Survey and Assessment of Computational Capabilities for Advanced (Non-LWR) Reactor Mechanistic Source Term Analysis

**Nuclear Science and Engineering Division,
Argonne National Laboratory**

**Nuclear Energy Safety Technologies Group,
Sandia National Laboratories**

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.

About Sandia National Laboratories

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

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.

Survey and Assessment of Computational Capabilities for Advanced (Non-LWR) Reactor Mechanistic Source Term Analysis

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

Sara Thomas
Chemical & Fuel Cycle Technologies Division, Argonne National Laboratory

Nathan Andrews, Andrew Clark, David Luxat, Michael Higgins, Mariah Smith
Nuclear Energy Safety Technologies Group, Sandia National Laboratories

September 2020

Acknowledgements

This report is part of a larger collaboration between Argonne National Laboratory and Sandia National Laboratories to assist in the development of modern mechanistic source term 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 Bixler, Brad Beeny, Daniel Clayton, Jennifer Leute, Raymond Fasano, Andrew Hahn, and Tina Nenoff from Sandia and Bo Feng, Tanju Sofu, Tom Fanning, Jim Jerden, Nathan Hoyt, Adam Nelson, Micheal Smith, Alisha Kasam-Griffith, and Scott Richards from Argonne for their support in this effort.

Executive Summary

A vital part of the licensing process for advanced (non-LWR) nuclear reactor developers in the United States is the assessment of the reactor's source term, i.e., the potential release of radionuclides from the reactor system to the environment during normal operations and accident sequences. In comparison to source term assessments which follow a bounding approach with conservative assumptions, a mechanistic approach to modeling radionuclide transport, which realistically accounts for transport and retention phenomena, is expected to be used for advanced reactor systems. As the designs of advanced reactors increase in maturity and progress towards licensing, there is a need to advance modeling and simulation capabilities in analyzing the mechanistic source term (MST) of a prospective reactor concept.

In the present work, a survey is provided of existing computational capabilities for the modeling of advanced reactors MSTs. The following reactors are considered: high temperature gas reactors (HTGR); molten salt reactors (MSR) which include salt-fueled reactors and fluoride salt-cooled high temperature reactors (FHR); and sodium- and lead-cooled fast reactors (SFR, LFR). A review of relevant codes which may be useful in providing information to MST analyses is also completed, including codes that have been used for source term analyses of LWRs, as well as those being developed for other aspects of advanced reactor system modeling such as reactor physics, thermal hydraulics, and chemistry. A discussion of MST modeling capabilities for each reactor type is provided with additional focus on important phenomena and functional requirements. Additionally, a comprehensive survey is provided of tools for consequence modeling such as atmospheric transport and dispersion (ATD).

Based on the survey of available modeling and simulation capabilities, key gaps and uncertainties were identified to aid in future code development. In general, there has been extensive research in modeling the reactor physics and thermal hydraulics of advanced reactors. Tools for fuel depletion and fuel performance are relatively robust but gaps remain for proper validation of certain fuel types and geometries. In addition, high-fidelity fuel performance codes have generally not been applied for the assessment of radionuclide behavior in the context of source term analyses. Determination of radionuclide chemical form is important to multiple areas of MST analysis, but modeling tool and database development is ongoing. Significant knowledge concerning aerosol behavior is available from LWR research, but there are areas of uncertainty regarding specific radionuclide aerosols and their chemical forms for different advanced reactor types. In the area of ATD, there are remaining development items concerning near-field dispersion and the behavior of reactor-specific radionuclides in the environment. Lastly, one overarching advanced reactor MST area of consideration is the potential increased importance of radionuclide sources located outside of the traditional core volume. This can be the result of the utilization of a liquid fuel, or the use of auxiliary processing or purification systems.

The findings of the study presented here will be utilized in FY21 to construct an advanced reactor MST modeling and simulation development pathway, which will outline a course to expedite gap resolution and capability expansion.

Table of Contents

1	INTRODUCTION	1
1.1	PROJECT OBJECTIVES	1
1.2	PROJECT METHODOLOGY	2
1.3	REGULATORY CONSIDERATIONS	3
1.4	ASME/ANS NON-LWR PROBABILISTIC RISK ASSESSMENT STANDARD	6
2	MODELING AND SIMULATION OVERVIEW	7
2.1	NEAMS CODES	8
2.2	SCALE SUITE	12
2.3	THERMOCHEMICAL MODELING	14
2.4	AEROSOL MODELING	16
2.5	MELCOR	18
2.5.1	REPRESENTATION OF LWR IN-VESSEL PHENOMENA IN MELCOR	18
2.5.2	REPRESENTATION OF LWR LOWER PLENUM IN-VESSEL PHENOMENA IN MELCOR	22
2.5.3	REPRESENTATION OF LWR EX-VESSEL PHENOMENA IN MELCOR	24
2.5.4	REPRESENTATION OF FISSION PRODUCT RELEASE MODELING IN MELCOR	25
2.6	NRC MODELING AND SIMULATION DEVELOPMENT PATHWAY	26
3	HIGH TEMPERATURE GAS REACTORS	29
3.1	SYSTEM MODELING CAPABILITIES	30
3.2	SOURCE TERM MODELING CAPABILITIES	31
3.2.1	RADIONUCLIDE INVENTORY	36
3.2.2	TRISO FUEL PARTICLE RELEASES	40
3.2.3	CIRCULATING ACTIVITY	42
3.3	CAPABILITY ASSESSMENT	43
4	MOLTEN SALT REACTORS	45
4.1	SYSTEM MODELING CAPABILITIES	47
4.1.1	SALT-FUELED MOLTEN SALT REACTORS	47
4.1.2	FLUORIDE SALT-COOLED HIGH TEMPERATURE REACTORS	53
4.2	SOURCE TERM MODELING CAPABILITIES	55
4.2.1	RADIONUCLIDE INVENTORY	62
4.2.2	SALT THERMOCHEMISTRY	68
4.2.3	BUBBLE TRANSPORT	73
4.2.4	CORROSION AND DEPOSITION MODELING	74
4.2.5	SALT RELEASE	76
4.2.6	AEROSOL MODELING	77
4.2.7	TRITIUM MODELING	79
4.2.8	SALT PROCESSING SYSTEM	82
4.2.9	OFF-GAS SYSTEM	84
4.3	CAPABILITY ASSESSMENT	85
5	SODIUM-COOLED FAST REACTORS	88
5.1	SYSTEM MODELING CAPABILITIES	88
5.2	SOURCE TERM MODELING CAPABILITIES	89
5.2.1	RADIONUCLIDE INVENTORY	93
5.2.2	FUEL MIGRATION AND RELEASE	94
5.2.3	SODIUM POOL	95
5.2.4	AEROSOL MODELING	96
5.2.5	SODIUM FIRES	96
5.3	LEAD-COOLED FAST REACTORS	97
5.4	CAPABILITY ASSESSMENT	98
6	CONSEQUENCE MODELING	101
6.1	OVERVIEW OF CONSEQUENCE MODELING	101

6.2	OFFSITE DISPERSION MODELING SOFTWARE AND ASSOCIATED CAPABILITIES	102
6.3	REGULATORY EFFORTS TO IMPROVE NON-LWR MODELING CAPABILITY OF ATD AND CONSEQUENCE ASSESSMENT SOFTWARE	105
6.4	PHENOMENOLOGICAL AND MODELING GAPS.....	105
6.5	CAPABILITY ASSESSMENT	107
7	CONCLUSION.....	108
	BIBLIOGRAPHY.....	110

Acronyms

Acronym	Definition
ADDER	Advanced Dimensional Depletion for Engineering of Reactors code
AHTR	advanced high temperature reactor (See FHR also)
ANL	Argonne National Laboratory
ANS	American Nuclear Society
AOO	anticipated operational occurrences
ASME	The American Society of Mechanical Engineers
ASTEC	Accident Source Term Evaluation Code
ATD	atmospheric transport and dispersion
ATR	Advanced Test Reactor
AVR	Arbeitsgemeinschaft Versuchsreaktor (German pebble bed reactor)
BDBA	beyond design basis accident
BDBE	beyond design basis events
BWR	boiling water reactor
CALPHAD	Calculation of Phase Diagrams method
CE	continuous energy
CFD	computational fluid dynamics
CFR	circulating fuel reactors
CNWG	Civil Nuclear Energy Research and Development Working Group
CR	control room
CRAB	Comprehensive Reactor Analysis Bundle of codes
CRAM	Chebyshev Rational Approximation Method
CSARP	Cooperative Severe Accident Research Program
CTF	Coolant Boiling in Rod Arrays-Two Fluid code
DBA	design basis accident
DBE	design basis events
DCC	depressurized conduction cooldown
DCF	dose conversion factors
DLOFC	depressurized loss of forced cooling
DNP	delayed neutron precursors
DNS	direct numerical simulations
DOE-NE	U.S. Department of Energy, Office of Nuclear Energy
EAB	exclusion area boundary
EBR-II	Experimental Breeder Reactor II
EPA	U.S. Environmental Protection Agency
ERANOS	European Reactor Analysis Optimized code System
FAI	Fauske & Associates, Inc
FATE	Facility flow, Aerosol, Thermal, and Explosion code
FFMEA	functional failure mode and effect analyses
FFTF	Fast Flux Test Facility
FHR	fluoride salt-cooled high temperature reactor
FOA	funding opportunity announcement
GEMS	Gibbs Energy Minimization Software
HALEU	high assay low enriched uranium
HEU	high enriched uranium
HFCA	high-fidelity criticality analysis
HTGR	high temperature gas-cooled reactor
HTR	See HTGR
IE	initiating events
IFR	Integral Fast Reactor
KAERI	Korea Atomic Energy Research Institute
LANL	Los Alamos National Laboratory
LBE	licensing basis events
LEU	low enriched uranium
LFR	lead-cooled fast reactor
LIBS	laser-induced breakdown spectroscopy
LLNL	Lawrence Livermore National Laboratory
LMP	Licensing Modernization Project
LMR	liquid metal-cooled reactor
LPZ	low-population zone
LWR	light-water reactor
MAAP	Modular Accident Analysis Program
MACCS	MELCOR accident consequence code system
MAEROS	multicomponent aerosol simulation code
MAR	material-at-risk
MCCI	molten core concrete interaction
MCFR	molten chloride fast reactor
MCNP	Monte Carlo N-particle Transport code
MELCOR	Methods for Estimation of Leakages and Consequences of Releases code suite

MG	multigroup
MGD	multigroup diffusion
MHTGR	modular high temperature gas reactor
MLD	master logic diagram
MOC	method of characteristics
MOOSE	Multiphysics Object Oriented Simulation Environment code framework
MOSART	MOlten Salt Actinide Recycler & Transmuter reactor concept
MOX	mixed oxide fuel
MPACT	Michigan Parallel Characteristics Transport code
MQM	modified quasichemical model
MSBR	Molten Salt Breeder Reactor
MSDR	Molten Salt Demonstration Reactor
MSFR	Molten Salt Fast Reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
MST	mechanistic source term
MSTDB	Molten Salt Thermodynamic Database
NARAC	National Atmospheric Release Advisory Center
NEAMS	Nuclear Energy Advanced Modeling and Simulation program
NEPA	National Environmental Policy Act of 1969, as amended
NEUP	Nuclear Energy University Program
NGNP	Next Generation Nuclear Plant
NMSS	NRC Office of Nuclear Material Safety and Safeguards
NRC	Nuclear Regulatory Commission
NRR	NRC Office of Nuclear Reactor Regulatory
ORIGEN	ORNL Isotope Generation and Depletion code
ORNL	Oak Ridge National Laboratory
PARCS	Purdue Advanced Reactor Core Simulator code
PBMR	pebble bed modular reactor
PBR	pebble bed reactor
PCV	primary containment vessel
PHSS	pebble handling and storage system
PIRT	phenomena identification and ranking table
PKE	point kinetic equations
PLOFC	pressurized loss of forced cooling
PNNL	Pacific Northwest National Laboratory
POSCA	Plate-Out Surface and Circulating Activities code
PRA	probabilistic risk analysis
PRK	point reactor kinetics
PyC	pyrolytic carbon
RADTRAD	Radionuclide, Transport, Removal, and Dose Estimation code
RAMP	NRC Radiation Protection Computer Code Analysis and Maintenance Program
RANS	Reynolds-Averaged Navier-Stokes equations
RASCAL	Radiological Assessment System for Consequence Analysis code
RCCS	reactor cavity cooling system
RELAP	Reactor Excursion and Leak Analysis Program code
RES	NRC Office of Nuclear Regulatory Research
RPV	reactor pressure vessel
RRM	Radionuclide Release Module within the FATE code
SAMOFAR	Safety Assessment of the Molten Salt Fast Reactor project
SCALE	Standardized Computer Analyses for Licensing Evaluation code suite
SDC	safety design criteria
SFR	sodium-cooled fast reactor
SHARP	Simulation-based High-fidelity Advanced Reactor Prototyping code toolkit
SNAP	Symbolic Nuclear Analysis Package code
SNL	Sandia National Laboratories
SOARCA	State-of-the-Art Reactor Consequence Analyses project
SRT	Simplified Radionuclide Transport code
SSC	structures, systems, and components
TFHR	transportable fluoride high temperature reactor
TMSR-SF	solid-fueled thorium-based molten salt reactor
TRACE	TRAC RELAP Advanced Computational Engine code
TRACER	Transport Phenomena of Radionuclides for Accident Consequence Evaluation of Reactor code
TRANSFORM	Transient Simulation Framework of Reconfigurable Modules code
TRISO	tri-structural isotropic fuel
TSLM	two-sublattice ionic liquid model
ULOF	unprotected loss of flow
ULOHS	unprotected loss of heat sink
UOCA	unprotected overcooling accident
URIA	unprotected reactivity insertion accident
VERA-CS	Virtual Environment for Reactor Applications, Core Simulator code suite

1 Introduction

A number of industry vendors are leading a resurgence in U.S. 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]. An MST attempts to realistically model the potential 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 for the advanced reactor industry by performing an assessment of the capabilities of currently available codes for the evaluation of advanced reactor MSTs.

1.1 Project Objectives

The current work is an integral 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 for the advanced reactor industry, as part of the U.S. Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. The MST development pathway outlined in Figure 1-1 is used to guide the project in advancing modeling and simulation tool readiness to support advanced reactor licensing applications.

The MST development pathway begins with the identification of radionuclide sources within the reactor system of interest, which potentially includes both core and non-core sources. This is followed by the identification of potential radionuclide release, transport, and retention phenomena, including radionuclide barriers. Once identified, modeling and simulation tools can be developed to account for those transport phenomena and barrier performance characteristics of high importance to offsite consequence or other metrics of interest. These tools are then utilized to evaluate specific event sequences as part of regulatory reactor licensing applications.

Previous work has focused on the first two steps of the MST development pathway, as outlined below:

- **Functional Requirements for the Modeling and Simulation of Advanced (Non-LWR) Reactor Mechanistic Source Term (ANL/NSE-20/17) [2]**
 - A preliminary development of functional modeling requirements for advanced reactor MST modeling and simulation tools
- **Mechanistic Source Term Considerations for Advanced Non-LWRs (SAND2020-6730) [3]**
 - An examination of potential licensing basis events (LBEs) for advanced reactors using function trees to identify event sequences of interest for MST development.

Figure 1-1: MST Development Pathway

The current work focuses on the third step of the MST development pathway, which is the modeling and simulation of radionuclide transport phenomena and barrier performance. The objective of the current work is to provide an assessment of currently available modeling and simulation tools that could be utilized for advanced reactor MST analyses. Through this survey, capability gaps can be identified for future prioritization and resolution.

In general, the current work is concentrated on MST assessments as part of the evaluation of radionuclide releases associated with accident event sequences and not routine operational releases of effluents. However, there are areas where the modeling and simulation capabilities may overlap, such as the assessment of tritium transport in certain types of molten salt reactors. In addition, for some advanced reactors that are in the preliminary design stage, operational releases may not yet be fully resolved and also require MST tools for further investigation.

1.2 Project Methodology

The current effort follows the general process outlined in Figure 1-2. First, a comprehensive survey of available modeling and simulation tools is conducted. As will be described in Section 2, this survey includes both high and low fidelity codes with capabilities spanning from radionuclide generation to radionuclide transport and offsite dispersion. Once the landscape of available modeling and simulation tools is established, their capabilities are compared to the functional requirements outlined in ref [2], within the context of the potential event sequences described in ref [3]. Based on this comparison, MST modeling and simulation capability gaps are identified both globally and for each advanced reactor type. As the importance of specific functional requirements is not yet known or varies by particular reactor design and event sequence, only a preliminary assessment of the identified capability gaps is provided here. Future efforts will seek to prioritize the identified gaps for resolution.

Figure 1-2: Overview of Project Methodology

The remainder of this section describes regulatory considerations associated with MST development and associated MST requirements from the ASME/ANS Non-LWR Probabilistic Risk Assessment (PRA) Standard. As outlined in Figure 1-3, this is followed by an overview of the existing code landscape in Section 2, including a description of the differing roles a modeling and simulation tool may play within the development of an MST. Sections 3 through 5 evaluate the available tools against the functional requirements for each of the main advanced reactor types. Section 6 performs a similar assessment but only for offsite dispersion analysis, which is viewed as applicable to all reactor types. Lastly, Section 7 provides a summary of the report findings, including an overview of identified gaps.

Figure 1-3: Report Outline

1.3 Regulatory Considerations

Source term considerations are a primary focus of the reactor licensing process, as the protection of the public and environment against accidental releases of radionuclides is central to the mission of the U.S. Nuclear Regulatory Commission (NRC). Historically, source term evaluations for light water reactors (LWRs) were developed based on prescribed core melt scenarios, with conservative, non-mechanistic assumptions regarding the release of radionuclides [4]. With the publication of NUREG-1465 in 1995 [5], the NRC began a shift towards attempting to account for specific radionuclide transport and retention phenomena in LWR accident scenarios based on insights gained from experimentation and modeling. This was followed by Regulatory Guide 1.183 [6], which provided guidance on the development of an alternative source term for LWRs following the initial mechanistic guidance utilized in NUREG-1465. For advanced non-LWRs, the NRC has long stated an expectation of the use of MST analyses as part of the reactor licensing

process [7, 8]. Therefore, recent developments in the establishment of a licensing pathway for advanced reactors has brought additional focus to MST development.

Central to MST considerations for advanced reactor licensing is the recent approval of the Licensing Modernization Project (LMP) process by the NRC [9]. The LMP approach, described in ref [10], provides guidance on a technology-inclusive, risk-informed, performance-based process for advanced reactor licensing. Specifically, risk information derived from the PRA is utilized to guide the identification and categorization of LBEs, the classification of structures, systems, and components (SSCs), and the evaluation of the adequacy of defense-in-depth. The frequency versus consequence targets shown in Figure 1-4 provides guidance in this process.

Figure 1-4: LMP Frequency-Consequence Targets [10]

As offsite dose is the central consequence metric in the LMP process, and therefore also the central consequence metric of the PRA, the development of event sequence-specific MST analyses is vital to the process. As shown in Figure 1-4, consequence targets are utilized for each category of LBEs: anticipated operational occurrences (AOOs), design basis events (DBEs), and beyond design basis events (BDBEs). Therefore, if a radionuclide release is possible for an LBE within each of these categories, some type of source term assessment is necessary to provide insights into offsite consequences and justify the satisfaction of the consequence targets. In addition, LBEs could also include the release of radionuclides from non-core sources of radioactivity, since the ASME/ANS Non-LWR PRA Standard, described in the following subsection, allows the inclusion of these sources within the PRA. Due to these factors, the development of MST analyses for advanced reactors may go beyond the historical focus of core-

centric severe accident analysis, but include higher frequency, less severe core events and radionuclide releases associated with support systems and fuel handling.

The NRC has recently sponsored several studies to assist with the development of source term analyses for advanced reactors. A barrier-centric analysis approach, outlined in Figure 1-5, was suggested as a possible pathway to characterize radionuclide transport and retention that would be generally applicable to all advanced reactor designs [11]. In addition, a technology-inclusive source term determination methodology was developed [12], which is depicted in Figure 1-6. This approach outlines the possible use of bounding, conservative source term analyses, in place of detailed mechanistic assessment, if it can be shown that the specified radiological controls are met. If an MST is required or desired, the PRA and LMP process is utilized to inform MST development.

