Dependent on reactor design, number of containment layers for reactor systems, and specific accident sequence, but likely utilizes temperature/pressure conditions in the compartment and a characterization of the radionuclide.
	Off-site dispersion of radionuclides	Ability to predict off-site dispersion and transport of the radionuclides released from the reactor building into the environment. Transport rates through soil and other surrounding materials.	Similar to that for other nuclear reactor designs see HTGR table) with additional focus on alkali metals.

4.1 Lead-Cooled Fast Reactor Considerations

For pool-type lead-cooled fast reactors (LFRs), the source term phenomena and functional requirements are sufficiently similar to those outlined in Table 4-1 for SFRs that a separate analysis and table are not necessary. However, there are several key differences and unique phenomena that should be considered for LFRs, which are reviewed here.

While U.S. SFR designs typically utilize sodium-bonded metal fuel, LFR concepts have been proposed that utilize oxide, nitride, and bondless metal fuels. The chosen fuel form impacts the phenomena related to radionuclide behavior within the fuel pin and radionuclide release during cladding failure. In addition to radionuclides present within the fuel, the production of Po-210 from Bi-209 within the lead coolant is another potential source of radioactive material. While polonium production in LFRs is not as high as in a reactor utilizing a lead-bismuth eutectic, it is still present from Bi-209 impurities and Bi-209 generation from the isotope Pb-208 [96]. Lastly, unlike sodium, energetic lead reactions with oxygen are not a concern at the normal operating temperatures of LFRs.

Bibliography

- [1] J. S. Merrifield, "U.S. Nuclear Industry Council Discussion of Policy Issues and Results of USNIC 2020 Advanced Nuclear Survey," ed: USNIC, 2020.
- [2] J. Taylor, "Issues Pertaining to the Advanced Reactor (PRISM, MHTGR, and PIUS) and CANDU 3 Designs and their Relationship to Current Regulatory Requirements," U.S. Nuclear Regulatory Commission, SECY-93-092, April 8 1993.
- [3] W. D. Travers, "Policy Issues Related to Licensing Non-Water Reactor Designs," U.S. Nuclear Regulatory Commission, SECY-03-0047, March 28 2003.
- [4] L. A. Reyes, "Second Status Paper on the Staff's Proposed Regulatory Structure for New Plant Licensing and Update on Policy Issues Related to New Plant Licensing," U.S. Nuclear Regulatory Commission, SECY-05-0006, January 7 2005.
- [5] "Memorandum: Staff Requirements - SECY-19-0117 - Technology-Inclusive, Risk-Informed, and Performance-Based Methodology to Inform the Licensing Basis and Content of Applications for Licenses, Certifications, and Approvals for Non-LWRs," U.S. Nuclear Regulatory Commission, SRM-19-0117, May 26 2020.
- [6] "Risk-Informed Performance-Based Technology Inclusive Guidance for non-LWR Licensing Basis Development, Revision 1," Nuclear Energy Institute, 18-04, August 2019.
- [7] "NRC Non-Light Water Reactor Vision and Strategy, Volume 3 - Computer Code Development Plans for Severe Accident Progression, Source Term, and Consequence Analysis," U.S. Nuclear Regulatory Commission, ML20030A178, January 2020.
- [8] "Probabilistic Risk Assessment Standard for Advanced Non-LWR Nuclear Power Plants," ASME/ANS, ASME/ANS RA-S-1.4-2013, December 9 2013.
- [9] J. Cao, L. Zhang, F. Xie, B. Xia, and S. T. T. Lam, "Source Term Study on Tritium in HTR-PM: Theoretical Calculations and Experimental Design," *Science and Technology of Nuclear Installations*, vol. 2017, pp. 1-11, 2017, doi: 10.1155/2017/3586723.
- [10] Y. Xu, H. Li, F. Xie, J. Cao, and J. Tong, "Source Term Analysis of Tritium in HTR-10," *Fusion Science and Technology*, vol. 71, no. 4, pp. 671-678, 2017, doi: 10.1080/15361055.2017.1290949.
- [11] X. Liu, X. Huang, F. Xie, F. Jia, X. Feng, and H. Li, "Source Term Analysis of the Irradiated Graphite in the Core of HTR-10," *Science and Technology of Nuclear Installations*, vol. 2017, pp. 1-6, 2017, doi: 10.1155/2017/2614890.
- [12] D. A. Petti, R. R. Hobbins, P. Lowry, and H. Gougar, "Representative Source Terms and the Influence of Reactor Attributes on Functional Containment in Modular High-Temperature Gas-Cooled Reactors," *Nuclear Technology*, vol. 184, no. 2, pp. 181-197, 2013, doi: 10.13182/nt184-181.

- [13] K. Verfondern, A. Xhonneux, H. Nabielek, and H.-J. Allelein, "Computational analysis of modern HTGR fuel performance and fission product release during the HFR-EU1 irradiation experiment," *Nuclear Engineering and Design*, vol. 273, pp. 85-97, 2014, doi: 10.1016/j.nucengdes.2014.01.026.
- [14] R. Moormann and K. Hilpert, "Chemical Behavior of Fission Products in Core Heatup Accidents in High-Temperature Gas-Cooled Reactors," *Nuclear Technology*, vol. 94, no. 1, pp. 56-67, 1991, doi: 10.13182/nt91-a16221.
- [15] R. Moormann, W. Schenk, and K. Verfondern, "Source Term Estimation for Small-Sized HTRs: Status and Further Needs, Extracted from German Safety Analyses," *Nuclear Technology*, vol. 135, no. 3, pp. 183-193, 2001, doi: 10.13182/nt01-a3215.
- [16] R. Moormann, "AVR prototype pebble bed reactor: a safety re-evaluation of its operation and consequences for future reactors," *Kerntechnik*, vol. 74, no. 1-2, pp. 8-21, 2009.
- [17] L. R. Zumwalt, "FREVA-8 Formulation for estimating the release of metallic fission products from HTGR fuel elements," Gulf General Atomic, Inc, GAMD-7535(Rev.), March 1967.
- [18] J. Appel and B. Roos, "A Study of the Release of Radioactive Metallic Isotopes from High-Temperature Gas-Cooled Reactors," *Nuclear Science and Engineering*, vol. 34, no. 3, pp. 201-213, 1968, doi: 10.13182/nse68-a21086.
- [19] R. Moormann, "Fission Product Transport and Source Terms in HTRs: Experience from AVR Pebble Bed Reactor," *Science and Technology of Nuclear Installations*, vol. 2008, pp. 1-14, 2008, doi: 10.1155/2008/597491.
- [20] G. H. Lohnert, "The consequences of water ingress into the primary circuit of an HTR-Module - From design basis accident to hypothetical postulates," *Nuclear Engineering and Design*, vol. 134, no. 2-3, pp. 159-176, 1992, doi: 10.1016/0029-5493(92)90137-k.
- [21] C. C. Stoker, L. D. Olivier, E. Stassen, F. Reitsma, and J. J. van der Merwe, "PBMR radionuclide source term analysis validation based on AVR operating experience," *Nuclear Engineering and Design*, vol. 240, no. 10, pp. 2466-2484, 2010, doi: 10.1016/j.nucengdes.2009.10.011.
- [22] H. Jeong and S. H. Chang, "Development of a method of evaluating an inventory of fission products for a pebble bed reactor," *Annals of Nuclear Energy*, vol. 35, no. 12, pp. 2161-2171, 2008, doi: 10.1016/j.anucene.2008.10.001.
- [23] C. H. Oh *et al.*, "Development of safety analysis codes and experimental validation for a very high temperature gas-cooled reactor," Idaho National Laboratory, INL/EXT-06-01362, March 2006.
- [24] W. Moe, J. Kinsey, K. Perry, D. Petti, and G. Gibbs, "Mechanistic Source Term White Paper," in "Next Generation Nuclear Plant Project," Idaho National Laboratory, INL/EXT-10-17997, July 2010.