Figure 1-5: Illustration of Source Term Reduction through Multiple Barriers [11]

Figure 1-6: Technology-Inclusive Source Term Determination Pathway [12]

1.4 ASME/ANS Non-LWR Probabilistic Risk Assessment Standard

The ASME/ANS PRA Standard for Advanced Non-LWR Nuclear Power Plants establishes the technical requirements for PRAs used in supporting risk-informed decisions. A trial use version of the standard was first published in 2013 [13]. Based on pilot user feedback, a revised version of the standard has been prepared, with formal publication expected in late 2020.

The non-LWR PRA standard includes separate technical elements for MST assessments and radiological consequence analyses, with both high-level and support requirements. The requirements contained within these sections are utilized by the current work for guidance on the necessary capabilities of MST modeling and simulation tools. Several authors of the current report are involved in the development of the standard, including the MST and radiological consequence elements. While not yet publicly available, the revised technical requirements to be included in the new version of the standard were used to aid in the MST capability assessment described herein.

2 Modeling and Simulation Overview

Source term modeling and simulation can require assessments from a variety of different types of computational tools, whether as part of the analysis itself or for the creation of necessary inputs. Before beginning a detailed discussion of modeling and simulation tools, it is important to clarify expectations and experience regarding source term models. Source term analysis for LWRs has historically focused on risk-significant scenarios in which radiological release to the environment could occur, including scenarios with extensive fuel melting as well as more extensive core degradation. In these scenarios, there are a multitude of complex physical and chemical phenomena that impact radionuclide release and transport. As highlighted in Section 1.3, initial source term assessments as part of early LWR licensing efforts utilized conservative models and bounding analyses rather than addressing these phenomena and their associated uncertainties in detail.

The NRC developed a mechanistic source term methodology for LWRs with the release of NUREG-1465 in the 1990s, and state-of-the-art accident analysis codes (e.g., MELCOR, MAAP, ASTEC, etc.) were developed to represent these physical and chemical phenomena mechanistically. However, it is important to note the meaning of mechanistic within this context. For source term analyses, “mechanistic” typically implies an attempt to capture the relevant radionuclide transport and retention phenomena rather than using conservative or bounding assumptions. Mechanistic source term analyses utilize models at the level of fidelity required to resolve the prevailing uncertainties and support regulatory decisions. Since source term assessments are primarily influenced by the representation of physical and chemical phenomena at an engineering scale, these analyses focus on mechanistic representation of phenomena and their interactions. These methods generally do not focus on refinement of spatial and temporal resolution in simulations, as regulatory decisions are typically not influenced by errors introduced from coarse spatial or temporal resolution.

As source term analyses include relatively large uncertainties regarding the phenomena of interest, detailed uncertainty assessments have been essential to ensure that regulatory decisions are robust. The uncertainties may be due to a lack of knowledge or data related to the phenomena or uncertainty concerning the evolution of the transient scenario itself. The state-of-the-art codes mentioned above have thus been developed to capture the spectrum of phenomenological uncertainties and accident scenarios necessary to support and expedite sensitivity and uncertainty analyses used to inform risk evaluations.

That is not to suggest that high-fidelity tools are not useful for source term analyses. Such tools may be used to resolve phenomena and result in simplified approaches that can be integrated into low-fidelity tools or as a benchmark of previously developed simplified models. These tools have primarily been applied to assess the likelihood and consequences arising from specific phenomena that can lead to rapid energy release and sudden failure of one or more fission product barriers (e.g., ex-vessel steam explosions in LWR systems). Such tools have also been utilized to develop reduced-order models that can be applied directly in systems-modeling tools such as MELCOR (see Reference [14] for further details on the application of CFD to developing reduced-order models to capture PWR hot leg natural circulation in MELCOR simulations).

The remainder of this section provides an overview of the current modeling and simulation landscape, as a foreword to the reactor-specific discussions in Sections 3 through 5. This includes an overview of codes historically used for LWR source term assessment along with newly available capabilities for advanced reactor modeling and simulation including both high-fidelity tools and system modeling codes, which may have application to advanced reactor mechanistic source term analysis. The following topics are covered in this section for the reasons described below:

- **NEAMS Codes:**
 - The NEAMS program offers a suite of high-fidelity modeling and simulation tools that could potentially be utilized as part of source term analyses. This could include their use for the creation of necessary input conditions, the development of modeling insights that could be integrated into low-fidelity tools, or direct integration into analyses.
- **SCALE:**
 - *SCALE* is a popular code system widely utilized for reactor physics, activation, depletion, and sensitivity analyses. This information is often needed for the determination of initial radionuclide conditions for source term analyses.
- **Thermochemical Modeling:**
 - Thermochemical phenomena are an important consideration into the assessment of radionuclide chemical forms, which can impact transport and retention. There are a host of modeling and simulations methods and tools available for these analyses.
- **Aerosol Modeling:**
 - Many source term assessments require the analysis of radionuclide aerosol behavior at some point during the analysis. An overview of aerosol phenomena and modeling approaches is provided here, with reactor type-specific considerations described in corresponding sections.
- **MELCOR:**
 - *MELCOR* has historically been utilized by the NRC for assessment of severe accident behavior and source term analyses. The NRC is now planning on adapting *MELCOR* for the assessment of non-LWR source terms.
- **NRC Modeling and Simulation Development Pathway:**
 - The NRC has recently established a modeling and simulation development pathway for the licensing of non-LWR designs. This subsection provides an overview of the proposed approach, with reactor with reactor type-specific considerations described in corresponding sections.

2.1 NEAMS Codes

The DOE NEAMS program supports and develops a wide range of computational codes aimed at all applications of nuclear reactor modeling [15]. NEAMS is developing capabilities in modeling neutron transport, thermal hydraulics, structural mechanics, fuel performance, and chemistry. Many of these codes are applicable to advanced reactor concepts and have relevance or potential use in source term modeling, as will be discussed here. This subsection is not meant to promote any one code for use in an MST development, but rather showcase the capabilities being developed under NEAMS, as some MST strategies may require the potential use of codes like these. An overview of some of these codes is provided in Figure 2-1 [15].

MOOSE, or *Multiphysics Object Oriented Simulation Environment*, is a parallel computational framework built with systems of coupled, nonlinear partial differential equations in mind [16]. Because such complex mathematical models are common in the simulation of physical processes, especially nuclear processes, many NEAMS codes are built on the *MOOSE* framework. Additionally, *MOOSE* includes a wide variety of physics modules that can be leveraged to build complex multiphysics simulations, supporting development in new areas of high-fidelity modeling. With all the recent advances in modern computing, *MOOSE* provides a highly scalable framework with robust solution algorithms allowing for multiscale, multi-physics simulations which can utilize these computing resources [17].

Figure 2-1: Overview of NEAMS Codes [15]

Fuel Performance Modeling

BISON [18] and *MARMOT* [19] are solid fuel performance codes with much of their initial development for UO_2 fuel modeling, but also provide modeling capabilities in other fuel types including TRISO fuel [20] and metallic fuels [21, 22]. Ongoing activities include code training and model development support [15]. Built on the *MOOSE* framework, *BISON* is continuum scale, finite element-based, and applicable to both steady state and transient fuel behavior. It can be used to analyze either 2D axisymmetric or 3D geometries. Capabilities include analysis of temperature- and burnup-dependent thermal properties, solid and gaseous fission product swelling, fuel densification, fuel thermal and irradiation creep, pellet fracture, fission gas production and release, fuel-clad gap mechanics, and gap/plenum gas behavior during irradiation. Also built on the *MOOSE* framework, *MARMOT* is a mesoscale phase field code and allows for capabilities such as the effect of grain and void growth on important material physical properties. For solid fueled reactors, the impact of mechanistic fuel performance modeling on source term analysis cannot be understated [23]. Therefore, coupling source term models to codes such as these, or using the output results of these simulations as input for source term models, is a possible starting point for mechanistic source term analyses.

Utilizing benefits of the *MOOSE* framework, *BISON* has also been coupled to the neutron transport solver *RATTLESNAKE*, providing pin level temperature fields for feedback effects on the neutron flux [24]. *RATTLESNAKE* is a 3D finite element-based neutron transport application featuring a deterministic radiation transport solver used to capture the impact of temperature and material density changes on time-dependent flux distribution, reaction rates and power profile. It can be used for lattice, pebble-bed and hexagonal fuels.

Neutronics Modeling

The *PROTEUS* neutronics suite is a high-fidelity reactor physics simulation toolset including multi-group cross section generation, two highly scalable neutron transport solvers, and a nodal transport solver. Included in the suite is *MC²-3*, a multigroup cross section generation code for fast reactor analysis [25]. Using different methods to solve the neutron transport equation, the three deterministic solvers include the discrete ordinate method (*S_N*) [26, 27], the method of characteristics (*MOC*) [28], and the nodal diffusion method (*NODAL*) [29]. It can also be coupled to *PERSENT*, a perturbation and sensitivity analysis code [30]. There are ongoing efforts in developing other neutronics solvers and codes as well. For example, *Griffin* is currently being developed for all advanced (non-LWR) reactors, and its use will be recommended for reactor physics modeling if coupling with other codes is envisioned (i.e., multiphysics).

Thermal Hydraulics & Computational Fluid Dynamics Modeling

Several thermal hydraulic codes are being developed under NEAMS, offering computational capabilities at plant, engineering, and fine scales, as can be seen in Figure 2-2 [15]. The *System Analysis Module (SAM)* is a plant-level system analysis tool for advanced reactors focusing on system thermal hydraulics, and recently many updates have been implemented to ease coupling in the *MOOSE* multi-physics framework [31, 32]. There is utility in using *SAM* for reactors with liquid coolant such as SFR, LFR, MSR, and FHR, and it has some application to gas reactors such as HTGR. There have been many developments to support transient analysis of advanced reactors using *SAM*, as well as increased user and developer engagement for the code from various licensees in industry, academia and the national labs, and in 2019, the NRC formally stated its intent to use *SAM* for design basis event analysis [33]. Initially developed for LMRs and FHRs, enhancements to the code have been made for application to MSRs, where a circulating fuel salt has unique effects on reactivity feedback modeling and system-level transient analysis. Efforts are ongoing in support of all of these reactor concepts, as will be discussed later.

Pronghorn is an engineering-level system analysis tool for advanced reactor thermal hydraulics, especially made for systems with flow through porous media such as pebble bed reactors like HTGR and FHR [34]. Using finite-element porous media models, *Pronghorn* does not provide the same level of flow detail around pebbles as CFD, but generally predicts fluid flow and fluid-solid heat transfer fairly well [35]. This compromise between plant scales and finer scales allows for an improvement in thermal hydraulics modeling in most situations for this unique core design.

For problems where a finer scale is needed to understand flow phenomena, the open source *Nek5000* is a parallel, spectral element, computational fluid dynamics code that accepts 2D or 3D meshes and couples to other codes through the *MOOSE* framework [36]. Its use with the spectral element method for wall-resolved large eddy simulations of LWR, LMR and MSR reactor core

flows has been shown [37, 38]. Although this higher computational expense may not be warranted for every source term modeling strategy, a high-fidelity reference solution may be necessary for certain problems such as complex species transport flows. Additionally, *Nek5000*, *Pronghorn*, and other codes have been cohesively coupled for multiscale problems using the *MOOSE* framework, proving the utility of using codes of similar software structure [39].

Figure 2-2: Scale of Analysis Offered by NEAMS Thermal Hydraulics Codes [15]

Structural Mechanics Modeling

Grizzly is a *MOOSE*-based structural mechanics simulation tool with the ability to model aging processes in plant systems, components and structures [40]. Advanced reactor development with *Grizzly* is ongoing, as there is interest in utilizing its capabilities for engineering-scale models at high temperatures [15]. Some examples of its use for LWRs include analyzing the effects of reactor pressure vessel embrittlement and aging of reinforced concrete structures, but its capabilities are being expanded for application to advanced reactors to address structural component integrity and degradation [41]. For example, this includes modeling crack propagation in a variety of structural materials for realistic fracture modeling, which obviously has serious implications for radionuclide releases. *Diablo* is another structural mechanics modeling tool under NEAMS, specifically an implicit finite element code with an emphasis on coupled structural and thermal analysis capabilities [42].

With the goal of coupling multiple NEAMS codes together to perform multi-physics calculations, the *SHARP* design and analysis toolkit was developed [43]. Neutronics is simulated with *PROTEUS*, fluid dynamics is simulated with *Nek5000*, and structural mechanics is simulated with *Diablo* [44]. It has been demonstrated for various problems with SFRs, LFRs, and fast-spectrum MSRs, as will be discussed later [45].

Chemistry Modeling

Several chemistry related codes or databases are currently being developed for advanced reactor applications. Today, many technology developers are taking advantage of different fuel types, coolant types, structural and component materials, and reactor operating conditions to utilize the full potential of various advanced reactor concepts. But this is a stark transition for nuclear reactor chemists where much of the focus of the past 50 years has been on water chemistry. In order to accommodate the unique chemistry modeling needs of advanced reactors, a thermochemical equilibrium tool called *Thermochimica* has been developed for chemical

speciation calculations [46, 47]. Initially developed as an upgrade to more traditional Gibbs free energy minimization solvers for LWR nuclear fuel applications, it has seen recent development for advanced reactors such as thermochemical modeling of molten salt systems [48]. And it can interface with the *MOOSE* framework, evidenced by coupling to *BISON* for fission product thermochemistry calculations in UO_2 [49].

The *Molten Salt Thermodynamic Database (MSTDB)* is being developed to compile molten salt thermodynamic data and provide it as input to *Thermochimica* and other applications [50, 51]. The need for a database of thermodynamic parameters that is not only comprehensive in salt systems but also accurate is absolutely vital for any molten salt reactor MST strategy. The topic of thermochemical modeling tools is discussed in more detail in Section 2.3, and the topic of salt chemistry is discussed in Section 4.2.2 for application to MSRs and FHRs.

Finally, the *Yellowjacket* code is in its infancy but being developed as a thermochemistry solver to model corrosion in advanced reactors by directly coupling thermodynamic equilibrium and kinetics with phase field models [52]. The valuable information provided by such a tool includes not only the impact of corrosion on the structural integrity of components, but also the dissolution of corrosion products from the component into the coolant salt, which can be a radionuclide that is now in the circulating source term.

2.2 SCALE Suite

SCALE is a modeling and simulation suite for nuclear safety analysis and design that is developed, maintained, tested, and managed by ORNL [53-57]. Included in the suite are a family of modular codes that provide capabilities in criticality safety, reactor physics, radiation shielding, radioactive source term characterization, and sensitivity and uncertainty analysis. *SCALE* can be used to provide initial reactor conditions for a given design, namely those conditions related to neutronics, isotopic composition, and radiation transport of the system. The majority of development in *SCALE* has been for LWR systems and oxide fuels, but *SCALE* can be, and has been, used to build reactor models for more complex, advanced reactors with unique material and fuel compositions. The inherently modular nature of the codes allows the user to build a reactor system model for many functional analyses and geometries. The code system is already used for many analyses related to nuclear security and safeguards, spent fuel transportation and storage, and other uses by the NRC. For all of these reasons, the NRC has recently outlined plans for continued development of *SCALE* for advanced reactors, with the hopes of coupling output to *MELCOR* for severe accident progression [58]. A brief introduction to some of the functionality that *SCALE* provides for a potential source term assessment is provided.

The main components in *SCALE* that are utilized to build a reactor system model include: a family of codes used to specify material compositions and process nuclear data; a family of radiation transport solvers for modeling reactor physics, either stochastically or deterministically; a family of material irradiation and decay solvers; and a family of codes used for sensitivity and uncertainty calculations. There is no built-in chemical or physical radionuclide transport functionality in *SCALE*, but *SCALE* can be used in a MST analysis to model the normal operating conditions or accident conditions, such as the changing radionuclide composition owing to depletion of the nuclear fuel and activation of surrounding materials, as well as parameters related to power, reactivity, and temperature. These characteristics are unique to the reactor concept and

constitute the very important initial conditions necessary for any source term assessment of an advanced reactor. Knowledge of the radionuclide inventory in the fuel and surrounding materials is the first and most important functional requirement for modeling source term [2].

TRITON is a reactor physics and depletion tool within *SCALE* which has multiple options for transport solvers including those using Monte Carlo methods such as *KENO* and *Shift*, and those using deterministic methods such as *NEWT* and *XSDRNPM* [53]. The *XSPROC* code is used for material specification and cross section processing to provide problem-dependent microscopic and macroscopic multigroup cross section data for *TRITON*. The reactor's design and characteristics must be taken into account when building the model, importantly including core materials and geometries. *TRITON* outputs a library file which can be used as input to *ORIGEN*.

ORIGEN (*ORNL Isotope Generation and Depletion Code*) has seen much development and support since it was first introduced in 1973 [59]. Recently, a second solver option based on the Chebyshev Rational Approximation Method (CRAM) has improved speeds for systems with larger datasets, but the MATREX solver still results in speeds on the timescale of seconds [53]. Taking *TRITON* depletion data as input, the code calculates depletion, decay, and activation as a function of irradiation time and material data. The code has functionality in tracking over 2,200 nuclides during irradiation, transmutation, decay, and/or depletion, where the initial nuclear data is sourced mostly from ENDF/B-VII. It can also model continuous addition or removal of materials at each timestep, representing potential fissile material additions or processing systems such as fission product separation streams for an MSR. As a standalone code, *ORIGEN* needs a library file of one or more problem-dependent transition matrices which can also be created with the *COUPLE* module, if a robust reactor system model is not created with *TRITON*. The *COUPLE*-created library files contain one-group reaction coefficients created from arbitrary flux spectra and arbitrary one-group cross sections. It is worth noting that *ORIGEN* does not take reactor geometries into account, as it is only a point depletion solver, and the solution to the transition matrix can be interpreted as a solution at a point in space or the spatial average over some volume.

OPUS is a post-processing module within the *ORIGEN* family which can sort the data by nuclide, element, activity, decay info, and other parameters, as well as convert the data to a format easily readable by graphic-creating software. *ORIGEN*'s default output options include grams, moles, activity, decay heat, radiological hazard factors, or radiation source spectra, but for a source term assessment, this additional processing of the *ORIGEN* data is necessary for carefully curated or sorted radionuclide concentrations. This data can then be used by a thermochemical modeling tool and a radionuclide transport tool in a source term modeling effort.

For LWRs and some advanced reactor configurations, *SCALE* has the capability to transfer data files to the severe accident analysis codes *MELCOR* and *MACCS* with the following information: time and space-dependent inventory, fundamental nuclide and decay data, and nuclide effective generation and destruction rate data [58]. The analyst has the ability to create datasets with any level of spatial fidelity. Further research and development are needed to bring this same level of functionality and fidelity, with sensitivity analyses, to all advanced reactors.

While *SCALE* has historically been developed for LWR applications and solid oxide fuels, recent developments are being made toward advanced reactor concepts such as HTGR, SFR, FHR and

MSR. Benefiting from a modular code suite and a large user base, *SCALE* has potential for continued support in developing reactor physics models specific to advanced reactor designs. These reactor physics models can be used to better understand the safety characteristics of the unique reactor, specifically the initial conditions used in a source term model, i.e., radionuclide inventory. It is worth reiterating that this radionuclide inventory is unique and should be calculated for each reactor design, based on many contributing factors including fissile material and enrichment, moderator type and density, neutron flux and energy spectra, and spatial characteristics, as well as other materials in the core that can produce activation products. The ability to calculate this inventory easily based on these input parameters for a given design of the advanced reactor is important to the mechanistic source term analysis.

2.3 Thermochemical Modeling

Source term analyses require knowledge of the chemical speciation and transport and retention behavior of key radionuclides within a nuclear reactor facility. Radionuclide speciation and phase distribution can be modeled thermodynamically by Gibbs free energy minimization calculations. A chemical equilibrium approach to source term assessment was recently applied to advanced reactors such as SFRs [60, 61] and MSRs [48, 62]. The input and information flow involved in thermodynamic modeling for a generic source term assessment is shown in Figure 2-3.

Figure 2-3: Gibbs Free Energy Minimization Pathway for a Source Term Model

A Gibbs free energy minimization calculation requires the elemental composition, temperature, and pressure of the system at a specific time point during reactor operation or a transient scenario.

The possible species and phases that may form must also be specified as input. The accuracy of the calculation relies on a complete thermodynamic database of system component Gibbs free energies, which can be computed from standard enthalpies, standard entropies, specific heat capacities, and Gibbs excess energy functions. The Gibbs excess energies account for non-ideality and can be quantified with activity coefficients or more advanced solution models. The output provides the equilibrium concentrations of the species and phases that were identified as input. Typically, the user categorizes and filters the output so that only the relevant radionuclides species above a specified threshold concentration are included in the source term model.

A variety of commercial and open-source software for the calculation of chemical speciation by Gibbs energy minimization exist (Table 2-1). The software with more advanced solution modeling capabilities (e.g., *FactSage*, *ThermoCalc*, and *Thermochimica*) are appropriate for reactor concepts that involve high concentrations of radionuclides in a solution phase (e.g., liquid-fueled MSRs). The open-source codes are suitable for direct coupling to other source term modeling codes. *Thermochimica* has been directly coupled to the solid-fuel performance code *BISON* to model the diffusion of oxygen in UO₂ fuel [49] and to the isotopic depletion software *ORIGEN* to model fission product vaporization based on MSRE power history [48]. Commercial software has aided the development of source term models for SFRs [60], although functionality was limited due to the inability to directly integrate with other codes.

Table 2-1: Summary of Select Gibbs Free Energy Minimization Software

Software	Open Source	Solution modeling capabilities	Databases	Algorithm for minimization	Language	Ref.
<i>FactSage</i>	No	Many (see Bale et al., 2016)	Internal	Lagrange multiplier method (Eriksson and Hack, 1990)	N/A	[63, 64]
<i>GEMS</i>	Partly	Ideal mixtures, regular solution model, Redlich-Kister	Internal, <i>HERACLES</i> for nuclear applications	Lagrange multiplier method (Karpov et al., 2001)	C/C++ (<i>GEMS3K</i>)	[65-67]
<i>HSC Chemistry</i>	No	Ideal mixtures, Redlich-Kister (with add-in)	Internal	Lagrange multiplier method (White et al., 1958)	N/A	[68, 69]
<i>OpenCalphad</i>	Yes	Ideal mixtures, regular solution model, compound energy formalism	Compatible with <i>Thermodynamic Database (TDB)</i> file format	Lagrange multiplier method (Hillert et al., 1981)	<i>Fortran standard 2008</i>	[70, 71]
<i>Thermo-Calc</i>	No	Many (see Andersson et al., 2002)	Internal	Lagrange multiplier method (Hillert et al., 1981)	Programming interface (C++, <i>Matlab</i> , <i>Fortran</i>)	[72]
<i>Thermochimica</i>	Yes	Ideal mixtures, regular solution model, compound energy formalism, modified quasichemical model	Compatible with <i>ChemSage</i> file format, <i>MSTDB</i>	Initialization procedures/ Lagrange multiplier method (Piro et al., 2013 and Poschmann et al., 2020)	<i>Fortran 90/2003 standard</i>	[49, 73]
<i>Yellowjacket</i>	Yes	Under development (see Bajpai et al., 2020)	Compatible with <i>ChemSage</i> file format, <i>MSTDB</i>	Under development (see Bajpai et al., 2020)	C++	[52]

It is important to note that these thermodynamic models assume chemical equilibrium, and thus, do not account for reaction kinetics, chemical and thermal gradients, or inhomogeneity of phases and species. In many cases, the assumption of instantaneous local equilibrium can be a good approximation for nuclear fuel systems, which are typically well-mixed due to the high system temperature and stochastic nature of nuclear fission. For example, kinetic studies on the fission product inventory of solid UO_2 fuel showed that chemical equilibrium in the vapor phase (i.e., fuel cladding gap and the containment atmosphere) is reached rapidly (< 0.1 s) at elevated temperatures (> 1000 K) [74, 75]. Less is known on liquid phase reaction kinetics and the homogeneity of molten coolant or molten fuel salt. Studies to elucidate chemical inhomogeneity as well as the time scales to reach chemical equilibrium in molten coolant or molten fuel salt are required to better understand the limitations of thermodynamic modeling in source term assessments for these reactor concepts. Assuming instantaneous equilibrium is conservative for some thermodynamically driven phenomena like volatilization or precipitation. However, radionuclide transport models for advanced reactor source terms should be as realistic as possible to not obscure the limitations of any mechanism or barrier [76].

2.4 Aerosol Modeling

The vast majority of potential radionuclide release events at nuclear power plants include the formation and transport of radionuclide aerosols. Significant research has been performed to characterize the formation, transport, agglomeration and deposition of aerosols for LWR systems [77]. The methods and techniques developed to capture this behavior will be directly applicable to non-LWR systems. However, each of the reactor systems in question will have technology specific factors that need to be addressed. Aerosol behavior will necessarily need to be captured both within the reactor system and external to it following release to the environment.

The status of the international understanding of nuclear aerosols is well summarized in the “State-of-the-Art Report on Nuclear Aerosols,” which was published in December 2009 [77]. This document primarily focuses on research that was conducted to better characterize radionuclide released for LWRs, but the methods used to capture aerosol behavior are technology agnostic, even though the specific aerosols present will be dependent on reactor type and specific accident scenarios.

Key features of aerosols that need to be well characterized so that they can be effectively modeled by a code system include: [77]

- Formation of aerosol particles
- Growth of aerosol particles
- Shape of aerosol particles
- Deposition of particles on surfaces
- Resuspension of particles from surfaces
- Removal of particles through engineered safety systems

Aerosols are formed through either mechanical processes that could occur during an accident scenario or through the nucleation of particles from supersaturated vapors, with the latter usually being the more important [77]. Typically, source term software does not model the creation of aerosols mechanistically due to the complicated nature of the physics that lead to their formation.

However, it is possible with CFD software to model such formation. This presents a potential way for advanced CFD software to provide reduced-order models to lumped parameter codes that are typically used for source term calculations.

Aerosol particles will grow by one of three methods: coagulation, condensation and hygroscopicity. Particles coagulate within nuclear systems primarily as a result of gravitation forces, Brownian diffusion, turbulent diffusion and turbulent inertia. Aerosols can also grow by condensing on an already existing aerosol particle. A third form of growth is of interest in nuclear systems: aerosol particles can also grow from water adsorption (hygroscopicity) [77].

Because aerosol particles are small, a substantial fraction of their atoms or molecules are on the particle surface, which can make them highly chemically reactive. When two particles are close, Van der Waals forces and chemical forces make it very likely they will interact and potentially adhere. When particles adhere, they will have fractal geometries; this means that the surface area and volume of aerosol particles do not have simple relations to the total mass of their components. In mechanistic models, a term known as a “shape factor” is used to account for this discrepancy. Shape factor is a parameter that may differ from LWRs to non-LWRs because of the difference in the elements and chemicals that could form aerosols [77].

As aerosols move through a system, they can become deposited on surfaces as a result of gravitational settling and diffusion, inertial deposition, phoretic deposition and turbulent deposition. After aerosols are deposited on surfaces, they can become resuspended as a result of a variety of chemical and mechanical processes. Some of these processes are technology specific; for instance, in an LWR steam generation associated with attempts to quench a degraded core would result in suspension of deposited aerosols within the primary circuit. Such phenomena would be technology and chemistry specific for non-LWRs. Another removal method addressed explicitly within modeling software is removal by engineered safety features, such as suppression pools and filtered vents. It may be necessary for explicit treatment of aerosol removal by safety components to be captured for advanced reactors for systems such as the off-gas within an HTGR, MSR, or FHR [77].

Aerosol behavior within nuclear systems is typically modeled in an integral manner or by looking at the primary circuit or containment in a stand-alone manner [77]. Representing the whole primary system, secondary system, and containment is an approach taken by integral severe accident softwares such as *MELCOR*, *ASTEC*, and *MAAP* [78, 79]. Such softwares have dedicated aerosol models such as the *MAEROS* [80] model within the *MELCOR* code. Within the reactor containment, aerosol transport may sometimes need to be treated in a more discretized manner, this has led to the widespread use of codes such as *GOTHIC* [81]. The behavior of aerosols outside of a reactor system in the atmosphere is different than within; radionuclides released to the environment must necessarily receive appropriate treatment so that transport and deposition are accurately accounted for. Software such as *RADTRAD* and *MACCS* (See Section 6) are currently used to capture such behavior for LWRs and would be useable for advanced reactors provided that technology-specific information is considered.

2.5 MELCOR

The following subsection is taken directly from the *MELCOR-ASTEC* crosswalk [78]. The contribution of IRSN authors is acknowledged.

MELCOR is a fully integrated, engineering-level computer code with the primary purpose of modeling the progression of accidents in nuclear power plants. A broad spectrum of severe accident phenomena in both boiling and pressurized water reactors is treated in *MELCOR* in a unified framework, and capabilities are being developed for advanced reactors. Uses of *MELCOR* include estimation accident progressions and the determination of fission product source terms. *MELCOR* can also be used to evaluate sensitivities and uncertainties of these calculations. *MELCOR* is developed by Sandia National Laboratories under contract to the U.S. NRC [78, 79, 82-85].

The *MELCOR* code is composed of an executive driver and a number of major modules, or packages, that together model the major systems of a reactor plant and their generally coupled interactions. Reactor plant systems and their response to off-normal or accident conditions include: [82-84]

- Thermal-hydraulic response of the primary reactor coolant system, the reactor cavity, the containment, and the confinement buildings
- Core uncovering (loss of coolant), fuel heat-up, cladding oxidation, fuel degradation (loss of rod geometry), and core material melting and relocation
- Heat-up of reactor vessel lower head from relocated fuel materials and thermal and mechanical loading and failure of the vessel lower head, and transfer of core materials to the reactor vessel cavity
- Core-concrete attack and ensuing aerosol generation
- In-vessel and ex-vessel hydrogen production, transport, and combustion
- Fission product release (aerosol and vapor), transport, and deposition
- Behavior of radioactive aerosols in the reactor containment building, including scrubbing in water pools, and aerosol mechanics in the containment atmosphere such as particle agglomeration and gravitational settling
- Impact of engineered safety features on thermal-hydraulic and radionuclide behavior

The various code packages have been written using a modular structure with interfaces between them. This allows the exchange of information among them so that all phenomena are explicitly coupled at every step.

2.5.1 Representation of LWR In-vessel Phenomena in MELCOR

2.5.1.1 MELCOR Representation of Core Region and Degradation

MELCOR represents the active core and lower plenum regions in terms of both control volumes and core cells. Within each core cell, *MELCOR* models both core materials and component physics. Built-in core cell materials are UO₂, Zircaloy, stainless steel, ZrO₂, stainless steel oxide, and B₄C. Core cells are apportioned between the following components: intact core components

(fuel, clad, etc.), structural components, particulate debris, oxidic molten pool and metallic molten pool [78, 79, 83].

For components other than fuel, *MELCOR* considers an additional field termed conglomerate debris. This represents previously molten debris that has refrozen onto an intact core component. It is treated as part of the intact core component onto which it has solidified [78, 83, 85].

Particulate debris is formed upon failure of embrittled core components. *MELCOR* assumes that this occurs for fuel rods when they persist at elevated temperatures for such an extended period that creep failure would be likely or when the fuel cladding metallic layer thickness is reduced below a critical thickness by the effects of oxidation and candling. Non-fuel rod structures are assumed to collapse into a particulate debris bed when the remaining metal thickness (either Zircaloy or stainless steel) decreases below a critical thickness. As in the case of fuel cladding, the loss of metal thickness can occur due to oxidation. For both fuel and non-fuel structures, the failure of a supporting structure will result in the supported structure collapsing into a particulate debris bed [78, 79, 83].

Molten debris that solidifies on particulate debris forms part of the particulate debris bed. This has the effect of decreasing the free volume (i.e., porosity) of the solid debris bed. *MELCOR* does not allow a particulate debris bed to become completely blocked to fluid flow [78, 79, 83].

Fuel cladding rupture is responsible for early release of fission products, primarily the gap inventory. It also accelerates cladding oxidation by exposing internal cladding surfaces to steam and results in the relocation of molten U-Zr-O outside of the fuel clad where it subsequently candles down the fuel rod. The oxidation of cladding internal structure is not currently accounted for within *MELCOR* [78, 79, 83].

Within *MELCOR*, the presence of an oxide scale can prevent relocation of molten U-Zr-O outside of the cladding. Molten material is assumed to be held up by an oxide scale when the thickness of the oxide scale is greater than a critical value, and when the component temperature is less than a critical value, typically above the Zr melting temperature. When either of these conditions are met, the oxide scale ruptures and the molten material inside is exposed and can candle down the fuel assembly [78, 79, 83].

MELCOR assumes that molten material relocates downward through a candling process until fuel component failure. The term candling refers to the downward flow of molten core materials and the subsequent refreezing of these materials as they transfer latent heat to cooler structures below. After fuel canisters failure, a significant amount of non-molten debris is generated. Both fuel debris and molten material will begin to redistribute radially based on a gravitational leveling algorithm. This becomes especially important in the distribution of material on the lower core plate and in the lower plenum [78, 79, 83].

MELCOR represents the interaction and degradation of core materials at high temperatures in one of two ways. The first is with the eutectics model, which represents eutectic interactions of key material pairs in reactor cores. It captures not only the early formation of molten material due to eutectic interactions, but more faithfully captures the evolution of stored energy in core debris beyond the point of initial eutectic melting. The second is the older interactive model, which has

been used in a number of past studies to identify the impact on core damage progression from the early formation of molten material due to eutectic interactions between core materials [78, 79, 83].

2.5.1.2 Eutectics Model

The eutectics model has become the default model used within *MELCOR* to capture the interactions between key material pairs as core structures experience temperature excursions under beyond design basis accident conditions. It captures the resulting early formation of molten material due to the interaction between dissimilar core materials, the formation of molten material at temperatures below those of the bare melting temperature for an isolated solid material. Since TMI-2 and the work of Hofmann [86], the interaction between dissimilar core materials has been recognized as a fundamental mechanism by which liquefaction of core structures occurs during LWR core damage accidents. The interaction between dissimilar materials in an overheated reactor core can occur due to a) two solid structures coming into mechanical contact at or above their eutectic interaction temperature, or b) the dissolution of an intact solid structure through contact with a liquid mixture [83].

Eutectic Reactions

Three eutectic reactions that can lead to the early failure of structural components, including the degradation of fuel rods and control blades, are currently considered:

- Early failure of fuel rods due to the reaction between Zircaloy cladding and Inconel grid spacers
- Early failure of PWR control rods due to the reaction between Zircaloy guide tubes and stainless steel clad of PWR control rods
- Early failure of BWR control blades due to the reaction between the B₄C powder and stainless-steel cladding of BWR control blades

The temperatures at which these material interactions are assumed to initiate in the *MELCOR* model are provided in Table 2-2 [83].

Composition-Dependent Mixture Properties

The eutectics model currently has six separate pseudo-binary material combinations that are used as approximations for more complicated, multi-component systems that would develop during an actual severe accident. These combinations are shown in Table 2-2 [83].

Table 2-2: Binary eutectic material pairs modeled within the core region of MELCOR [83]

Material Pairs		Molar Ratio	Eutectic Temperature
Zr	Inconel	0.76 / 0.24	1210
Zr	steel	0.76 / 0.24	1210
ZrO ₂	UO ₂	0.50 / 0.50	2800
Zr	B ₄ C	0.43 / 0.57	1900
Steel	B ₄ C	0.69 / 0.31	1420
Zr	Ag-In-Cd	0.67 / 0.33	1470

The eutectics model calculates the solidus temperature of the mixture as the mole-weighted combination of every binary combination of material pairs in the mixture. For the materials in Table 2-2, the solidus temperature of mixtures is treated as the mole-weighted average of eutectic temperature and solidus temperature of constituent in excess of the eutectic composition. The difference between the liquidus and solidus enthalpies in the material mixture is used as the latent heat of fusion of the mixture [83].

MELCOR does not perform a Gibbs free energy minimization to evaluate the multi-component material mixture state. In the simplified approach currently implemented in *MELCOR*, a mixture formed from eutectic interaction of two materials is assumed to have properties that are the mass-weighted averages of the constituent properties. For temperatures below the calculated solidus, the mass weighted enthalpies are summed with the exception that extrapolated solid enthalpies are used for any material that would ordinarily be liquid. For temperatures greater than the liquidus, mass-weighted enthalpies are summed, with the exception that extrapolated liquid enthalpies are used for any materials that would ordinarily be solid. Figure 2-4 shows how a liquidus/solidus line is constructed in this manner [83].

Figure 2-4: Two-phase construction of materials mixtures within the MELCOR eutectics model [83]

Chemical Dissolution of Solids

Within a core cell, if the enthalpy of a liquid mixture exceeds its liquidus enthalpy then certain solids within the core cell will begin to be dissolved. The dissolution proceeds sequentially, and at most two solid structures will be dissolved simultaneously. The order in which the dissolution proceeds is based on Table 2-3; if a molten mixture of high enthalpy were present in a “Canister” component then dissolution would first occur for ZrO_2 from the intact canister, followed by ZrO_2 from intact cladding and finally of UO_2 in intact fuel in the core region. The rate at which the dissolution occurs is limited by mixture enthalpy limits and parabolic rate limitations, which are based on experimental values. The mass of dissolved material is first calculated with the parabolic rate relation and then the predicted mass is then used to calculate the enthalpy of the mixture,

ensuring conservation of energy. If the mixture enthalpy is too high, then the mixture enthalpy is set to be equal to the liquidus of the new mixture composition [83].

Table 2-3: Solid dissolution hierarchy within MELCOR [83]

Component	Solids Dissolved by Mixture
Cladding	UO ₂ from intact fuel
	ZrO ₂ from intact cladding
Canister	ZrO ₂ from intact canister
	ZrO ₂ from intact cladding (A)
	UO ₂ from intact fuel
Other structure SS or NS (steel only)	steel oxide from the same other structure
Other structure NS (BWR control rod)	steel oxide from the same other structure
	ZrO ₂ from intact canister (A)
	Zr from intact canister (A)
Other structure NS (PWR control rod)	steel oxide from the same other structure (B)
	Zr from the same other structure
	ZrO ₂ from intact cladding (A)
	UO ₂ from intact fuel (A)
Particulate debris	UO ₂ from particulate debris
	ZrO ₂ from particulate debris
	ZrO ₂ from intact cladding
	UO ₂ from intact fuel
(A)	indicates solid is attacked only if there is no holdup of the mixture in the component.
(B)	indicates solid is attacked only if the mixture is being held up by the component

2.5.1.3 MELCOR Interactive Materials Model

In a number of earlier studies, including those of the Fukushima Daiichi accident, the *MELCOR* interactive materials model was used. This model was implemented to specifically capture the depressed melting temperature for B₄C, UO₂ and ZrO₂ caused by the interactions that occur between those materials and other core materials. These take place within intact fuel pins, control blades and damaged core representations. MELCOR implements three unique material classes ZrO₂-INT, UO₂-INT, and B₄C-INT, that have the same physical characteristics as pure ZrO₂, UO₂ and B₄C. For these materials, users are able to modify the liquefaction temperatures based on physical approximations and experimental evidence. This approximates the early onset of melting that would occur due to material interactions during the core degradation process. These modifications are global and not local [83].

2.5.2 Representation of LWR Lower Plenum In-vessel Phenomena in MELCOR

Within the lower plenum, particulate debris and molten pool layers are modeled similarly to how they are modeled within the core region, with separate oxidic and molten pools and a field for particulate debris. Within the formed molten pools, there is natural circulation. Heat is also transferred from formed molten pools to particulate debris and the lower head. A representation of material treatment within the lower plenum of *MELCOR* can be seen in Figure 2-5. However, *MELCOR* generally does not predict molten material formation in the lower plenum for BWR calculations [78, 83].

Figure 2-5: Representation of lower plenum characterization in MELCOR [83]

In *MELCOR*, the lower head wall can be divided into nodes or segments along the surface of the lower head wall, with nodal locations specified by the user. This enables users to capture the non-uniform distribution of heat flux from any lower plenum debris bed along the extent of the lower head wall. For each of these lower head segments, a further subdivision of the structure is performed to capture the through-wall variation of wall temperature. The through-wall subdivision extends from the internal RPV (reactor pressure vessel)-facing surface to the external PCV (primary containment vessel)-facing surface of each lower head wall segment. The number of through-wall sub-divisions can be selected by the user. The manner in which *MELCOR* discretizes the lower head wall is represented in Figure 2-6 [78, 83].

Figure 2-6: Characterization of bottom vessel head modeling in MELCOR, showing different radial nodes and discretization within the shell [83]

In this manner, *MELCOR* enables users to capture the distribution of temperature in the lower head wall, critical to evaluating lower head thermal-mechanical stress distributions. In addition to thermal stresses in the lower head wall arising from the thermal loads imposed by lower plenum debris beds, *MELCOR* represents the mechanical load from the RPV internal pressure as well as structures and mass supported by the lower head. *MELCOR* uses a Larson-Miller approach to capture creep of the lower head wall under the thermal-mechanical loads at this phase of the accident [78, 83].