- [25] A. C. Kadak, "The Status of the US High-Temperature Gas Reactors," *Engineering*, vol. 2, no. 1, pp. 119-123, 2016, doi: 10.1016/j.Eng.2016.01.026.
- [26] D. Grabaskas, T. Fei, and J. Jerden, "Technical Letter Report on The Assessment of Tritium Detection and Control in Molten Salt Reactors: Final Report," Argonne National Laboratory, ANL/NSE-20-15, May 2020.
- [27] R. Yoshioka, K. Mitachi, Y. Shimazu, and M. Kinoshita, "Safety criteria and guidelines for MSR accident analysis," presented at the PHYSOR 2014 – The Role of Reactor Physics Toward a Sustainable Future The Westin Miyako, Kyoto, Japan, September 28 – October 3, 2014.
- [28] B. Chisholm, S. Krahn, A. Afzali, and E. Harvey, "Application of a Method to Estimate Risk in Advanced Nuclear Reactors- A Case Study on the MSRE," presented at the Probabilistic Safety Assessment and Management PSAM Los Angeles, CA, September 14, 2018.
- [29] D. Gérardin *et al.*, "A methodology for the identification of the postulated initiating events of the Molten Salt Fast Reactor," *Nuclear Engineering and Technology*, vol. 51, no. 4, pp. 1024-1031, 2019, doi: 10.1016/j.net.2019.01.009.
- [30] M. H. Mohsin, K. Qureshi, and T. Ashfaq, "Safety assessment of MSR concept using INPRO methodology," *Progress in Nuclear Energy*, vol. 117, 2019, doi: 10.1016/j.pnucene.2019.103099.
- [31] Z. Guo, D. Zhang, Y. Xiao, W. Tian, G. Su, and S. Qiu, "Simulations of unprotected loss of heat sink and combination of events accidents for a molten salt reactor," *Annals of Nuclear Energy*, vol. 53, pp. 309-319, 2013, doi: 10.1016/j.anucene.2012.09.009.
- [32] B. R. Betzler, J. J. Powers, and A. Worrall, "Molten salt reactor neutronics and fuel cycle modeling and simulation with SCALE," *Annals of Nuclear Energy*, vol. 101, pp. 489-503, 2017, doi: 10.1016/j.anucene.2016.11.040.
- [33] K. Zhuang, Y. Zheng, L. Cao, T. Hu, and H. Wu, "Improvements and validation of the transient analysis code MOREL for molten salt reactors," *Journal of Nuclear Science and Technology*, vol. 54, no. 8, pp. 878-890, 2017, doi: 10.1080/00223131.2017.1323689.
- [34] L. Cao, K. Zhuang, Y. Zheng, T. Hu, and H. Wu, "Transient analysis for liquid-fuel molten salt reactor based on MOREL2.0 code," *International Journal of Energy Research*, vol. 42, no. 1, pp. 261-275, 2018, doi: 10.1002/er.3828.
- [35] A. Lindsay, G. Ridley, A. Rykhlevskii, and K. Huff, "Introduction to Moltres: An application for simulation of Molten Salt Reactors," *Annals of Nuclear Energy*, vol. 114, pp. 530-540, 2018, doi: 10.1016/j.anucene.2017.12.025.
- [36] D. Wooten and J. J. Powers, "A Review of Molten Salt Reactor Kinetics Models," *Nuclear Science and Engineering*, vol. 191, no. 3, pp. 203-230, 2018, doi: 10.1080/00295639.2018.1480182.