2.5.3 Representation of LWR Ex-vessel Phenomena in MELCOR

The *Cavity (CAV)* package in *MELCOR* models the attack on the basemat concrete by hot, often molten, core materials. The effects of heat transfer, concrete ablation, cavity shape change, gas generation, and debris/gas chemistry are included. The package consists of models taken from the *CORCON-Mod3* code [82] together with all necessary interfaces to the *MELCOR* database and to other packages in *MELCOR*. Beyond the *CORCON-Mod3* package, SNL has recently implemented a treatment of the *CORQUENCH* and *MELTS SPREAD* models. These models take into account more complex physics that occurs during MCCI (Molten Core Concrete Interaction), such as water ingress [78, 83].

Boundary conditions for temperature and pressure used by the cavity models are obtained from an associated control volume. The treatment of any overlying coolant (water) pool is represented using *MELCOR* models distinct from the *CAV* package. Overlying coolant is applied within the *CAV* package model as a boundary condition. Heat and evolved gases are delivered as sources to the associated cavity control volume, with thermal hydraulic response treated by the *Control Volume Hydrodynamics (CVH)* package [78, 83].

When debris is deposited into the cavity from the lower plenum, the default is for a no spreading calculation to be performed; the corium is assumed to instantaneously conform to the maximum area permitted by the cavity geometry. However, the user can specify a different spreading treatment through the use of a user-defined function or make use of the *MELTS SPREAD* model contained within *MELCOR* [78, 83].

The release of fission products and the generation of aerosols from debris in the cavity is calculated using the *VANESA* model which was integrated into *CORCON-Mod3* and subsequently *MELCOR*. Within the cavity, corium is divided into five possible types of debris layers; each has a conventional three-letter designation in the associated documentation. In order of increasing density, they are: [78, 83]

- Pure oxide, less dense than the metallic phase;
- Mixed phases, less dense than the metallic phase;
- Pure metal;
- Mixed phases, denser than the metallic phase; and
- Pure oxide, denser than the metallic phase.

A user can model the debris in the debris pool as either stratified, generally into the above debris layers, or elect to enforce layer mixing.

The chemistry considered in the *CAV* package of *MELCOR* involves interactions between concrete decomposition products and metallic species in the debris pool. Equilibrium chemistry is assumed, without consideration of rate limiting effects. The calculational method is very general and is based on minimization of the total Gibbs function for a metallic phase, a gaseous phase, and an oxidic phase. Each of the three phases is treated as an ideal solution; that is, the entropy of mixing is considered, but any heat-of-solution effects are ignored [78, 83].

Two separate reactions are considered. The first involves reactions in the interior of the debris. For a pure metal layer, it is modeled as mutual equilibrium among the metal layer with the gas bubbles and concrete decomposition oxides passing through it. For a mixed-phase layer, the oxidic constituents of the layer are included as reactants. The primary effect is the oxidation of metals by the H_2O and CO_2 in the bubbles. However, if the metallic phase contains significant amounts of Zr, it can also reduce the concrete oxides to produce metallic Al, Ca, and Si. The second reaction involves mutual equilibrium among the metal layer, the gas film at its radial boundary, and the products of metal oxidation. Concrete decomposition (and other) oxides are not included in this reaction [78, 83].

2.5.4 Representation of Fission Product Release Modeling in MELCOR

The *MELCOR* code is intended to represent integral plant response for a broad range of beyond design basis accidents in traditional LWRs and a broad range of advanced reactor technologies. It has traditionally been applied to inform prescriptive radionuclide source term assessments and probabilistic risk assessments (PRAs). In order to assess the radiological source term for a range of beyond design basis accident scenarios, it is necessary for *MELCOR* to model the behavior of radionuclides as they are released from damaged fuel, transported through the plant, and ultimately released into the environment. *MELCOR* does not track all fission product isotopes, but instead tracks the behavior of chemically similar elemental groups and chemical species. The initial radionuclide inventory is calculated based on whole core neutronic calculations, using software such as the *ORIGEN* code. The release from fuel is controlled by a *CORSOR* or *CORSOR-Booth* model for each radionuclide class (Table 2-4), dependent on temperature and vapor pressure of the species. Under ex-vessel conditions, after corium has entered the containment, release from corium is controlled by the *VANESA* model [83].

Table 2-4: Radionuclide classification groups used in MELCOR [82]

Class Name	Representative	Member Elements
1. Noble Gases	Xe	He, Ne, Ar, Kr, Xe, Rn, H, N
2. Alkali Metals	Cs	Li, Na, K, Rb, Cs, Fr, Cu
3. Alkaline Earths	Ba	Be, Mg, Ca, Sr, Ba, Ra, Es, Fm
4. Halogens	I	F, Cl, Br, I, At
5. Chalcogens	Te	O, S, Se, Te, Po
6. Platinoids	Ru	Ru, Rh, Pd, Re, Os, Ir, Pt, Au, Ni
7. Early Transition Elements	Mo	V, Cr, Fe, Co, Mn, Nb, Mo, Tc, Ta, W
8. Tetravalent	Ce	Ti, Zr, Hf, Ce, Th, Pa, Np, Pu, C
9. Trivalent	La	Al, Sc, Y, La, Ac, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Am, Cm, Bk, Cf
10. Uranium	U	U
11. More Volatile Main Group	Cd	Cd, Hg, Zn, As, Sb, Pb, Tl, Bi
12. Less Volatile Main Group	Ag	Ga, Ge, In, Sn, Ag
13. Boron	B	B, Si, P
14. Water	H_2O	H_2O
15. Concrete	- -	- -
16. Cesium Iodide	CsI	Classes 2 and 4
17. Cesium Molybdate	CsM	Classed 2 and 7

After being released from the fuel, radionuclides are treated through a variety of models depending on their assumed physical form. Specific treatment of aerosol dynamics, condensation and evaporation, pool scrubbing, filters, sprays and chemisorption is included among other models. At the point radionuclides are released to the environment, the physical characteristics of each radionuclide class is passed to an atmospheric transport and deposition code or consequence tool. The chemistry within the code is informed by results of numerous experiments, against which the radionuclide models are validated. These include the *LOFT* and *PHEBUS* programs among others [83].

2.6 NRC Modeling and Simulation Development Pathway

Relevant to the current effort, the NRC has recently issued their vision and strategy for the development of computer code capabilities for the assessment non-LWR licensing applications. The development pathway is planned to be published in five volumes, with the first three volumes now available. The first volume focuses on computer code capabilities for the analysis of DBEs¹ [87]. As shown in Figure 2-7, this centers on the formation of the Comprehensive Reactor Analysis Bundle (CRAB), which is a code suite composed of commercial, NRC, and DOE codes. The current and planned capabilities of these codes in relation to non-LWR modeling will be discussed in detail in later sections of the report. The second volume of the development pathway describes the utilization of the *FAST* and *BISON* codes for fuel performance analyses [88].

Figure 2-7: Comprehensive Reactor Analysis Bundle (CRAB) Proposed by the NRC for the Analysis of Design Basis Events in Non-LWRs [87]

¹ See Figure 1-4 regarding the location of DBEs in frequency-space.

Of particular interest to the current project is the third volume of the NRC code development pathway, which describes the proposed codes for severe accident progression, source term, and consequence analyses [58]. Included in this document is a description of the interfaces and responsibilities between the NRC offices, with an overview provided in Figure 2-8. The Office of Nuclear Regulatory Research (RES) is responsible for code development, which would then be utilized by the Office of Nuclear Material Safety and Safeguards (NMSS) for storage and transport analyses and the Office of Nuclear Reactor Regulatory (NRR) for reactor licensing and siting evaluations. The NRC Vision and Strategy document also outlines the computer codes proposed for the evaluation of accident progression, source term, and consequence analysis for each advanced reactor type, which will be reviewed here in the appropriate subsections. In addition, development pathways for codes such as *MELCOR* are also provided in the NRC document.

Figure 2-8: Role of Accident Progression, Source Term, and Consequence Analysis Computer Codes and Applicable Regulatory Requirements [58]

As part of the NRC's efforts to support the safety analysis of advanced reactors, they are funding the development of generic plant models. This will enable universities, labs, and other experts to analyze the safety behavior of advanced systems without needing to develop decks and models from scratch. This includes the development of advanced reactor plant decks for the *MELCOR* program.

Volume 4, which has yet to be published, will focus on codes utilized for licensing and siting dose calculations. This mainly includes codes under the NRC Radiation Protection Computer Code Analysis and Maintenance Program (RAMP). As shown in Figure 2-9, the RAMP program includes many codes utilized for radiological and chemical hazards across the spectrum of potential events. In general, the RAMP program is pursuing a consolidation effort, which is seeking to reduce to the total number of codes by centralizing capabilities within select tools.

Figure 2-9: Overview of NRC Licensing and Siting Dose Code Landscape [89]

Some of the RAMP codes will be discussed individually in the corresponding sections of this report. However, the *RADionuclide, Transport, Removal, and Dose Estimation (RADTRAD)* code is briefly described here, as its use for source term analyses has been pursued recently by several advanced reactor vendors. *RADTRAD* was developed by the NRC for the analysis of radionuclide release to the exclusion area boundary (EAB) during design basis accidents [90]. The current version of *RADTRAD* (RADTRAD-AC version 5.0) has been coupled with the *Symbolic Nuclear Analysis Package (SNAP)* to provide a graphical user interface (GUI) and pre- and post-processing capabilities and is generally referred to as *SNAP/RADTRAD*². The main purpose of *SNAP/RADTRAD* is to show compliance with nuclear siting and control room dose limits. The code contains general models for the release of radionuclides from fuel based on TID-14844 and NUREG-1465, although custom release fractions can be used. In addition, radionuclide transport and retention are simulated utilizing a compartment model, with simplified aerosol deposition and spray models available. *RADTRAD* performs dose assessments based on user-specified values.

² Versions of *RADTRAD* are also maintained by industry, such as *RADTRAD-NAI* by Zachry Nuclear Engineering [91].

3 High Temperature Gas Reactors

Over the past six decades, seven experimental and commercial scale high-temperature gas reactors (HTGRs) have been built and operated in the United Kingdom, United States, Germany, Japan, and China [92]. A significant amount of work has been done to date to characterize the mechanistic source terms of HTGR, including recent work as part of the Next Generation Nuclear Plant (NGNP) project. Of note is the publication of a mechanistic source term white paper by INL [93], several characterizations of the functional containment concept for HTGR systems [94, 95], significant experimental efforts of fission product release, and the development of both system-level software and code suites for source term analysis.

HTGR systems use a gas as a coolant, typically helium under elevated pressures. This review examines the primary system of a generic HTGR that uses helium as a coolant and graphite as a moderator. The fuel is a compact of spherical tri-structural isotropic (TRISO) fuel either in a pebble or prismatic geometry [3].

TRISO fuel is a multilayer fuel form that contains a fuel kernel (typically uranium oxycarbide (UCO) but other fuels have been used, such as UO_2) surrounded by a porous carbon buffer, silicon carbide, and two pyrolytic carbon layers. TRISO fuel can then be compacted into any designed shape, with the most common two fuel matrix designs being pebbles and prismatic fuel elements. A diagram of TRISO fuel can be seen in Figure 3-1, where an example of fuel particles being used in a pebble compact form is provided [93]. It should be noted that some designs may even move or reconfigure pebble locations during operation. A prismatic fuel matrix design will use cylindrical compacts filled with the TRISO fuel particles that are organized into a solid lattice.

Figure 3-1: TRISO fuel layers and final compact form as a pebble [93]

Licensing Basis Events

The selection of potential licensing basis events (LBEs) has been performed for many HTGR designs [96]. One of the unique characteristics of many HTGR LBEs, and therefore also a challenge for associated MST modeling, is the relatively long duration of some identified event sequences, which could exceed 10 days. The difficulty in modeling long event sequences is the selection of appropriate time scales in the models, especially when considering that the time scales for different phenomenon can vary by several orders of magnitude. The primary LBE of interest for offsite consequence, a depressurized loss of forced cooling event, is an example of this complexity. In the first phase of the accident, there is an initial primary system blowdown, followed by a long core heat-up. The blowdown and subsequent events related to the blowdown occur relatively quick, on the order of minutes, whereas the core heat-up occurs over the course of days.

As will be further discussed in this section, many of the additional MST modeling challenges are the result of their dependency on multiple factors, as is the case for assessing mass-transport modeling through TRISO particles/pebbles. Parameters such as natural convection flow and temperature-dependent diffusivity coefficients for radionuclide classes may require significant experimental validation. Other LBEs of interest, such as events that include air and water ingress into the primary system, require complex radiochemistry modeling.

Several of the phenomena important to MST can be modeled with existing capabilities such as: in-service TRISO failures; distribution, settling, condensation in the primary system and reactor building; and the functional containment approach. The functional containment approach is considered a key safety design characteristic of the HTGRs.

3.1 System Modeling Capabilities

A set of tools within the CRAB framework is one method that has been identified for use in capturing the behavior of the reactor system during a DBE. These include:

- *BISON* for fuel performance and fission product diffusion within TRISO fuel
- *MAMMOTH* for neutronics of the reactor core
- *PRONGHORN* for primary system modeling
- *TRACE* for characterization of the RCCS (reactor cavity cooling system)
- *SERPENT* for cross-section generation

The interfacing of these software can be seen in Figure 3-2. Currently the largest gaps identified for the CRAB suite for HTGR modeling is the perceived maturity level of solution verification, model validation and uncertainty quantification being low [87].

Figure 3-2: Modeling Approach for Design Basis Event Simulation in an HTGR [87]

3.2 Source Term Modeling Capabilities

An overview of HTGR radionuclide transport and retention pathways is provided in Figure 3-3, with the following subsections detailing the main phenomena of interest. In general, two separate approaches have been examined to capture the behavior of fission products within HTGR systems. As part of the NGNP, DOE and industry partners developed two separate sets of software suites (one for prismatic and one for pebble beds) to model the release of fission products from the fuel, their transport through the primary circuit, and their behavior within reactor containment [93]. The NRC has taken an approach of coupling the *SCALE* neutronics package, for initial inventory development, with the *MELCOR* system level code for fuel releases, transport through the primary circuit, and release to the environment [58]. It is also worth noting that X-Energy, a domestic developer of an HTGR concept, has indicated that they will likely utilize their proprietary software *XS-Term* for a portion of their source term analysis [12, 97].

Figure 3-3: Summary Diagram for HTGR Radionuclide Retention Phenomena [93]

The “Mechanistic Source Terms White Paper” [93], authored by INL as part of the NGNP, details the code suites employed by General Atomics for a prismatic core design and the code suites employed by PBMR Ltd. for pebble bed HTGRs. A description of these codes is provided within the white paper in Appendix D and Appendix E, respectively. Thus, descriptions are not provided in this document. Additionally, these exact software suites are no longer the state-of-practice and are not expected to be employed by either licensees or the NRC for source term modeling. Nonetheless, Figure 3-4 provides a high-level overview of the *PBMR* software suite for reference.

Figure 3-4: PBMR fission product release software analyses suite [93]

In comparison to LWRs, a different set of radionuclides must be considered for HTGR source term modeling. As shown in Table 3-1, analysis performed as part of the NGNP effort identified characteristic radionuclides, where radionuclides assumed to have the same release and attenuation factors based on physical and chemical properties were organized into groups or classes [94]. These radionuclides generally align well with the established fission product classes from source term modeling software. Of particular note is the importance of silver within the HTGR source term. Silver is not considered a radionuclide of significant importance for LWR source term characterization.

Table 3-1 Fission Product Classes of Similar Properties with Characteristic Nuclides [94]

Fission Product Class	Characteristic Nuclides
Noble Gases	Kr-85, Kr-88, Xe-133
I, Br, Te, Se	I-131, I-133, Te-132
Cs, Rb	Cs-134, Cs-137
Sr, Ba, Eu	Sr-90
Ag, Pd	Ag-110m, Ag-111
Sb	Sb-125
Mo, Ru, Rh, Tc	Ru-103
La, Ce	La-140, Ce-144
Pu, actinides	Pu-239

MELCOR

As shown in Figure 3-5, the NRC has proposed the use of *MELCOR* for LBE transient analysis for HTGRs. As part of the NGNP Program, significant effort was taken to update the *MELCOR* software package to include HTGR specific models. The NGNP effort lasted from 2005 to 2013, during which time the NRC conducted pre-licensing interactions with reactor designers. To ensure the NRC would be able to perform source term and consequence analysis when necessary, significant resources were directed to add new models to the *MELCOR* framework at SNL. Resources were also directed to universities to develop scientific models and plant decks that could be used for analysis of publicly available designs. Following a hiatus of a few years, development of *MELCOR* is once again being performed to adequately capture all HTGR relevant phenomena. Models within *MELCOR* can be validated by experiment or benchmarked against other software such as those within the NEAMS suite [83, 98, 99].

Specific models and phenomena that have been implemented within *MELCOR* that are relevant to HTGR modeling include: [83]

- Helium coolant gas properties
- Graphite oxidation models
 - Graphite in steam model
 - Graphite in air model
 - Multipliers for the effects of catalysis and pore diffusion
- Heat transfer and thermodynamic models for pebble beds and prismatic compacts
 - Zehner-Schlunder-Bauer formulation of pebble bed radial core heat conduction
 - Tanaka-Chisaka formulation for prismatic fuel radial heat conduction
 - Addition of pebble bed and prismatic specific treatments of fuel to clad heat transfer
- Fission product release and transport models
 - Fission product distribution within TRISO particles
 - Fission product release from TRISO particles
 - Fission product release from pebbles and prismatic compacts
 - Ability to account for initial TRISO particle defects
 - TRISO failure model based on temperature criteria

- Graphite dust transport models
 - Defined by used and tracked as an aerosol
 - Turbulent deposition and resuspension models
- Point kinetics model
 - Six delayed neutron precursor group model
 - Allows for zero-power neutron source
 - Reactivity feedback effects of fuel and graphite.

Figure 3-5: Proposed NRC Evaluation Model for HTGRs [58]

Recent gap analysis by the NRC and SNL have indicated that there is an existing gap in the modeling of the TRISO fuel, which can be seen in Table 3-2 with other key accident progression phenomena [58]. Currently TRISO fuel is modeled with UO_2 fuel properties within *MELCOR*, and this will necessarily need to be updated to include UCO properties. Additionally, more extensive code verification, validation, and uncertainty quantification will need to be performed to ensure that the models have the appropriate level of fidelity. These efforts are planned for FY21 and FY22. An extensive test matrix has been prepared for the software to ensure that these needs are met [58].

Table 3-2: Key Accident Progression Phenomena for HTGRs [58]

Key Phenomenon	Importance	Existing Capabilities	Modeling Gaps
Modeling of TRISO fuels	Determining release of fission products from fuel and fuel material properties	<ul style="list-style-type: none"> Analytic release model Multi-zone diffusion model Account for FP recoil, matrix contamination, and initial TRISO defects 	<ul style="list-style-type: none"> Current modeling uses UO₂ material properties, needs to be extended to UCO (Development Items M2.1 and M2.2)
Heat Transfer in Graphite block (PMR)	Thermal response of fuel components and failure of TRISO fuel particles	<ul style="list-style-type: none"> Tanaka-Chisaka effective radial conductivity 	
Heat Transfer in fuel pebbles (PBR)	Thermal response of fuel components and failure of TRISO fuel particles	<ul style="list-style-type: none"> Zehner-Schlunder-Bauer effective thermal conduction 	
Reactivity temperature feedback coefficients.	Neutronics power feedback	<ul style="list-style-type: none"> Point kinetics model Reactivity coefficients specific to an application can be implemented via control functions 	
Ability to model two-sided reflector component	Heat transfer from overheated core	<ul style="list-style-type: none"> Two-sided reflector component 	
Modeling graphite dust transport	Pathway for fission product transport and release	<ul style="list-style-type: none"> All relevant mechanisms for graphite dust transport, deposition, and resuspension 	
Graphite oxidation	Heat generation and release of combustible gases	<ul style="list-style-type: none"> Graphite oxidation model and oxidation products 	
Air/moisture Ingress modeling	Air/moisture ingress can lead to oxidation of the graphite structures and release of radionuclides	<ul style="list-style-type: none"> Momentum exchange model 	

MELCOR has been assessed for use in HTGR accident scenarios by multiple nuclear energy research organizations, including SNL [83, 100], Texas A&M University [101, 102], the Korean Institute of Nuclear Safety [103] and the Paul Scherrer Institute [104]. These organizations have performed analyses for a wide array of accident scenarios, including pressurized loss of forced cooling (PLOFC), depressurized loss of forced cooling (DLOFC), pressurized conduction cooldown (PCC), and depressurized conduction cooldown (DCC). Core temperatures as modeled by MELCOR during a DLOFC scenario can be seen in Figure 3-6. along with the nodalization used (Figure 3-7) for a 300 hr transient.

Figure 3-6: MELCOR DLOFC transient simulation of axial core temperature for ring 2 [58]

Figure 3-7: PBR MELCOR model with simplified primary and secondary loops [58]

3.2.1 Radionuclide Inventory

The development of a radionuclide inventory is important for developing accurate models for the source term. This subsection looks at the various models that have been proposed for modeling the depletion of the radionuclide inventory. First an example simulation is shown and explained to give an understanding how these processes work, followed by a study that compared the results of eight organizations for various HTGR set ups.

HTGR core neutronics modeling in *SCALE* is robust, as evidenced by the recently characterized maturity levels seen in Table 3-3. *SCALE* has been used to investigate three approaches to modeling TRISO fuel particles: a uniform array of particles, a random-mesh, and random location of particles. The uniform array of particles is the fastest approach, but can introduce user bias, which can lead to problems in the fuel to moderator ratio. The random mesh approach is between uniform and random, where fuel particles are randomly distributed throughout the fuel and if the fuel particles interact with the mesh, they are resampled. Random is where the fuel particles are randomly dispersed throughout the fuel region [105].

Table 3-3: SCALE Maturity for HTGR Analysis [58]

Element	Maturity Level ¹	Comments
Representation and Geometric Fidelity	3	<ul style="list-style-type: none"> Under NGNP effort a double-heterogeneous treatment has been developed to support the two levels of heterogeneity (i.e. TRISO micro-spheres dispersed in the graphite matrix, and the rod-to-rod or sphere-to-sphere neutron interactions). SCALE has been used to model various aspects of HTGRs Reviewed by ACRS as part of NGNP
Physics and Model Fidelity	2	<ul style="list-style-type: none"> Need for more complete test data to support depletion validation. SCALE has been used to model various aspects of HTGRs Reviewed by ACRS as part of NGNP
Code Verification	2	<ul style="list-style-type: none"> Extensive SQE, many capabilities have been benchmarked and some peer review.
Solution Verification	2	<ul style="list-style-type: none"> Some informal assessments both internally as well as assessment by code users.
Model Validation	2	<ul style="list-style-type: none"> Extensive validation of most physics models though not all within the domain of HTGRs. External assessment
Uncertainty Quantification and Sensitivity Analysis	2	<ul style="list-style-type: none"> Uncertainties and numerical propagation of errors has been examined extensively for LWR applications though not for HTGR application

¹Maturity Levels

- level 0, little or no assessment of accuracy and completeness and highly reliant on personal judgment and experience;
- level 1, some informal assessment of accuracy and completeness, and some assessment has been made by an internal peer review group;
- level 2, some formal assessment of accuracy and completeness, and some assessments have been made by an external peer review group; and
- level 3, formal assessment of accuracy and completeness, and essentially all assessments have been made by an independent, external peer review group.

The *SCALE* model of an HTGR was benchmarked with the HTR-10, the Chinese test PBR. Depletion calculations are calculated using the *TRITON* sequence within *SCALE*. The *TRITON* sequence performs the cross-section processing, neutron transport, and depletion/decay by calling the adequate modules as applicable. The depletion and decay of isotopes is shown in Figure 3-8 for the ORNL study on various neutronic codes for modeling depletion in HTGRs. The results showed that the results for the MG and CE are very similar, with all actinides shown having less than 3% difference between the two methods [105].

A study by ORNL looked at a code-to-code benchmark for HTGR fuel element depletion [106]. This study presented 21 research efforts that were submitted by eight organizations, both domestic and international. The approaches varied for the treatment of the fuel elements and they used various codes to model the reactor type. This study compared three fuel types: infinite lattice of grains, generic pebble bed configuration, and a third based on the characteristics of the MHTGR prismatic fuel element [106]. Table 3-4 lists the various organizations and codes that have been used to model HTGR designs. The study compared many aspects of these different models, from calculating the multiplication factor to the depletion of isotopes. The depletion calculations were modeled out to 120 GWd/t [106].

Figure 3-8: HTGR pebble depletion as a function of burnup. The results for the various studies analyzed by ORNL [106]

The depletion calculations showed good agreement for the various isotopes of interest shown in the study. This study showed the following isotope inventories: U-235, U-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, Cm-244, and Cm-245 for the three different fuel types. For the three types of fuel, all of the organizations saw the same trends for the isotopes [106]. Along with actinide isotopes, specific fission products were also of interest.

The fission products of interest included: Kr-85, Sr-90, Ag-110m, Cs-137, Xe-135, and Sm-149. These plots showed higher variance between the various codes used. The codes showed the same general trends and reasonable agreement, though one code system was an outlier that skewed the average values away from most of the results. The pebble bed design and prismatic design showed higher agreements, although the study showed that pebble bed modeling is more advanced than the neutronic modeling of prismatic fuel [106].

Table 3-4: The ORNL study organized work with the following organizations and codes [106].

Name of Study	Type Modeled	Organization	Code Used
INL-Studsvik	Prismatic	INL/Studsvik Scandpower	<i>HELIOS</i> Cross-Section Library based on ENDF/B-VII.0
FZD 1	Grain, Prismatic	FZD	<i>BGCore</i> (MCNP + MG depletion)
FZD 2	Prismatic	FZD	<i>BGCore 3D</i> (MCNP + MG depletion)
FZD 3	Grain, Prismatic	FZD	<i>HELIOS 1.9</i>
IKE 1	Grain, Pebble, Prismatic	IKE	<i>MCNP</i> coupled with module <i>Abbrand</i> (simplified burn-up model with 86 fission products and 20 actinides)
IKE 2	Grain, Pebble, Prismatic	IKE	<i>Microx2.2</i> coupled with <i>Origen2.2</i>
IKE 3	Grain, Pebble, Prismatic	IKE	Grain: <i>SCALE 6</i> , <i>TRITON</i> t6-depl sequence (<i>KENO-VI</i>) Pebble and Prismatic: <i>SCALE 6</i> , <i>TRITON</i> t5- depl sequence (<i>KENO V.a</i>)
GRS	Grain, Pebble, Prismatic	GRS	<i>MONTEBURNS 2.0</i> (<i>MCNP5</i> + <i>ORIGEN2.2</i>)
LLNL	Grain, Pebble, Prismatic	LLNL	<i>MOCUP</i> , <i>MCNP5</i> Version 1.40 + <i>ORIGEN2.2</i>
VTT1	Pebble, Prismatic	VTT Technical Research Centre of Finland	<i>Serpent</i> Monte Carlo reactor physics burn-up calculation code, version 1.1.2
VTT 2	Grain, Pebble, Prismatic	VTT Technical Research Centre of Finland	<i>Serpent</i> Monte Carlo reactor physics burn-up calculation code, version 1.1.2
ORNL 1	Pebble, Prismatic	ORNL	<i>Scale 6.1</i> , <i>TRITON</i> t-depl sequence
ORNL 2	Grain, Pebble, Prismatic	ORNL	<i>Scale 6.1</i> , <i>TRITON</i> t-depl sequence (<i>KENO</i> <i>V.a</i>)
ORNL 3	Pebble	ORNL	<i>Scale 6.1</i> , <i>TRITON</i> t-depl sequence (<i>XSDRN</i>)
ORNL 4	Pebble	ORNL	<i>Scale 6.1</i> , <i>TRITON</i> t-depl sequence (<i>XSDRN</i>)
LANL	Grain, Prismatic	LANL	<i>MCNPX2.7b</i>

3.2.2 TRISO Fuel Particle Releases

Computational tools exist and are currently being validated for their ability to model radionuclide releases from TRISO fuel particles and the fuel matrices. For example, *BISON* has been used to model Cs, Sr, and Ag releases and was found to be in good agreement, after slight parameter modification, with legacy German post-irradiation annealing data as well as more recent AGR-1 data [107]. The current state of fuel performance modeling of TRISO fuel seems to be sufficient for source term modeling purposes, and as irradiation and annealing testing of TRISO fuel continues to shed light on radionuclide release fractions, codes such as *BISON* should be correspondingly validated against such data.

Irradiation testing of coated particle fuels has been conducted in materials test reactors and used in operating gas-cooled reactors like the AVR and the Fort St. Vrain HTR. These experiments developed a list of potential mechanisms that could lead to fuel particle failure: [108]

- (1) Coating damage during fuel manufacture, particularly during the sphere pressing stage.
- (2) Pressure vessel failure during irradiation or during a temperature transient caused by the internal gas pressure of the irradiated particles, resulting in complete coating failure.
- (3) Failure of the pyrocarbon layer due to neutron-induced embrittlement.
- (4) Failure of the SiC layer due to fuel kernel migration and interaction with the coating.
- (5) Failure of the SiC layer by chemical reaction with internally generated fission products.
- (6) Thermal decomposition of the SiC at extremely high temperatures.

Item (1) is based on the quality of the fuel entering the reactor. A study comparing the differences of fabrication process between the United States and Germany linked stages in production with quality of the fresh fuel [109]. A major difference in the fabrication of TRISO fuel between the United States and Germany was on the scale of production. The German fabrication process was on the industrial scale, their production produced TRISO fuel with ~100 defects in 3.3 million particles. Whereas the United States fabrication process combined lab fabrication and larger scale productions to produce TRISO fuel. Splitting the fuel fabrication into work led by both General Atomics and Oak Ridge National Laboratory led to the facilities using different fuel and coating types; this collaboration led to more variation in the defect level that varied greatly between the two teams and was greater than that for the TRISO fuel produced by Germany [109].

Pressure-induced failure is caused by the internal pressure buildup of xenon, krypton, and CO (in the UO₂ fuel). The pressure increases with increasing gas and if the induced circumferential stress exceeds the ultimate tensile strength of this layer. The dominant 1D failure mechanism for TRISO fuel particles involves pressure vessel failure, where the SiC layer develops a through-thickness crack resulting from a tensile stress that exceeds the fracture strength of the material [110]. The fracture stress for SiC varies but is on the order of 350-400 MPa [110]. Modern approaches consider a pebble failed if all three layers are cracked. In the model, modeling all three layers, the tangential stress between the IPyC/SiC interface usually is the limiting stress [110]. Once the fuel is under flux, both PyC experience irradiation-induced radial shrinkage; following the densification, irradiation creep all acts outward on the layers. This creates a situation where the max compressive stress is -100 MPa based on fluence, and the stress increases during the lifetime of the fuel in the reactor. If the stress becomes tensile (greater than zero) it is presumed the fuel

will fail [110]. Experiments have shown by increasing the thickness of the buffer layer this failure mechanism is contained [108].

Pyrocarbon embrittlement has been eliminated by a change in the fabrication process; the fabrication change included changing the method of adding layers to the particles, switching from methane to propene [108, 111].

The migration of fuel kernels is based on the temperature gradient, and is independent of burn-up [112]. Figure 3-9 shows an example of kernel migration. The fuel kernels move until they interact and attack the coating. Fuel kernel migration increases with temperature; if the fuel temperature increases from 1100°C to 1300°C the propensity for kernel migration increases by a factor of 1.7 [112]. However, fuel kernel migration is mitigated using spherical fuel elements to generate a homogenous fuel distribution, and to have low power densities in HTGRs to decrease the presence of strong temperature gradients [105].

Relative Thermal stability of
HTGR Candidate Recycle
Oxide Fuel Kernels Irradiated
in HRB-7. Time-average
temperature, 1200-1220EC;
thermal gradient,
1000-1030EC/cm; fast
fluence, $6 \times 10^{21} \text{ n/cm}^2$.

Figure 3-9: Migration of a UO_2 kernel [113]

During studies on accident scenarios where chemical reactions occur between fission products and the SiC layers, palladium has been shown to be the key element in the degradation of the SiC layers [105]. In studies on palladium, high burnup LEU fuel produces 25-50 times more palladium compared to either high burnup HEU fuels or low burnup LEU fuels [112]. Palladium penetration in SiC shows an Arrhenius temperature dependence, therefore palladium attack on the SiC layers is a concern for HTGR designs.

SiC degrades at 2100 °C and in experiments using temperature ramps, it was shown that the SiC layer completely degrades at a temperature of 2500 °C. Although, it is expected that these high temperature will not occur in small, modular HTGR designs [105].

In 2010, a report looking at the material problems facing next generation reactors stated that there are multiple studies around the world on implementing TRISO fuel for next gen reactors [114]. As nuclear material experts continue to test TRISO fuel, the issue becomes ensuring the quality of the fuel is maintained at higher burnups and higher operating temperatures [114].

Although published results have shown that most fission products are contained within the fuel pebbles, certain fission products are able to escape through the SiC and PyC layers. The Next Generation Nuclear Plant (NGNP) Fuel Development and Qualification Programs ran a series of fuel irradiation experiments in the Advanced Test Reactor (ATR) at INL. AGR-1 ran from December 2006 to November 2009 and used deconsolidation-leach-burn-leach and individual particle gamma counting to identify the fission products released from the particles [115]. This experiment found Eu-154 and Ag-110m present in the fuel compact matrix, indicating a release through intact pebbles. In a fuel compact matrix, silver was released completely from certain pebbles and completely contained in others. Future work is needed to understand the effect of temperature gradient on silver release. There were fractional releases of Ce-144 and Sr-90 at smaller rates compared to Ag-110m and Eu-154, and 1% of the total Pd-105 inventory was found to be in the fuel compact matrix also. As an example, Figure 3-10 shows metal release during post-irradiation heating of LEU UO₂ TRISO particles at 1700°C.

Figure 3-10: Metal Release During Post-irradiation Heating of UO₂ TRISO Particles [93]

3.2.3 Circulating Activity

One concern with HTGR is the distribution of radionuclides that deposit on wetted surfaces over long periods of reactor operations. Developing realistic models for normal operations and accident scenarios is important for the design of the purification system, shielding, maintenance of the components of the coolant systems, and safety analysis [116]. Korea Atomic Energy Research Institute (KAERI) developed a computer code *POSCA* (*Plate-Out Surface and Circulating Activities*) to evaluate the fission product plate-out and circulating coolant activities within the primary circuit of a HTGR [116].

Radioactive nuclides enter the primary coolant as a result of release from wetted surfaces in the reactor core or by direct production in the coolant [116]. A fear is that during DBA-like depressurizations, stand alone or followed by a core heat-up phase, steam/water ingress accidents may lead to a partial release of fission products accumulated in the coolant circuit [117]. A plate-out model needs to take into account the deposition of material, and the various ways the radioactive nuclides are removed from the primary coolant system: nuclide decay, deposition on wetted surfaces, coolant purification, coolant leak, and nuclear reactions with neutrons [116].

The AVR has been studied to understand the impact of nuclides released from the fuel elements which are reabsorbed on surfaces or on graphitic dust [117]. Two experimental studies were performed on the AVR: VAMPYR-I and VAMPYR-II. VAMPYR-I was an experiment on the hot-gas filter. The experiment looked at the condensable nuclides' activity in the hot gas and were measured via a plate out section and the dust activity in the filter. VAMPYR-II was a plate out experiment in the hot gas region.

The AVR showed that radionuclides were reabsorbed on the fuel elements [117]. Activities of long-lived metallic fission products were seen in the graphite/carbon brick core top (approximately 10% of the total inventories of Sr-90 and Cs-137 released from active core are found on this component). Another area of concern is deposition in the steam generator. VAMPYR-I showed that most condensable fission products are deposited along the steam generator. Cs-137 and Cs-134 were found on the hot and cold paths of the steam generator. Co-60 occurs only as a dust borne product, formed by corrosion from activated metallic components. Cs, Sr, and Co were the main isotopes found in the steam generator and on fuel components in reactor experiments. However, other elements have been shown in experimental studies outside of the AVR reactor. I-131 was also seen in the HTR-like reactor DRAGON and in the COMEDIE loop.

The *POSCA* code breaks the region into three areas: bulk, thin boundary layer, and structural surface. The code solves the following equations to obtain a solution: mass conservation in the coolant bulk, mass conservation of reversible nuclides on the wall surface, and mass conservation of irreversible nuclides on the wall surface. The code was validated by simulating the VAMPYR-1 and OGL-1 experiments and the simulated results are in good agreement with the experimental data. A problem related to accurately modeling plate-out activities is the lack of experimental data, therefore the model needs further verification [116].

3.3 Capability Assessment

As detailed in the previous subsections, the current status of analysis software for the full spectrum of accident modeling, from AOO to BDBE is relatively advanced and complete. However, based on the review of previous studies and the evaluation performed as part of the current work, several key gaps were identified regarding the current capabilities of source term modeling and simulation tools for HTGRs, described below:

- **Core Neutronics and Fuel Depletion:**
 - There have been many efforts in modeling core neutronics and fuel depletion for HTGRs, however, gaps exist regarding the modeling of prismatic fuel.
- **TRISO Fuel Particle Releases**
 - In general, there is lack of a refined TRISO model (within a framework such as *MELCOR*) that is based on UCO properties and not just a UO₂ approximation.

Accurately capturing the failure rate of TRISO particles within any software suite is of high priority and may be batch- and manufacturer-dependent.

- The effect of burnup on the quality of TRISO fuel particles as well as the temperature-dependent behaviors of radionuclide releases are key to understanding the element-specific release fractions of TRISO particles.
- **Circulating Inventory**
 - Despite having a chemically inert coolant, radionuclides may still transport within the bulk flow of the primary circuit in HTGRs. Therefore, further research appears needed on both computational modeling and experimental studies of fission product plating in the primary coolant loop.
 - Due to the ease with which radionuclides can adsorb onto graphite, the transport of radionuclide-containing dust throughout the reactor, as well as the deposition or settling, may be an important source of radioactivity. Experimental studies may be needed to better characterize the phenomenon. Modeling the transport of dust with the bulk fluid can conceivably be accomplished with existing gas-phase thermal hydraulic modeling tools for HTGRs.
- **Software Quality Assurance**
 - An identified gap is software verification, validation and uncertainty quantification for HTGR-specific models. This is particularly true of the NEAMS software suite, which will necessarily have to perform more of these exercises for use in licensing.

There have been many efforts in modeling core neutronics and fuel depletion for HTGRs but there still exist gaps in modeling capabilities. This is especially true for pebble bed reactor (PBR) cores, whereas additional development of the prismatic fuel neutronic codes is likely needed. The ORNL study showed that the neutronic codes are more in agreement with each other for the PBR style reactor, with the results having larger variance for the prismatic design. Identifying the factors affecting the larger variance for the prismatic design is a gap.

Another gap related to both types of HTGRs is the effect of temperature and burnup on the quality of the TRISO fuel. Many of the fuel failure mechanisms are related to the temperature of the TRISO fuels. Modeling temperature spectrums throughout the reactor is an important step, and then having dependencies based on the failure mechanism impacts the fission products that are released. Currently the burnup behavior of TRISO particles has many gaps and progress in materials science will need to be made. Experimental studies, notably fuel irradiation testing, are required to see how fuel evolves with increasing burnup; burnup may change the amount of fission products released from the HTGR.

Once fission products are released from the TRISO particles, modeling fission product plating in the coolant circuit loop needs to be accomplished. A major issue related to the fission products absorbed on structures is the lack of experimental data. The fission products have been identified; however, properly modeling the behavior has been limited due to the lack of operational source term data to validate the models.

4 Molten Salt Reactors

Molten salt reactors (MSR) utilize a broad class of liquid salts as the coolant, with both solid fuel and dissolved fuel in the coolant salt as options. MSR designs are quite diverse, given the ability to choose many different combinations of dissolved-fuel material, coolant salt, core design, auxiliary systems, and flow parameters. One variant of molten salt-cooled reactors combines the heat transport properties of molten salts with the positive fuel performance benefits of TRISO fuel particles. First introduced as the Advanced High-Temperature Reactor (AHTR) [118, 119], design modifications over the past two decades have led to a version called the fluoride salt-cooled high temperature reactor (FHR) [120-125]. Although this solid-fueled, salt-cooled reactor does not share all of the same characteristics as salt-fueled MSRs (e.g., a circulating distribution of most of the reactor's radioactivity) some of the FHR systems and phenomena are shared with liquid-fueled MSRs. This includes a low pressure, high temperature coolant salt with chemistry similar to that of many other MSR concepts, auxiliary systems such as salt and off-gas clean-up systems, and interactions of the salt with graphite and other core materials. Therefore, source term modeling capabilities for the FHR are discussed in this section along with salt-fueled MSRs.