- [37] B. R. Betzler, K. B. Bekar, W. A. Wieselquist, S. W. Hart, and S. G. Stimpson, "Molten Salt Reactor Fuel Depletion Tools in SCALE," presented at the Global 2019, Seattle, WA, September 22-27, 2019.
- [38] A. Graham, B. Collins, R. Salko, Z. Taylor, and C. Gentry, "Development of Molten Salt Reactor Modeling and Simulation Capabilities in VERA," pp. 1-9, 2019.
- [39] C. Wan, T. Hu, and L. Cao, "Multi-physics numerical analysis of the fuel-addition transients in the liquid-fuel molten salt reactor," *Annals of Nuclear Energy*, vol. 144, 2020, doi: 10.1016/j.anucene.2020.107514.
- [40] M. S. Greenwood and B. Betzler, "Modified Point-Kinetics Model for Neutron Precursors and Fission Product Behavior for Fluid-Fueled Molten Salt Reactors," *Nuclear Science and Engineering*, vol. 193, no. 4, pp. 417-430, 2018, doi: 10.1080/00295639.2018.1531619.
- [41] Z. Taylor, "Implementation of Multi-phase Species Transport into VERA-CS for Molten Salt Reactor Analysis," MS, Nuclear Engineering, University of Tennessee, Knoxville, 2019.
- [42] T. J. Price, O. Chvala, and Z. Taylor, "Xenon in molten salt reactors: The effects of solubility, circulating particulate, ionization, and the sensitivity of the circulating void fraction," *Nuclear Engineering and Technology*, 2019, doi: 10.1016/j.net.2019.11.026.
- [43] B. Zhou *et al.*, "Study on dynamic characteristics of activated products source term in secondary loop of integrated molten salt reactor," *Radiation Physics and Chemistry*, vol. 171, 2020, doi: 10.1016/j.radphyschem.2020.108700.
- [44] D. J. Diamond, "Phenomena Important in Molten Salt Reactor Simulations," Brookhaven National Laboratory, BNL-114869-2017-INRE, January 2018.
- [45] J. McFarlane *et al.*, "Fission Product Volatility and Off-Gas Systems for Molten Salt Reactors," Oak Ridge National Laboratory, ORNL/TM-2019/1266, August 2019.
- [46] J. McFarlane, C. F. Weber, M. S. Greenwood, and A. L. Qualls, "Thermochemical and transport properties important to molten salt reactor operation: Off-gas performance and the fission product mechanistic source term," Oak Ridge National Laboratory, ORNL/TM-2018/958, October 2018.
- [47] M. S. Greenwood, B. R. Betzler, A. L. Qualls, J. Yoo, and C. Rabiti, "Demonstration of the Advanced Dynamic System Modeling Tool TRANSFORM in a Molten Salt Reactor Application via a Model of the Molten Salt Demonstration Reactor," *Nuclear Technology*, vol. 206, no. 3, pp. 478-504, 2020, doi: 10.1080/00295450.2019.1627124.
- [48] R. Hu, "A fully-implicit high-order system thermal-hydraulics model for advanced non-LWR safety analyses," *Annals of Nuclear Energy*, vol. 101, pp. 174-181, 2017, doi: 10.1016/j.anucene.2016.11.004.
- [49] G. Zhang and R. Hu, "Development of MSR Transient Safety Analysis Capability in SAM," presented at the 2018 ANS Annual Meeting, Philadelphia, PA, 2018.