Due to their design diversity, it can be difficult to develop a standard source term modeling strategy applicable to all MSRs. Nonetheless, a survey of computational capabilities in modeling and simulation is described here. While a detailed mechanistic source term analysis has not yet been completed for an MSR, much work has been published regarding system modeling, transient analysis, and multiscale modeling, which is the focus of Section 4.1. Knowledge of these capabilities can assist in the development of a robust source term modeling approach, which is discussed in Section 4.2. Considerations for the FHR are provided throughout where applicable. Finally, the current gaps in MST computational capabilities are outlined in Section 4.3.

Licensing Basis Events

For salt-fueled MSRs, the major sources of radioactivity are the circulating primary salt, which includes the dissolved fuel and fission products, and the potential for tritium generation and activation products. There are also sources of radioactivity in auxiliary systems such as the fuel processing system and the off-gas system. In general, the LBEs of interest for source term assessment include a breach or release from one of these systems.

Holcomb et al. hosted an MSR initiating event (IE) and LBE workshop [126] that included MSR subject matter experts and nuclear industry experts from several technical areas. 140 IEs were identified as part of the ORNL-hosted workshop, with extended discussion on seven deemed to be critically important: [126]

- Primary fuel salt heat exchanger failures
- Primary fuel salt boundary breaches
- Primary fuel salt composition changes
- Primary fuel salt void fraction changes
- Drain tank/decay heat removal failure
- Drain tank breaches
- Off-gas system breaches and other failures

The results of the workshop are important as it includes a wide range of generic IEs that could be used by reactor designers and regulators. Frequencies and consequences were estimated qualitatively for some IE classes, which is a necessary step when employing the LMP. The results also provide critical discussion notes and commentary that have impacts on source term modeling. The conclusions of the workshop highlighted several activities that intersect with the goals of this report, such as assessing the phenomena that impact LBEs and event sequence progressions.

Of the identified LBEs, breaches of the primary fuel salt piping and drain tank would result in the release of radionuclides into the reactor containment cell and represent LBEs that are qualitatively estimated to be DBEs and BDBEs, respectively [126]. The conditions at the time of a breach are important as unique conditions—such as reactor containment cell open or not properly inerted—could result in high consequences. These LBEs and the functional requirements needed to adequately model phenomena of the MST intersect with many of the modeling capabilities described in this section and have been the focus of historical maximum credible accident analysis.

In addition, there may be AOOs with resulting consequences. For example, the failure of the fuel salt pump—likely considered a relatively frequent IE—could alter the reactivity due to loss of forced circulation (i.e., reactor dynamics change as a result of fuel salt residence times changing), resulting in fuel salt heat-up and material challenges throughout the primary system that are difficult to assess without an integral MST modeling capability. Additionally, the scattering of radionuclides across many diverse auxiliary systems during normal operation create unique MST modeling challenges.

Gérardin et al. published an exhaustive study on postulated IEs for the MSFR using both a Master Logic Diagram (MLD) top down approach and the Functional Failure Mode and Effect Analyses (FFMEA) bottom-up approach [127]. They conclude that an update on the safety methodologies and requirements from LWRs is needed due to the lack of potential for core damage and the non-applicability of fuel meltdown in MSRs. Although, the criteria for physical barriers of MSRs need to be stricter than that for LWRs given the circulating fuel salt and fission products. The physical barrier of the core has more potential for leakage than a standard LWR due to the larger volume and more auxiliary systems used such as fuel processing.

For salt-cooled MSRs (such as the FHR), the major sources of radionuclides include the solid fuel and activity present in the circulating salt, such as activation products of impurities, coolant, and corrosion products, of which tritium is a major concern. Similar to salt-fueled MSRs, any auxiliary systems being used will represent sources of radionuclides as well, including an off-gas system, salt cleaning system, and the pebble handling and storage system in the case of the FHR.

In 2013, a whitepaper based on a workshop on LBEs and functional requirements for licensing for the FHR was published [123]. This preliminary investigation into FHR LBEs identified over 30 IEs based on categories used for LWRs and used approaches similar to those taken during NGNP. Understandably, many of the LBEs are similar to ones selected for salt-fueled MSRs above. Additionally, the whitepaper discusses an approach taken for BDBE selection based on previous experiences had by other developers when attempting to license reactors with new and

incomplete technologies. The experts of the workshop suggested eleven bounding events for the FHR:

- Unprotected transient overpower events
- Station blackout
- Protected loss of heat sink
- Unprotected loss of heat sink
- Protected loss of forced circulation
- Unprotected loss of forced circulation
- Large loss of primary coolant
- Overcooling
- Flow blockage
- Failure of hold-down structures
- External events

An analysis of the FHR during very low frequency accidents has also been published [128, 129] and highlights potential design features that can provide enhanced safety of the reactor during a reactivity excursion. This includes discussion on fission product behavior in the fuel and coolant salt, as well as a heat transfer analysis of the system with the CFD code *STAR-CCM+*. This exploratory study concludes that such a system is feasible for use in mitigating radionuclide releases, but further research and development of the concept and models are needed.

4.1 System Modeling Capabilities

As the state of MST assessments for MSRs and FHRs is largely immature, it is prudent to review and highlight code development in reactor system modeling (neutronics, computational fluid dynamics, system-level thermal hydraulics, etc.) that has been recently published. Due to the vast array of designs and the unique modeling functional requirements, the following is meant to be a fairly exhaustive summary of such efforts to provide a comprehensive review of the various approaches taken in modeling circulating fuel reactors and FHRs. As discussed previously, a successful source term assessment will require a deeper understanding of the IEs and event sequence development. Therefore, certain aspects of MST assessments will depend directly on the system modeling code results. For example, during a reactivity insertion accident, the temperature of the fuel salt will theoretically increase up to a limit based on various neutronics, flow, and salt-related properties. Because temperature has arguably the biggest impact on physicochemical properties (i.e., transport), this time- and event-dependent information is necessary to accurately model radionuclide transport for that specific time and event considered.

4.1.1 Salt-Fueled Molten Salt Reactors

A multitude of nuclear systems codes have been developed for modeling salt-fueled molten salt reactors. A summary of recent advances in some of these codes is provided here, and an attempt is made to relate these studies to source term modeling and simulation. To the best of the authors' knowledge, no multi-physics solver has been used to model source term releases in a molten salt reactor. Despite this, they can provide useful information with steady-state and transient analyses, which is typically one of the first steps in an MST analysis. This can include the spatial and time distribution of temperature, power, decay heat, and various flow related parameters. Recently,

Diamond et al. reviewed and tabulated the phenomena important to modeling neutronics and thermal hydraulics in molten salt reactors [130].

Many researchers have built in-house codes with the goal of modeling thermal hydraulics coupled with neutron transport capabilities in circulating fuel reactors such as MSRs. Cammi et al. provides an overview of the efforts completed up to the time of publication and reviews the selected approaches [131]. It should be noted many of the in-house codes may not necessarily be openly available but still showcase the wide number of capabilities in modeling MSRs. For example, Guo et al. developed code that includes a single channel model, a heat transfer model, a heat sink model and a liquid-fuel point kinetic model that takes into account the effect of circulation [132]. They validated the code against MSRE pump startup and coast-down data, and then analyzed an unprotected loss of heat sink (ULOHS) transient with and without a combined unprotected loss of flow (ULOF) transient. They expanded on this work by coupling MCNP to their in-house *MAC* code to calculate effective multiplication factor, neutron flux and power distribution based on MSRE experimental data [133]. Shi et al. added a point kinetic model to *RELAP5* to solve coupled neutronic-thermal hydraulic problems in MSRs with delayed neutron precursor flow, benchmarking against MSRE experimental data [134]. Brovchenko et al. added a point kinetic model to in-house code *REM* coupled to *MCNP* to evaluate safety transients due to delayed neutrons [135].

Wooten and Powers provide a comprehensive review of reactor kinetic models for circulating fuel reactors (CFR) [136], categorizing the three main groups as multigroup diffusion (MGD), point reactor kinetics (PRK), and quasi-static models. They note that time-resolved MGD approaches coupled to a thermal hydraulics code will produce the highest degree of accuracy for transient simulations. PRK models are preferred in situations where computational speed is most important and a high degree of accuracy is not needed, such as transients that do not occur at low powers or involve strong spatial effects. In certain situations when PRK models are not accurate enough, quasi-static-based approaches may be chosen providing computational savings that are at least modest compared to MGD methods.

Hu et al. have utilized *SAM*, described in Section 2.1, for salt-fueled molten salt reactor applications [33]. The code has been shown to model the 1D thermal hydraulic characteristics of MSRE with good agreement to legacy data in terms of temperature and velocity profiles, among other parameters [137]. They note that future work should focus on improvements to the salt properties and equation-of-state in *SAM*, and that coupling the code to a CFD code will improve the accuracy of certain parts of the model. They show that *SAM* can be used to model a postulated loss of flow accident caused by a pump failure. Recently, a delayed neutron precursor drift model and modified point kinetic equations were added to *SAM*, providing some functionality in modeling basic neutronics characteristics of the circulating fuel salt during transients [138]. They model decay heat generation and reactivity feedback in the core. The newly implemented model was benchmarked against MSRE data in three experiment simulations: zero power pump start-up, zero power pump coast-down, and thermal convection heat removal test [139]. The results show good agreement with the experimental measurements, but the authors note that coupling a more robust core neutronics tool to *SAM* may achieve better agreement due to more accurate spatial parameters.

Zhuang et al. developed the *MOREL* code, and later updated it to *MOREL2.0* with various improvements in kernels and methods used to solve neutron dynamics and thermal hydraulics [140]. The code is validated with MSRE data, proves to be of similar accuracy as the previous version, and has increased speedups for both steady-state and transient scenarios by considerable amounts. They later modeled the power and temperature as a function of time for scenarios such as a pump driven transient, an unprotected fuel pump startup and coast-down at zero and normal power, an inlet fuel temperature driven transient, and overheating and overcooling of inlet fuel [141]. This work shows the importance of system modeling for source term modeling, as time and space dependent temperature modeling during transient scenarios has a very important and direct impact on radionuclide speciation, transport, and many other phenomena that affect its evolution during a severe accident. Similarly, Li and Cai developed another in-house code that solves neutron transport with thermal-hydraulic feedback dynamics and uses Serpent-calculated few-group cross sections as input, exploiting different calculation methods to improve efficiency [142]. Comparing results of the MSRE pump start-up and coast-down transients validates the performance of the code for modeling channel-type MSRs.

In 2017, Collins et al. used the deterministic neutron transport solver *MPACT* coupled to the two-fluid thermal hydraulic subchannel solver *CTF* within *VERA* to model MSRs [143]. Also in 2017, Betzler et al. developed a *Python* script called *ChemTriton* which utilized neutron transport solvers and fuel depletion capabilities in *SCALE/TRITON* [144, 145]. Gentry et al. later reported an initial benchmarking of the two code's capabilities, also utilizing the continuous energy (CE) Monte Carlo solver *Shift* with *ChemTriton* to provide a reference solution [146]. They reported that the two capabilities are performing lump depletion and material transfer consistently, but cross section accuracies may vary depending on the system flux spectrum. Continued work in *VERA* has resulted in published preliminary results of the *MPACT-CTF* code for a representative fast spectrum MSR [147], as well as an updated summary of the state of the code for MSR applications which includes species transport capability [148] and the integration of a thermophysical properties database into the simulation tool [149].

This multi-phase species transport capability provides information on the effects of fission product poisons to the neutronics solver but can also be very informative for severe accident source term modeling. Taylor provides a comprehensive summary of the implementation of species transport in the coupled *VERA* codes, *MPACT-CTF* [150]. The species transport capability is used to model bubble growth and removal, as well as the xenon, iodine and helium transport through the salt and/or bubbles. They note that work is ongoing to add in thermochemical modeling capabilities to the code suite, utilizing the *Thermochemica* solver.

As will be discussed further in Section 4.2.1, Betzler et al. have implemented the *ChemTriton* script into the *SCALE/TRITON* lattice physics tool, providing a more efficient and accurate material accountability methodology that is accessible to external *SCALE* users with a defined input [151]. They note that direct implementation into *SCALE* benefits a large number of users as many rely on *ORIGEN* for point depletion calculations, and the CE Monte Carlo solver *Shift* can provide necessary high-fidelity solutions of MSR fuel depletion. The newly developed ability for reactor physics solutions and fuel depletion in liquid-fueled MSRs allows for depletion with continuous removals and delayed neutron precursor flow. Analogous to the work done with *VERA* by other researchers, Vicente-Valdez et al. used the new MSR modeling capabilities in *TRITON* to study the build-up and effect of xenon behavior on reactivity in MSRs, comparing to

MSRE experimental data [152]. They used the deterministic solver *NEWT* and *TRITON* with *ORIGEN* to estimate production rates of ^{135}Xe , also adding a term for effective removal rates based on decay and off-gas system removal. This allowed for analysis of the off-gas system's efficiency in removing the fission product poison and the effect on the poison fraction. Using a unit cell approximation, they created models for the MSRE, MSBR, and MSDR, validating *SCALE*'s new capabilities against *Serpent-2* for the MSRE data.

Similarly, Price et al. examined the physical behavior of xenon in MSRs, including its solubility in salt, the effect of circulating particulates on xenon, and the size, ionization and circulating void fraction of xenon [153]. They expanded on this work by developing a dynamic model of xenon behavior in MSRs accounting for various types of mass transport in a model built in *MATLAB/Simulink* [154]. The model accounts for mass transfer from salt to graphite as well as to bubbles, and factors in decay characteristics and removal due to the xenon stripper. These studies provide relevant discussion on the behavior of volatile radionuclides in molten salt reactors that is directly applicable to understanding source term releases.

Singh et al. studied the dynamic behaviors of MSRE and the two-fluid MSBR concept (which is representative of the design being pursued by Fluide Energy) using models built with *MATLAB/Simulink* [155]. Expanding on the linearized MSRE model built [156], the non-linear MSBR model [157] is based on neutron dynamics (modified point-reactor kinetics equations) coupled to thermal hydraulics with reactivity feedback, including heat transfer in the fuel, graphite, and fertile salt. They analyze MSBR under both positive and negative reactivity perturbations and conclude that the reactor concept is stable and operable at all power levels investigated. The point kinetic equations in the presented model have inherent limitations which include only being able to model reactors of relatively high power (>1 MW) and for transients longer than the core fuel transit time. Additionally, they complete a plant-level dynamics modeling of a prototypical commercial molten salt reactor similar to ORNL's Molten Salt Demonstration Reactor (MSDR) [158].

Greenwood et al. developed at ORNL the *TRANSFORM* code, a flexible *Modelica*-based library of physical models relevant to advanced energy systems, especially circulating fuel reactors such as MSRs [159]. The tool was designed to be as generic as possible to MSRs, but the authors note that its application to different reactor concepts such as fast spectrum reactors or pebble bed reactors may require a swapping out of the neutronics module or thermal hydraulic module, owing to the code's inherent ability to be tailored to the user's needs. For example, they implement a modified point-kinetics model with neutron precursor drift into a system modeling tool for application to a thermal spectrum, fluoride salt-fueled MSR, and further adapt that model for the generation, decay, reactivity feedback, and decay heat modeling of fission products [160]. By treating fission products as trace substances in the bulk flow, they allow for mass accountancy of these low concentration substances without having to worry about its effect on the other physical and chemical properties. They also use the *TRANSFORM* code to model the MSBR concept as a basis for more modern MSR designs [161]. Taking advantage of the publicly available design data, *TRANSFORM* is used to model system dynamics including the distribution of neutron precursors, tritium, and a representative fission product, xenon. The ability of the code interchange numerical models may aid its application for prototyping different source term analysis strategies. This is further evidenced by the code's interfacing with *RAVEN* [162] for a steady-state sensitivity and statistical analysis [161].

Fratoni et al. published the first MSR-related reactor physics benchmark which was reviewed by the International Reactor Physics Experiment Evaluation Project and is included in the handbook starting with the 2019 edition [163, 164]. They used *Serpent-2* to develop a three-dimensional, high fidelity benchmark of the MSRE; methodologies were developed to account for the additional effects on reactivity from having circulating fuel, as opposed to coupling to a separate thermal hydraulics code. The benchmark serves as a baseline foundation for future efforts in validating models against experimental data derived from MSRE, such as the behavior of other fissile isotopes of fuel or the effect of xenon poisoning.

Recently, Fei et al. modeled the thermal spectrum, graphite-moderated ThorCon core in 2D and 3D at steady state, utilizing all three *PROTEUS* neutron transport solvers for their specific capabilities [165]. They used the open-source, stochastic code *OpenMC* to generate cross sections and provide a reference solution, resulting in generally good agreement with the *PROTEUS* solvers at the benefit of computational cost, as well as delayed neutron drift modeling capability with the *PROTEUS-SN* solver. Also recently, Carter and Borrelli modeled Terrestrial Energy's Integral Molten Salt Reactor (IMSR) in *MCNP*, using Idaho National Laboratory's high performance computing resources [166]. The authors note the study is the first in a series to evaluate the effect of fuel burn-up on waste stream source terms and radiotoxicity amongst different potential salt systems that may be used for this reactor. They also recommend future work which may involve coupling to other codes for a more robust multi-physics analysis including *Serpent*, *RELAP7*, *BISON*, *Pronghorn* and *Griffin*.

Recently, multi-physics codes developed at four institutions supporting the SAMOFAR project were benchmarked against each other for their ability to model steady state coupling and time-dependent modeling of a simple representation of the MSFR [167]. Although standardized, the benchmark is still representative of the main features of the MSFR, including coupling between thermal hydraulics and neutronics codes in the fast spectrum with delayed neutron precursor transport. The codes, conveniently referred to by the name of the institution they were developed at, include *CNRS* (Centre National de la Recherche Scientifique-Grenoble), *PoliMi* (Politecnico di Milano), *PSI* (Paul Scherrer Institute), and *TUD* (Delft University of Technology). The results show excellent agreement among the codes for most parameters measured, providing differences of a few tenths of percent in many cases, up to a maximum of a few percent difference in some cases. The authors note differences can be attributed to, among other things, the different meshes used, the neutronics model used, the discretization technique used, as well as the approaches taken to account for various cross-section corrections. For the *CNRS* code, they provided data for two different neutronics solvers, utilizing both the first order (SP_1) and third order (SP_3). Similarly, for the *TUD* code, they provided results from neutronics solvers of order S_2 and of order S_6 . They conclude that this benchmark provides a very useful systematic process for comparing multi-physics codes of all types of MSRs, although thermal spectrum reactors will need reconfigured models to factor in differences in phenomena due to moderated-core designs.

A complete description of work done by all four institutions is omitted for brevity although recently published work on the *TUD* code is provided here as an example of some of the capabilities these codes can provide in multi-physics modeling. In this study, Tiberghien et al. provide a brief history of neutronics coupling to thermal hydraulics codes for MSRs [168]. They note the progression from early models that combined neutron dynamics and fluid dynamics with strongly simplifying assumptions, to codes with full multi-physics solvers that coupled more

advanced neutronics codes (multi-group diffusion) with thermal hydraulics codes that model incompressible flow and Reynolds-Averaged Navier-Stokes equations (RANS). They provide background on the *TUD* code, which involves coupling of an incompressible RANS/k- ϵ model with a solver for the S_N multi-group neutron transport equation with delayed neutron precursors. The code is utilized in simulating the MSFR at steady-state and during an unprotected total loss of power accident. Meshes for distributions of power density, temperature, fluid velocity, DNP, eddy viscosity, and turbulent kinetic energy are provided, as well as distributions with respect to time for most parameters during the transient. Additional work on these codes was done to quantify the uncertainty associated with various input parameters, and their effect on the maximum temperature and k_{eff} [169]. Using Polynomial Chaos Expansion, it is found that the maximum temperature is mainly sensitive to the heat transfer coefficient and reactor power, while the effective multiplication factor is more sensitive to the neutronics parameters, rather than fluid or heat transfer parameters.

Similarly, Wan et al. also developed an in-house multi-physics solver to model the MSFR during steady-state and transients, providing spatial and time distributions of various parameters [170]. Specifically, they simulate the effect of both fissile and fertile fuel additions to the reactor, noting the evolution of power and core-averaged temperature during the transient. These tools may provide useful analysis capabilities for severe accidents. For example, modeling salt temperature and power distributions during a fuel salt spill accident, or conversely during a fuel addition from an on-line processing system, will be necessary when constructing a mechanistic source term assessment for MSRs.

Shemon et al. are developing multiphysics capabilities within the *SHARP* toolkit under NEAMS for fast reactors [45]. Solving the delayed neutron precursor distribution has been outlined as a major challenge in modeling fast spectrum MSRs. Therefore, efforts are ongoing to couple *PROTEUS* neutronics solvers to the CFD code *Nek5000*. Two simultaneous efforts are being pursued: coupling the *PROTEUS-NODAL* solver to *Nek5000* via one approach, and the *PROTEUS-SN* solver via another. The various advantages of each of these solvers are outlined in the report, but Figure 4-1 displays the coupling approach for *PROTEUS-NODAL/Nek5000*.

Figure 4-1: PROTEUS-NODAL/Nek5000 Coupled Solution Approach for DNPs in Fast MSRs [45]

Several researchers have used CFD to simulate detailed flow patterns or particle transport in MSRs. Pauzi et al. used the *ANSYS FLUENT* software to model natural circulation flow inside a fuel salt tube [171]. This can be useful in providing details about the location of laminar and turbulent flows as well as the temperature profile which may have an impact on radionuclide deposition on the wall, as will be briefly discussed in Section 4.2.4. Podila et al. report the first high fidelity 3D thermal hydraulics modeling of the MSRE core using stand-alone CFD simulations [172]. They use the *STAR-CCM+* code [173], noting the benefits of its implemented turbulence models and ability to be coupled with the system thermal hydraulics code *RELAP5-3D* and the reactor physics code *Serpent*. Predictions of the salt's local flow and heat transfer properties were compared against MSRE data with excellent agreement. The authors conclude that in order to accurately predict the primary system temperature distribution in an MSR, complete knowledge of 3D flow behavior around plenums is needed to provide fuel channel flow conditions, as opposed to assuming uniform flow conditions for all channels, especially due to the important role temperature reactivity feedback effects play in core neutronics.

Finally, it should be noted that many studies have been performed on modeling MSR fuel cycles and fuel processing systems, but a discussion of these studies is omitted here and addressed in Section 4.2.8, as they relate to source term modeling of the fuel processing system of an MSR.

4.1.2 Fluoride Salt-Cooled High Temperature Reactors

Just as with salt-fueled MSRs, considerable work has been completed in modeling various aspects of the reactor system for FHRs. Many of the codes used in modeling reactor physics and thermal hydraulics of MSRs that were discussed in the previous subsection may also be applicable to FHRs, but with alterations to the core design and parameters. Additionally, because many of the studies on thermal hydraulics beyond the core of MSRs are potentially applicable to FHRs with minimal modification, the reader is directed to Section 4.1.1 for additional insights in modeling high temperature, liquid salt systems.

Rahnema et al. have published considerable literature on the phenomenology of FHRs, including the challenges in modeling and simulation of FHRs [174], the required level of multiphysics coupling corresponding to various phenomena [175], and important phenomena and gaps for neutronics modeling [176]. Similarly, Lin et al. published a phenomena identification and ranking table (PIRT) for thermal hydraulics modeling [177], and Singh et al. published a PIRT for metallic structural materials [178].

In 2013, four white papers summarizing corresponding FHR-related workshops were published, and in the second report, Cisneros et al. provide an outline of potential methods and experiments relevant to modeling the FHR, including thermal hydraulics and neutronics [124]. They mention the applicability of *RELAP5* for thermal hydraulic modeling, as well as *Flownex* for CFD, similar to its use in modeling the pebble bed modular reactor (PBMR) [179]. The use of *RELAP5/MOD4.0* for pebble bed FHRs has also been shown by Yang et al. for a reactivity insertion transient analysis [180]. Candidate codes for high-fidelity criticality analysis (HFCA) include the stochastic codes *MCNP*, *SCALE/KENO-VI*, and *Serpent*, as well as the deterministic code *DRAGON*. The candidate tools for depletion analysis and transient analysis include the *SCALE* codes *ORIGEN* and *TRITON*, *REBUS3/DIF3D*, *Serpent*, *PEBBED-THERMIX*, *WIMS*,

and *PARCS*. The reader is directed to this document for a more detailed discussion on candidate codes for thermal hydraulic analyses and neutronics analyses.

In 2017, a white paper titled “The Challenges in Modeling and Simulation of Fluoride-Salt-Cooled High-Temperature Reactors” was published [174]. It provides a high-level overview of reactor system modeling codes that are applicable to FHR modeling. The reader is directed to this document for a complete discussion on all of these codes, but the neutronics and thermal hydraulics codes are listed here for brevity:

Neutronics codes:

- *SCALE*
- *MCNP*
- NEAMS Toolkit (*PROTEUS*, etc)
- *Argonne Reactor Codes (ARC)*
- *COMET*
- *Serpent*

Thermal hydraulics codes:

- *RELAP5, RELAP5/MOD3, RELAP5-3D*
- *TRACE*
- *SAM*
- *ANSYS FLUENT*
- *STAR-CCM+*
- *COMSOL*
- *OpenFOAM*
- *Nek5000*

Ge et al. completed a CFD study of the flow in a pebble bed FHR using *FLUENT*, providing results via two approaches: the porous media approach and the realistic model approach [181]. The porous media approach calculates the temperature and pressure distribution but can only provide a uniform velocity distribution due to the averaged porosity characteristics used in the model, so the detailed local flow and heat transfer is not calculated. In the realistic model, distributions of temperature, pressure, velocity, and non-dimensional velocity are obtained, yet at the expense of higher computational cost. The authors conclude the porous media approach provides valid information relevant to the design and operation of an FHR, yet the realistic model with more detailed local flow capabilities is best for safety analyses and specialized problems. Recently, the culminating report of a U.S. DOE NEUP (Nuclear Energy University Program) project on experimental and computational analysis of pebble bed reactors using the CFD code *Nek5000* was published [182]. High-fidelity direct numerical simulations (DNS) were performed using experimental datasets and reconstructed geometries of randomly packed beds. Compared with correlations in literature were characteristics of the flow domain such as volume averaged porosity, axial porosity, and radial porosity, as well as friction factors.

As mentioned previously, the *SAM* code was developed for advanced reactors with liquid coolants of single-phase, incompressible but thermally expandable flow [31, 32, 183]. Ahmed et al. showed its use in modeling a natural circulation cooling system of the FHR, benchmarking

against *RELAP5-3D* [184]. They also note that one of the code's benefits is the ability to easily couple other *MOOSE*-based codes, for example, such as a CFD code for high-fidelity comparisons of specific regions of flow or heat transfer within the system.

Pronghorn is a finite element (FE), porous media thermal-hydraulics simulation code built on the *MOOSE* framework made for pebble bed reactors [34]. As mentioned previously, porous media models cannot capture flow details around the pebbles or the highly asymmetric drag and heat transfer in the bed, therefore *Pronghorn* is best utilized for design scoping studies or core boundary condition elucidation at a fraction of the computational cost of higher fidelity codes. Advantages of the code over other porous media model codes include an arbitrary equation of state, unstructured mesh capabilities, compressible flow models, the ability to couple to other *MOOSE* codes such as a fuels performance code or a systems-level thermal-hydraulics code. The code was validated against experimental data from the SANA pebble bed experiments which ran in the 1990's. *Pronghorn* predicted lower average error and standard deviation than the codes *Flownex* and *GAMMA* [35, 185].

Brown et al. published core design and safety analysis of a preconceptual FHR demonstration reactor [186], utilizing *Serpent* and a modified version of the nodal diffusion code *PARCS* [187]. *PARCS* has spatial kinetics capability with a standalone thermal hydraulics module for single phase coolant flow. They note confidence in *PARCS* results due to comparisons with *COMSOL*, *RELAP5-3D*, *TRACE*, and the FHR version of the *NESTLE* nodal core simulator.

Wang utilized *Serpent*, *COMSOL*, and *Python* to build multiple computational models of the FHR with coupled neutronics and thermal hydraulics and different levels of spatial and energy resolution [188]. These models include coupled heat diffusion and point kinetics unit cell models, Monte Carlo neutronics models, and coupled multigroup neutron diffusion and multi-scale porous media model. They model two popular designs of FHRs at steady state power, neutron flux, and temperature distributions, as well as reactivity insertion and overcooling transients.

Recently Shen introduced a code-to-code verification benchmark for FHR using *Serpent-2*, *MCNP6*, and *SCALE6.1* [164]. Because no variation of the FHR has ever been operated, these computational benchmarks will be important when comparing future neutronics analyses such as design scoping completed by a developer. For example, Kairos Power is developing a pebble bed FHR concept, and recently they acknowledged the use of *Serpent-2* as an important computational tool in their MST strategy [189]. Additionally, they are developing modified, in-house versions of *SAM* for system thermal hydraulics of the KP-FHR.

4.2 Source Term Modeling Capabilities

Figure 4-2 provides a summary of radionuclide transport phenomena for a salt-fueled MSR [190], with a focus on radionuclide releases from the primary fuel salt. As can be seen in the figure, there are multiple physical and chemical processes that require consideration for the analysis. Some of the most important focus areas of a related MST assessment are expanded on in the following subsections, based on phenomena that were discussed in a previous report [2]. These focus areas include:

1. Radionuclide inventory calculation
2. Salt thermochemistry
3. Bubble transport modeling

4. Corrosion and deposition modeling
5. Salt release scenarios
6. Aerosol modeling
7. Tritium modeling
8. Salt processing system considerations
9. Off-gas system considerations

Figure 4-2: Summary Diagram for Salt-Fueled MSR Radionuclide Transport Phenomena [190]

Preliminary evaluations of the source term of MSRs during severe accidents have been published. Vaporization experiments of cesium and iodine binary systems with lithium fluoride were completed to determine vapor pressure for corresponding volatile species [191]. The same study also assessed the corrosion behavior of Hastelloy-N with fluoride salts containing cesium iodide and under air.

More recently, Kalilainen et al. published the first study which couples analyses from a fuel depletion code, a salt evaporation numerical model, and a thermodynamic modeling tool in order to assess the temperature dependent release of fission products during a salt spill [62]. They use the *EQL0D* routine to estimate fuel isotopic equilibrium, numerical models within *MELCOR* to assess salt evaporation, and the *GEMS* thermodynamic solver with the *HERACLES* database to

calculate vapor species distributions. This work is representative of the modeling strategy necessary for a salt spill scenario, which represents one aspect of the overall MST methodology.

A summary diagrams of radionuclide transport phenomena for a salt-cooled MSR can be seen in and Figure 4-3 [58]. The source term strategy for FHRs will be similar to MSRs as the choice of coolant has a considerable impact on radionuclide release and the main adjustment needed is in estimating the radionuclide inventory that results from TRISO particle releases, as opposed to assuming all fission products and activation products end up in the salt. Cisneros et al. outlined safety design criteria (SDC) for the FHR in the first of four white papers released in 2013 [123, 192]. The first SDC discussed is “Maintain Control of Radionuclides”, stressing the importance of source term modeling to the safety of the reactor and its impact on licensing. They outline seven engineered safety functions related to this SDC:

- Tristructural-isotropic (TRISO) particle fuel integrity
- Primary coolant chemistry, particulates, and inventory control
- Tritium control and recovery
- Cover gas chemistry, particulates, and inventory control
- Reactor cavity low-pressure containment
- Fuel transfer and storage
- Reactor citadel filtered confinement

Figure 4-3: Summary Diagram for Salt-Cooled MSR Radionuclide Retention Phenomena [58]

MELCOR

MELCOR has been outlined by the NRC for regulatory use in the in assessment of both FHRs and MSRs source term analyses. The models within *MELCOR* are generic and can be adapted to different reactor types by implementing relevant thermal fluid properties and other physical models. Recently *MELCOR* was assessed for use in modeling molten salt systems by the NRC and contracted experts. These reviews highlighted a pathway forward for the *MELCOR* program to accurately capture behavior in these systems. Of particular importance is a better experimental database of chemical and thermal fluid behavior of molten salt itself and entrained radionuclides. Modeling gaps and existing capabilities within *MELCOR* are highlighted in Table 4-1. There are significant commonalities within core structures of FHR reactors and HTGR reactors, meaning that the majority of the core component models already exist for FHR systems [58]. This includes the models for capturing the behavior of TRISO fuel.

Table 4-1: Key Accident Progression Phenomena for MSRs [58]

Key Phenomenon	Importance	Existing Capabilities	Modeling Gaps
Physical Properties	Fundamental to simulation of steady state temperature and flow distributions.	FLiBe EOS and properties already implemented in MELCOR.	Validation of properties (development items M3.1, M3.4 and M3.6)
Heat Transfer Coefficients	Transfer of heat to calculate heat loads to structural materials	Existing generic correlation forms	Implement and validation of heat transfer coefficients (development items M3.4 and M3.6)
Track the flow of gas through the molten salt	Important for calculating entrainment of fission products from molten salt (next item)	SPARC model for aerosol scrubbing in liquid pools exists in MELCOR	Extend the SPARC model and bubble rise model (development items M3.2).
Entrainment of contaminated molten salt droplets in the gas flow	The primary mechanism for such entrainment of droplets is of course the rupture of gas bubbles at the molten salt surface.	Similar capability exists for molten corium pool	Use of correlations derived from data for droplet formation during bubble bursting in aqueous systems. This phenomenon is described further in Appendix C and is part of development Item M3.2 MSR
Vaporization of fission products from the molten salt.	Release of volatile fission products to cover gas.	Similar capability exists for molten corium pool	This phenomenon is described further in Appendix C and is part of Development Items M3.2 and M3.3 MSR

To date, equations of state for FLiBe have been implemented into *MELCOR*. However, other salts that may be used in advanced reactor systems have not been added to *MELCOR* yet, including chloride salts, nitrate salts and other fluoride systems. Generic forms of heat transfer models are currently implemented within *MELCOR*, and salt-specific heat transfer coefficients are currently being implemented as experimental data becomes available [58].

The framework to capture radionuclide behavior also exists within the *MELCOR* framework as part of the *SPARC* model, in-vessel molten core models and ex-vessel corium models. As with thermomechanical models within *MELCOR* additional experimental data will be needed to validate any new models implemented within *MELCOR*. Such data is needed for each relevant salt system since each system due to inherent differences in chemical behavior.

Future development of *MELCOR* for MSR systems includes:

- Validation of molten salts within control volumes as a working fluid
- Implementation of the ability to simultaneously model multiple working fluids within the same control volume to capture condensable species behavior
- Chemistry models to capture salt thermodynamics and salt/material interactions
- New aerosol physics models specifically designed for molten salt systems
- Natural circulation models for molten salt within large pools
- MSR and FHR-specific component models for both primary and secondary systems [58].

The proposed NRC evaluation model for salt fueled reactor systems can be seen in Figure 4-4. Within this model, *AMPX* is to be used for cross section generation, *SCALE* is to be used for reactor physics simulations, *MELCOR* is to be used for system behavior and fission product transport within the system, *DIF2* is to be used for fission product diffusion within the fuel, and *MACCS* is to be used for consequence analysis. The tool to be used for fuel fracture modeling has yet to be determined [58].

Figure 4-4: Proposed NRC Evaluation Model for Salt Fueled Reactors [58]

Path Forward

Recently, Jerden et al. outlined a path forward for the development of a MST approach for MSRs that is both rigorous and technology neutral [193]. They conclude there remains a need for a thorough, step-by-step source term analyses that includes parametric sensitivity analyses and trial calculations. It is recommended that the following two types of models be developed:

1. A simplified mass balance model that is compiled from existing information and tools which can be used for sensitivity studies to rank the importance of specific radioisotopes and release pathways in term of off-site dose consequence.
2. A mechanistic model that provides a realistic, science-based assessment of the radionuclide release and retention that accounts for all key transport phenomena. This mechanistic MSR source term model would provide feedback to reactor design teams regarding the impact of design features and could eventually provide a means of justification for reducing the size of emergency planning zones and site boundaries for new MSR reactors.

Item (1) was recently fulfilled when a simplified approach for a scoping assessment for non-LWR source terms was published [11]. The present report aims to assess the current state of computational capabilities needed to complete item (2). Additionally, while developing these MST modeling tools, it is recommended to use the following risk analysis tools to determine postulated accident initiating events: master logic diagrams (MLD) and functional failure mode effect analyses (FFMEA). This can narrow the focus of the MST strategy as well as allow the source term model to provide feedback into the reactor design process. A conceptual pathway for MSR source term development is provided in Figure 4-5 [193].

Figure 4-5: Conceptual Pathway for Developing a Rigorous MST Analysis for MSRs [193]

4.2.1 Radionuclide Inventory

4.2.1.1 Salt-Fueled Molten Salt Reactors

The first step of any source term modeling strategy is to estimate the inventory of radionuclides, which can be a complex analysis for fuel-salt MSRs. This includes not only the production of radionuclides due to fissions but also the activation, or neutron absorption, of various materials such as the coolant, moderator, and structural materials in the core and peripheral systems. The capability of fuel depletion and neutron activation modeling for source term analysis is arguably more important for MSRs than for solid fueled reactors because of the larger variety and volume of materials in a flux of neutrons when you have a fuel that is not stationary. Although this will require a detailed understanding of the reactor core design and associated systems, some tools are being developed to be generic enough to be used by most developers of MSRs. As highlighted in Figure 4-4, the proposed NRC evaluation pathway relies on the use of the *SCALE* suite for this task. A brief discussion of work that has been published on modeling depletion and activation analysis for MSRs is provided here, including *SCALE* as well as other codes.

Owing to its modular nature with various solvers and codes, the highly customizable *SCALE* modeling suite can be used to provide the following information to a severe accident analysis code: radionuclide inventory, decay heat, power distribution, kinetics data including six-group delayed neutron precursor kinetics with precursor drift, and temperature and void reactivity coefficients. MSR-specific capabilities and models in *TRITON* are available in version 6.3 beta 1, and a more complete implementation is expected when version 6.3 fully releases [58]. This new MSR capability in *SCALE/TRITON* is able to calculate the scalar flux, power, and isotopic distribution as a function of core operation, but a more formal assessment with validation work is needed to assess accuracy limits of the geometric representation. Volume 3 of the NRC Non-LWR Vision and Strategy discusses development plans for *SCALE* for MSRs, including delivering tested and quality-assured *ORIGEN* reactor libraries for MSRs, providing a simple and more straightforward approach to analyzing isotopic distributions. Table 4-2 summarizes the maturity levels of *SCALE* for MSR analysis.

As mentioned previously, a neutronics modeling capability for MSRs has been implemented into the *TRITON* depletion modeling tool within *SCALE*, including continuous removal and tracking of isotopes [151]. Time-dependent modeling of isotope concentrations provides necessary information to source term tracking. Using this tool, a fuel cycle uncertainty analysis was completed using parameter perturbation with the *SAMPLER* tool, also within *SCALE* [194]. The authors completed this sensitivity study on the MSDR, showcasing the tool's ability to remove certain elements from the salt during depletion simulations. *SCALE*'s MSR capability was utilized in a study of fuel cycle analysis and depletion modeling for four fast reactor concepts [195]. The authors compared the tool, which used a 2D unit cell approximation, against full core models in *Serpent-2*, showing very good agreement.

Earlier versions of *SCALE* have also been used to model MSRs by coupling the tools in *SCALE* that are normally used for stationary cores to in-house codes that iteratively solve burn-up after each timestep. Sheu et al. developed methods to utilize *SCALE/TRITON* with the stochastic solver *KENO-VI* to solve neutronics for the fast reactor concept Molten Salt Actinide Recycler & Transmuter (MOSART) [196]. With *SCALE*'s point depletion solver *ORIGEN*, they used an in-

house script to account for fuel loading and fission product removal. Other researchers have used similar methods utilizing *SCALE* to do comparable burn-up analyses for the thermal spectrum Thorium Molten Salt Reactor (TMSR) [197] and a smaller MSFR [198].

Table 4-2: *SCALE* Maturity for MSR Analysis [58]

Element	Maturity Level ¹	Comments
Representation and Geometric Fidelity	2	<i>SCALE</i> 6.3 includes a translation of the ChemTRITON capability into TRITON-MSR, described in a peer-reviewed journal article, which allows initial characterization of MSR designs to support MELCOR data needs. However, further assessment/validation work is required to assess accuracy limits of the geometric representation.
Physics and Model Fidelity	1	Higher-fidelity solutions are possible through VERA-MSR. Neutronics and depletion physics are the same as in the rest of <i>SCALE</i> , which should receive 2 or 3, however the fluid flow model is highly simplified, and designed to be applicable for estimating fuel cycle parameters (e.g. thorium feed required for a particular design). Assessment of the accuracy compared to higher-fidelity solutions are possible through VERA-MSR.
Code Verification	2	<i>SCALE</i> SQA requires review of implementations by an external party not involved in the development of that particular capability (hence external). However, this does not approach the rigor for LWRs of implicit external verifications by one of the thousands of code users who have received and used <i>SCALE</i> for LWR analyses.
Solution Verification	2	<i>SCALE</i> SQA requires review of capabilities by an external party not involved in the development of that particular capability (hence external). However, this does not approach the rigor for LWRs of explicit external verifications as part of a document such as an NRC NUREG.
Model Validation	1	Some comparisons of ChemTRITON to MSRE were made. The methodology in TRITON-MSR is essentially the same with some improvements but re-evaluation has not yet been formally performed.
Uncertainty Quantification and Sensitivity Analysis	1	Uncertainty and sensitivity capabilities for any model in <i>SCALE</i> are available through <i>SCALE</i> /Sampler, however specific sensitivity and uncertainty analyses for the MSR have not been performed yet.