- [50] R. Hu, Y. Zhu, and A. Kraus, "Advanced Model Developments in SAM for Thermal Stratification Analysis during Reactor Transients," Argonne National Laboratory, ANL/NSE-18/7, September 2018.
- [51] G. Hu, G. Zhang, and R. Hu, "Reactivity Feedback Modeling in SAM," Argonne National Laboratory, ANL/NSE-19/1, February 2019.
- [52] T. Fei, T. Hua, B. Feng, F. Heidet, and R. Hu, "MSRE Transient Benchmarks using SAM," presented at the PHYSOR 2020: Transition to a Scalable Nuclear Future, Cambridge, U.K., March 29 - April 2, 2020.
- [53] A. M. Leandro, F. Heidet, R. Hu, and N. R. Brown, "Thermal hydraulic model of the molten salt reactor experiment with the NEAMS system analysis module," *Annals of Nuclear Energy*, vol. 126, pp. 59-67, 2019, doi: 10.1016/j.anucene.2018.10.060.
- [54] B. R. Betzler, F. Heidet, B. Feng, C. Rabiti, T. Sofu, and N. R. Brown, "Modeling and simulation functional needs for molten salt reactor licensing," *Nuclear Engineering and Design*, vol. 355, 2019, doi: 10.1016/j.nucengdes.2019.110308.
- [55] J. Jerden, D. Grabaskas, and M. Bucknor, "Development of a mechanistic source term approach for liquid-fueled Molten Salt Reactors," Argonne National Laboratory, ANL/CFCT-19/4, July 2019.
- [56] J. Jerden, "Liquid Fueled MSR Source Term Summary Diagram," ed. Argonne National Laboratory, 2020.
- [57] D. E. Holcomb, A. Huning, A. G. Yigitoglu, M. D. Muhlheim, W. P. Poore, and G. F. Flanagan, "Molten Salt Reactor Initiating Event and Licensing Basis Event Workshop Summary," Oak Ridge National Laboratory, ORNL/TM-2019/1246, July 2019.
- [58] M. H. A. Piro, S. Simunovic, T. M. Besmann, B. J. Lewis, and W. T. Thompson, "The thermochemistry library Thermochemica," *Computational Materials Science*, vol. 67, pp. 266-272, 2013, doi: 10.1016/j.commatsci.2012.09.011.
- [59] S. Delpech, C. Cabet, C. Slim, and G. S. Picard, "Molten fluorides for nuclear applications," *Materials Today*, vol. 13, no. 12, pp. 34-41, 2010, doi: 10.1016/s1369-7021(10)70222-4.
- [60] M. Yamawaki, Y. Arita, K.-i. Fukumoto, M. Taira, and R. Fujimura, "Evaluation Study of Source Term for Severe Accident Analysis of Molten Salt Reactors," *Journal of Plasma and Fusion Research SERIES*, vol. 11, 2015.
- [61] J. Zhang *et al.*, "Redox potential control in molten salt systems for corrosion mitigation," *Corrosion Science*, vol. 144, pp. 44-53, 2018, doi: 10.1016/j.corsci.2018.08.035.
- [62] A. M. Kruizenga, "Corrosion Mechanisms in Chloride and Carbonate Salts," Sandia National Laboratories, SAND2012-7594, September 2012.

- [63] R. I. Olivares, C. Chen, and S. Wright, "The Thermal Stability of Molten Lithium–Sodium–Potassium Carbonate and the Influence of Additives on the Melting Point," *Journal of Solar Energy Engineering*, vol. 134, no. 4, 2012, doi: 10.1115/1.4006895.
- [64] E. Capelli, "Thermodynamic Characterization of Salt Components for Molten Salt Reactor Fuel," Delft University of Technology, 2016.
- [65] Y. Sekiguchi, K. Uozumi, T. Koyama, and T. Terai, "Fundamental study on the vaporization of cesium and iodine dissolved in LiF–NaF–KF molten salt," *Journal of Nuclear Materials*, vol. 522, pp. 136–143, 2019, doi: 10.1016/j.jnucmat.2019.05.014.
- [66] J. D. Stempien, "Tritium transport, corrosion, and Fuel performance modeling in the Fluoride Salt-Cooled High-Temperature Reactor," PhD, Nuclear Science and Engineering, Massachusetts Institute of Technology, 2015.
- [67] W. L. Carter and E. L. Nicholson, "Design and cost study of a fluorination-reductive extraction-metal transfer processing plant for the MSBR," Oak Ridge National Laboratory, ORNL-TM-3579, May 1972.
- [68] R. C. Robertson, "Conceptual Design Study of a Single-Fluid Molten-Salt Breeder Reactor," Oak Ridge National Laboratory, ORNL-4541, June 1971.
- [69] M. W. Rosenthal, P. N. Haubenreich, and R. B. Briggs, "Development Status of Molten-Salt Breeder Reactors," Oak Ridge National Laboratory, ORNL-4812, August 1972.
- [70] C. W. Forsberg, L.-W. Hu, P. F. Peterson, and K. Sridharan, "Fluoride-Salt-Cooled High-Temperature Reactor (FHR) for Power and Process Heat," Massachusetts Institute of Technology, Cambridge, MA, MIT-ANP-TR-157; DOE/NEUP-11-3272, December 2014.
- [71] G. Cao *et al.*, "Fluoride-Salt-Cooled, High-Temperature Reactor (FHR) Materials, Fuels and Components White Paper," University of Wisconsin, Madison, UCBTH-12-003, July 2013.
- [72] D. M. Carpenter *et al.*, "Fluoride-Salt-Cooled, High-Temperature Reactor (FHR) Development Roadmap and Test Reactor Performance Requirements White Paper," Massachusetts Institute of Technology, UCBTH-12-004, June 2013.
- [73] A. T. Cisneros, M. R. Laufer, R. O. Scarlat, N. Zweibaum, and J. E. Seifried, "Fluoride-Salt-Cooled, High-Temperature Reactor (FHR) Subsystems Definition, Functional Requirement Definition and Licensing Basis Event Identification White Paper," University of California, Berkeley, UCBTH-12-001, August 2013.
- [74] A. T. Cisneros, N. Zweibaum, M. R. Laufer, R. O. Scarlat, and J. E. Seifried, "Fluoride-Salt-Cooled, High-Temperature Reactor (FHR) Methods and Experiments Program White Paper," University of California, Berkeley, UCBTH-12-002, May 2013.
- [75] F. Rahnema, D. Diamond, D. Serghiuta, and P. Burke, "Phenomena, gaps, and issues for neutronics modeling and simulation of FHRs," *Annals of Nuclear Energy*, vol. 123, pp. 172–179, 2019, doi: 10.1016/j.anucene.2018.08.035.

- [76] F. Rahnema *et al.*, "The Challenges in Modeling and Simulation of Fluoride-Salt-Cooled High-Temperature Reactors," Georgia Institute of Technology, CRMP-2017-9-001, September 2017.
- [77] F. Rahnema *et al.*, "Phenomena identification and categorization by the required level of multiphysics coupling in FHR modeling and simulation," *Annals of Nuclear Energy*, vol. 121, pp. 540-551, 2018, doi: 10.1016/j.anucene.2018.07.039.
- [78] P. M. Singh *et al.*, "Phenomena Identification and Ranking Table (PIRT) study for metallic structural materials for advanced High-Temperature reactor," *Annals of Nuclear Energy*, vol. 123, pp. 222-229, 2019, doi: 10.1016/j.anucene.2018.08.036.
- [79] H.-C. Lin *et al.*, "Phenomena identification and ranking table study for thermal hydraulics for Advanced High Temperature Reactor," *Annals of Nuclear Energy*, vol. 124, pp. 257-269, 2019, doi: 10.1016/j.anucene.2018.08.038.
- [80] C. W. Forsberg *et al.*, "Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Status, Challenges, and Path Forward," *Nuclear Technology*, vol. 197, no. 2, pp. 119-139, 2017, doi: 10.13182/nt16-101.
- [81] S. T. Lam, J. Stempien, R. Ballinger, and C. Forsberg, "Tritium Management and Control Using Carbon in a Fluoride-Salt-Cooled High-Temperature Reactor," *Fusion Science and Technology*, vol. 71, no. 4, pp. 644-648, 2017, doi: 10.1080/15361055.2017.1290945.
- [82] J. Zhang, "Impurities in Primary Coolant Salt of FHRs: Chemistry, Impact, and Removal Methods," *Energy Technology*, vol. 7, no. 10, 2019, doi: 10.1002/ente.201900016.
- [83] J. J. Lee *et al.*, "Chemical compatibility of silicon carbide in molten fluoride salts for the fluoride salt-cooled high temperature reactor," *Journal of Nuclear Materials*, vol. 524, pp. 119-134, 2019, doi: 10.1016/j.jnucmat.2019.07.001.
- [84] S. R. Greene *et al.*, "Pre-Conceptual Design of a Fluoride Salt-Cooled Small Modular Advanced High-Temperature Reactor (SmAHTR)," Oak Ridge National Laboratory, ORNL/TM-2010/199, December 2010.
- [85] D. E. Holcomb, S. M. Cetiner, G. F. Flanagan, F. J. Peretz, and J. G. L. Yoder, "An Analysis of Testing Requirements for Fluoride Salt-Cooled High Temperature Reactor Components," Oak Ridge National Laboratory, ORNL/TM-2009/297, November 2009.
- [86] N. Zweibaum *et al.*, "Phenomenology, methods and experimental program for fluoride-salt-cooled, high-temperature reactors (FHRs)," *Progress in Nuclear Energy*, vol. 77, pp. 390-405, 2014, doi: 10.1016/j.pnucene.2014.04.008.
- [87] J. Ge *et al.*, "Thermal-hydraulic analysis of a fluoride-salt-cooled pebble-bed reactor with CFD methodology," *Progress in Nuclear Energy*, vol. 91, pp. 83-96, 2016, doi: 10.1016/j.pnucene.2016.01.011.
- [88] N. R. Brown *et al.*, "Preconceptual design of a fluoride high temperature salt-cooled engineering demonstration reactor: Core design and safety analysis," *Annals of Nuclear Energy*, vol. 103, pp. 49-59, 2017, doi: 10.1016/j.anucene.2017.01.003.

- [89]J. E. Seifried, R. O. Scarlat, P. F. Peterson, and E. Greenspan, "A general approach for determination of acceptable FLiBe impurity concentrations in Fluoride-Salt Cooled High Temperature Reactors (FHRs)," *Nuclear Engineering and Design*, vol. 343, pp. 85-95, 2019, doi: 10.1016/j.nucengdes.2018.09.038.
- [90]D. S. Grabaskas, A. J. Brunett, M. D. Bucknor, J. J. Sienicki, and T. Sofu, "Regulatory Technology Development Plan - SFR Mechanistic Source Term Development," Argonne National Laboratory, ANL-ART-3, February 2015.
- [91]D. S. Grabaskas, M. D. Bucknor, and J. L. Jerden, "Regulatory Technology Development Plan - SFR Mechanistic Source Term Metal Fuel Radionuclide Release," Argonne National Laboratory, ANL-ART-38, February 2016.
- [92]D. S. Grabaskas *et al.*, "Regulatory Technology Development Plan - SFR Mechanistic Source Term Trial Calculation," Argonne National Laboratory, ANL-ART-49, October 2016.
- [93]D. Grabaskas, M. Bucknor, and T. Sofu, "Assessment of Regulatory Technology Gaps for Advanced Small Modular Sodium Fast Reactors," Argonne National Laboratory, ANL-SMR-9, May 2014.
- [94]D. Grabaskas, A. J. Brunett, and M. Bucknor, "A Methodology for the Integration of a Mechanistic Source Term Analysis in a Probabilistic Framework for Advanced Reactors," presented at the International Conference on Nuclear Engineering ICONE24, Charlotte, NC, USA, June 26-30, 2016.
- [95]D. Grabaskas, J. Jerden, and M. Bucknor, "A New Computational Tool for Rapid Uncertainty and Sensitivity Analyses of Sodium Reactor Source Term," presented at the ANS Annual Meeting, 2019.
- [96]"Liquid metal coolants for fast reactors cooled by sodium, lead, and lead-bismuth," in "IAEA nuclear energy series; ISSN 1995–7807," International Atomic Energy Agency (IAEA), Vienna, No. NP-T-1.6, 2012.

Nuclear Science and Engineering Division

Argonne National Laboratory
9700 South Cass Avenue, Bldg. 208
Argonne, IL 60439

www.anl.gov

Argonne National Laboratory is a U.S. Department of Energy
laboratory managed by UChicago Argonne, LLC