¹Maturity Levels

- level 0, little or no assessment of accuracy and completeness and highly reliant on personal judgment and experience;
- level 1, some informal assessment of accuracy and completeness, and some assessment has been made by an internal peer review group;
- level 2, some formal assessment of accuracy and completeness, and some assessments have been made by an external peer review group; and
- level 3, formal assessment of accuracy and completeness, and essentially all assessments have been made by an independent, external peer review group.

Besides *SCALE*, there have many efforts with other tools for estimating the inventory of radionuclides in MSRs, whether for fuel cycle analyses or otherwise. In 2005, Nuttin et al. developed an in-house program which used *MCNP* iteratively to calculate static core neutronics and calculate isotopic distributions in MSBR [199]. The evolution code was later updated and named *REM*, being used for numerous MSFR analyses in conjunction with *MCNP* [135, 200-204]. For example, Brovchenko et al. [135] used the coupled codes to calculate the radionuclide inventory, using the information to estimate contributions to decay heat. Additionally, Doligez et al. used the tool with a numerical model of the reprocessing system to calculate radionuclide inventories in various stages of the auxiliary system [203].

Within the *ERANOS* code platform, a depletion analysis tool named equilibrium fuel cycle procedure for fast reactors (*EQL3D*) was developed by Krepel et al. [205]. Although developed for solid-fueled reactors, Fiorina et al. modified the code's procedure to simulate on-line fuel reprocessing with physical decay constants and blanket salts for application to MSFR [206, 207]. Assuming constant core power, the code can calculate radionuclide inventories at equilibrium for

various fuel and feed types but assumes a constant total concentration of actinides and fission products.

Aufiero et al. extended the capabilities of the Monte Carlo code *Serpent-2* by adding continuous on-line fuel reprocessing to permit MSR fuel burn-up and evolution, thereby reducing uncertainties that had previously been assumed for the more-approximating calculation methods [208, 209]. They compare the code's new capability against the *EQL3D* procedure mentioned above, showing good agreement, but noting that the less energy groups used by *ERANOS-EQL3D* underestimated the production of ^{232}U over time. This is one of the most important isotopes with dose considerations, as its daughter ^{208}Tl has very high energy gamma rays.

Recently, Hombourger et al. used the *MATLAB*-based *EQL0D* code, a modification of the *EQL3D* code for MSRs which has point-depletion to approximate the mixing of circulating fuel, and modeled fuel cycle equilibria of both MSBR concepts and the MSFR [210]. Instead of using the fuel cycle procedure with the *ERANOS* code, it was used here with *Serpent-2*, and the authors note *EQL0D* version 3 is not limited to only some actinides like previous versions. The procedure is capable of feed and removal streams and was designed to either handle fuel evolution over a finite number of steps like a standard depletion code or can search for equilibrium directly and conclude there. Limitations include the inability to account for DNP drift and the inability to account for depletion (or lack thereof) in regions of low or zero neutron flux.

Serpent-2 was used to model a breed & burn MSR [211], initially configured from the design of a Stable Salt Reactor concept representative of the reactor pursued by Moltex Energy. The author also compares the *Serpent-2* reference solution to calculations by a neutronics software package called *WIMS* [212] to assess the accuracy of its deterministic solver for several different pin cell model geometries and fuel configurations; they note good agreement for the 2D geometries. Separately, *WIMS* was also used to model a fast-spectrum, breeder MSR and showed good agreement of the actinide inventory over time when benchmarked against *ERANOS* [213]. The authors also note the code's ability to provide on-line refueling and removal as continuous processes or batch.

Several MSR depletion analyses have been completed by using *MCNP* to model the neutronics of a thermal spectrum core and then coupling to a depletion solver. Ahmad et al. used *MCODE*, which links *MCNP* to the point depletion code *ORIGEN2*, to calculate reactor burn-up in different variants of the Denatured Molten Salt Reactor (DMSR) [214]. Unfortunately, only relatively infrequent refueling is modeled with no fission product removal streams. The MSBR core has also been modeled using *MCNP* neutronics with *CINDER90* depletion and an in-house *Python* script to account for batch reprocessing or refueling at each time step for both whole core analysis [215] and multi-zone modeling [216].

Recently, Nelson et al. developed *Advanced Dimensional Depletion for Engineering of Reactors (ADDER)* to meet general reactor design and fuel cycle analysis needs [217]. Written in *Python*, *ADDER* couples to external neutronics solvers such as *MCNP* and provides point depletion capability either by coupling to *ORIGEN2* or uses its internal depletion solver. *ADDER* is meant to be agnostic to the reactor type, but MSR-specific capabilities are included, and further development is ongoing. Additionally, it allows for fuel management with continuous feed and

removal, has support for multiple salt loops, and provides depletion capability in flows outside the core as well.

For a salt-fueled MSR, the radionuclide inventory may not consist only of fission products and transmuted actinides. Modeling the activation of materials outside the fuel are needed, and *SCALE* provides many such capabilities in activation analysis of stationary geometries but combining this with salt loop modeling could prove to be beneficial. For example, Zhou et al. developed a numerical model in *Mathematica 7.0* for the generation, decay, and flow of activation products in the secondary loop of a MSR [218]. Based on the spatial flux distribution generated from *KENO-VI*, the model estimates the production of coolant activation products such as ^{19}O , ^{20}F , ^{16}N , as well as activation of key structural materials such as Fe, Ni, Co, Cr, and Mn. The model divided the loop into six solution regions to estimate time- and spatial-dependent activity, assuming 1D flow in the loop. To allow for conservative estimations of circulating activity, the model did not account for deposition of activation products within the loop nor leakage from the loop.

After the radionuclide inventory is estimated with one of the tools mentioned above, it would be beneficial to use a thermochemical modeling tool to predict chemical species formation of the activation and fission products. This is especially important in MSRs, as chemistry plays a major role in the phenomenology of the reactor. Figure 4-6 shows a generalized scheme of fission product elements that may be connected via beta decays for certain isotopes, along with the expected chemical group designation for that element in an MSR. It would be beneficial to have simulation capabilities with thermochemical modeling coupled with time-dependent fuel depletion and decay. For example, there are plans to integrate the Gibbs free energy minimization solver *Thermochimica* into *ORIGEN* [48, 58], as well as coupling *Thermochimica* to the *VERA* codes *MPACT-CTF* [48, 150]. A more detailed discussion on thermochemical modeling for molten salt systems is provided in Section 4.2.2.

where (1) - salt seeker,

(2) - noble gas,

(3) - noble metal, and

(4) - noble metal or salt seeker depending on oxidation-reduction state of fuel salt.

Figure 4-6: Generalized Beta Decay Scheme of Fission Products with Chemical Class Designation [219]

4.2.1.2 Fluoride Salt-Cooled High Temperature Reactors

Estimating the radionuclide inventory in FHRs is very different than that for salt-fueled MSRs. Due to the use of TRISO fuel, neutronics modeling of the core will more closely resemble that for the HTGR than any other type of reactor. Therefore, the reader is directed to Sections 3.2.1 and 3.2.2 for discussions on radionuclide inventory calculation and TRISO fuel particle releases, respectively, for HTGRs. Depending on the following two main factors, the same tools that are used for a corresponding HTGR reactor can generally also be used for an FHR with simple modifications made in core and coolant materials properties:

- TRISO fuel particle type (UCO, UO₂, etc.)
- Fuel geometry and type (pebble bed, prismatic, etc.)

For example, Kairos Power is designing a packed pebble bed FHR which uses high assay low enriched uranium (HALEU) fuel kernels in the form of uranium oxycarbide (UCO) [189]. They are using *Serpent-2* for neutronics and fuel depletion modeling. One difference between FHR and HTGR source term inventory calculations will be the contributions from activation of the coolant (or other core materials used) that may differ from helium-cooled HTGRs. Whether using the *SCALE* suite, for which much development has been completed in modeling HTGR neutronics, or using *Serpent-2*, the modeler will need to be cognizant of radionuclides formed by neutron activation.

In Kairos Power's source term methodology report, they also note the very important use of a fuel performance tool to assess the mechanical integrity of the TRISO-coated particles and the retention of fission products by intact or potentially failed particles [189]. For this, they are using *BISON* with in-house modifications and will verify and validate as part of their fuel performance methodology. This is a crucial part of the MST strategy as the source term is composed of radionuclide releases from failed TRISO fuel particles, as well as activated core and coolant materials and contaminants or impurities of the coolant salt. *KP-BISON* will be used to determine the material-at-risk (MAR), or source term, associated with the fuel.

The FHR is similar in design to many HTGR pebble bed reactors which have TRISO fuel particles embedded in spherical fuel compacts. Both reactors envision most fission products being captured in the fuel; however, some fission products have been shown to escape the fuel during irradiation and as a function of temperature and burnup. These TRISO particle release fractions were discussed in more detail in Section 3.2.2 and have been covered extensively in literature [95, 220]. ³H, or tritium, is a unique problem to FHR and MSR designs, and is initially in the form of ³HF (aka TF) in fluoride salt coolants. A concern is that the ³H can diffuse and escape through most metals at FHR temperatures (~700 °C). The ³H is generated by neutrons leaving the fuel and activating certain lighter nuclei in the fluoride coolant salt. Carbon-based materials such as graphite are a sink for ³H where it is adsorbed on the graphite fuel pebbles and along the graphite structure in the core and may transport further into the graphite. Tritium modeling is discussed further in Section 4.2.7.

As discussed in Section 3.2.1, the use of *SCALE* for core neutronics modeling of HTGR/FHR has already begun with further development planned [58]. *SCALE* provides radionuclide inventories, decay heat, power distributions, kinetics parameters, and reactivity coefficients for thermal feedback. *SCALE*'s established history of modeling solid-fueled reactors allows for

straightforward model building while providing unique capabilities for continuous-energy and multigroup neutronics as well as source term analysis. A 3D Monte Carlo model of the HTR-10 benchmark is shown in Figure 4-7 as an example.

The most recent version of *SCALE* has the capability to calculate the core spatial flux spectrum and power distribution of an FHR given the representative core model, calculate tritium content and generation, and analyze isotopic distribution and fuel depletion [58]. Planned development tasks include delivering production-quality *ORIGEN* reactor libraries for FHR pebbles, tritium modeling in *ORIGAMI*, and calculating core distributions as direct input for *MELCOR*. A preliminary evaluation of the maturity levels for the *SCALE* code for FHRs is displayed in Table 4-3.

Figure 4-7: 3-D Monte Carlo Model of the Gas-cooled HTR-10 Benchmark in SCALE [58]

Table 4-3: SCALE Maturity for FHR Analysis [58]

Element	Maturity Level ¹	Comments
Representation and Geometric Fidelity	2	<ul style="list-style-type: none"> • The 3D continuous energy Monte Carlo neutronics + depletion capabilities essentially allow the maximum fidelity of representation within nuclear engineering for predicting neutron and nuclide distributions throughout the system, but with an associated high cost of calculation. Depending on the system, only the formality of assessment varies. • The 1D/2D multi-group energy deterministic capabilities allow a much faster running capability ideal for design and safety analysis, where many perturbations/variations are needed. Depending on the system, the modeling strategy may need refinement, e.g. the double-het treatment to support multi-group, deterministic TRISO calculations.
Physics and Model Fidelity	2	<ul style="list-style-type: none"> • Need for more complete test data to support depletion validation. • Validation data for the working fluid to support model approximations would be needed, and support nuclear data refinement
Code Verification	2	<ul style="list-style-type: none"> • Extensive SQE, many capabilities have been benchmarked and some peer review.
Solution Verification	2	<ul style="list-style-type: none"> • Some informal assessments both internally as well as assessment by code users.
Model Validation	2	<ul style="list-style-type: none"> • Extensive validation of most physics models though not all within the domain of HTGRs. • Aspects of FHRs which are markedly different from NGNP designs require additional verification of modeling fidelity.
Uncertainty Quantification and Sensitivity Analysis	1	<ul style="list-style-type: none"> • Uncertainties and numerical propagation of errors has been examined extensively for LWR applications though not for FHR application

¹Maturity Levels

- level 0, little or no assessment of accuracy and completeness and highly reliant on personal judgment and experience;
- level 1, some informal assessment of accuracy and completeness, and some assessment has been made by an internal peer review group;
- level 2, some formal assessment of accuracy and completeness, and some assessments have been made by an external peer review group; and
- level 3, formal assessment of accuracy and completeness, and essentially all assessments have been made by an independent, external peer review group

4.2.2 Salt Thermochemistry

The ability to accurately model radionuclide speciation and the chemical behavior of molten salts is essential for the design and safety evaluation of MSR concepts. In a recently published report on MSR fuel salt qualification methodology, the liquid fuel salt was deemed to have three safety functions: to retain radionuclides, to act as heat transfer medium for both operational and decay heat removal, and to provide net negative reactivity feedback [221]. The salt thermochemistry is a dominant factor in all three of these safety functions, and for source term modeling, it is especially important in estimating radionuclide retention in the salt. The functional needs of thermochemical models include the ability to predict:

- ***Vapor pressure and boiling point.*** The volatilization of radionuclide species, as determined by their vapor pressures above the fuel salt, is the leading thermodynamically driven transport phenomenon that can result in radionuclide release from reactor containment. The vapor pressures of radionuclide species above molten fuel salt can be determined by Gibbs free energy minimization calculations according to the CALPHAD approach, as described below. The molten salt compositions of interest to MSR concepts have low boiling points, and therefore enable low operating pressures. However, a transient involving excessive salt heating could result in a build-up of reactor containment pressure. Phase diagrams and vapor pressure data have been experimentally measured for many fluoride and chloride salt systems, especially for systems of interest during MSRE. Some of these fluoride systems were more recently updated and verified [222].
- ***Melting temperature.*** A lower fuel salt melting point is beneficial to MSR concepts because it allows for a lower reactor operating temperature and decreases the risk of salt component precipitation. Changes in fuel salt composition (i.e., the build-up of fission products) can affect the overall fuel salt melting temperature [223], and thus, the effect of fuel salt composition on melting point must be adequately modeled.
- ***Actinide solubility.*** Fuel salts must dissolve enough fissile material to be critical but avoid actinide precipitation, which may lead to particulates circulating in the fuel salt loop, the blockage of flow, and a violation of the reactivity control fundamental safety function. Solubility is highly dependent on salt redox potential, temperature, and the presence of other metals (i.e., lanthanides). It is important to note that actinide solubility is much higher in chloride salts compared to fluoride salts and that the solubility of actinide trifluorides is more limited than actinide tetrafluorides.
- ***Plate out of noble metals.*** Noble metal fission products (e.g., Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, and Te) may plate out onto containment surfaces (e.g., structural alloys and graphite moderator) as evidenced from the MSRE [224]. The plate out of these radionuclides can lead to concentrated areas of decay heat on structures and interfere with the graphite moderator.
- ***Redox potential.*** The speciation and phase partitioning of radionuclides in molten salts can drastically change as a function of redox potential, so the redox potential must be maintained within a narrow operating range [225-227]. The generation of radiolytic oxidants can increase salt redox potential over time, which is typically counteracted with the addition of reducing metallic species to the melt. The corrosion of salt-facing structural materials is controlled by salt redox potential. Thermodynamic, electrochemical, and kinetic models may be integrated to model corrosion, which is discussed in more detail in Section 4.2.4.

Salt-fueled MSR concepts are unique in comparison to other advanced reactor concepts because the majority of radionuclides will be present in a liquid phase (the molten salt). The behavior of radionuclides in molten salt solutions can stray from ideality due to the ionic nature and short-range ordering of molten salts [228, 229]. Thus, an accurate depiction of salt chemistry and

radionuclide speciation in salt-fueled MSRs requires a thermodynamic modeling approach that can capture the non-ideality of the solution phase.

The recommended approach for thermodynamically modeling the solution phase of salt-fueled MSRs is the CALPHAD method [230], which stands for CALculation of PHase Diagrams. In this method, thermodynamic equilibrium at a defined temperature and pressure is specified by the minimum of the Gibbs energy (G) for each phase in the system:

$$G = \sum n^{\varphi} G_m^{\varphi} = \text{minimum} \quad \text{Equation 1}$$

where n^{φ} is the number of moles and G_m^{φ} is the molar Gibbs energy of phase φ . The minimization of G in Equation 1 is constrained by mass balance equations. The molar Gibbs energy of each phase can be written as:

$$G_m = G_m^{\circ} + G_m^{id} + G_m^{xs} \quad \text{Equation 2}$$

where G_m° is the weighted molar Gibbs energies of the phase constituents (the end members), and G_m^{id} is the ideal entropy of mixing term. These first two terms account for ideal mixing, while the Gibbs excess energy term (G_m^{xs}) accounts for the effects of non-ideal mixing. In general, the vapor phase for an MSR system can be treated as an ideal gas mixture, and the G_m^{xs} term can be neglected. In addition, the G_m^{xs} term can be neglected for pure substances and stoichiometric compounds in the solid state. However, accounting for G_m^{xs} is essential for an accurate thermodynamic description of the molten salt solution phase and is usually described by Gibbs excess energy functions.

The foundation of the CALPHAD approach rests on the fact that phase diagrams are derived from the thermodynamic properties of a system. Gibbs excess energy functions are parameterized by optimizing fits of solution models to experimentally obtained data including phase equilibria and enthalpies of mixing for binary and ternary systems. These thermodynamic descriptions of binary and ternary systems in combination with standard interpolation methods can then be used to describe higher order systems. Once the Gibbs excess energy functions are obtained and the full Gibbs free energy of the system is accounted for, the system can be thermodynamically modeled with accuracy. The *Molten Salt Thermodynamic Database* or *MSTDB* is a collection of thermodynamic models for molten salts formulated according to the CALPHAD approach that is currently under development and intended for public release in the future [51].

Of the numerous solution models that have been developed to represent Gibbs excess energy functions, the modified quasichemical model (MQM) and the two-sublattice ionic liquid model (TSLM) are used most frequently to model molten salts [231]. Of the two, the TSLM is more convenient to use and can represent the Gibbs energy of ionic melts well in many cases. However, the MQM better describes the configurational entropy by accounting for first-nearest-neighbor and second-nearest-neighbor short-range ordering in solutions and is the recommended solution model for molten salts [231]. Few commercial software (*FactSage* and *Thermo-Calc*) and one open-source software (*Thermochimica*) currently have the capability of implementing the MQM for molten salt solution modeling (Table 2-1; Section 2.3). These molten salt solution models can be integrated with other physics models to predict vapor phase transport, nucleation, precipitation, plate out, and corrosion of salt-facing structural materials.

Most chemical compositions of molten salts of interest for nuclear reactor applications experience a negative deviation from ideality in terms of Gibbs excess energies due to the mixing of salt components. Thermodynamically, this is due to negative heats of solution and positive excess entropies of mixing for the component in a mixture [232]. The deviation is best represented by the difference in the charge-to-radius ratio between the cations in the ionic liquid. Physically, the reduced heat of solution can be explained by noting that liquid lattices of alternating, differently sized cations exhibit less electrostatic repulsion between nearest neighbor cations than would cations of the same charge and radius. Additionally, the excess entropy of mixing can be represented by a distorted lattice of cations of different charge and size, as well as a distorted anion lattice, to accommodate the unique configuration of cations. This entropy contribution can be especially significant for cations of differing charge as complex ion formation has an especially distorting effect on the anion configuration in the liquid lattice. These two thermodynamic effects lead to negative Gibbs excess energies and result in reduced melting temperatures and decreased vapor pressures when compared to that of the pure component [233, 234].

Kalilainen et al. recently modeled the evaporation of radionuclides from a LiF-ThF₄-UF₄ fuel salt and emphasized the importance of considering non-ideal mixing to accurately reflect radionuclide retention [62]. Specifically, the authors calculated the mass of Cs, I, U, and Th species released into the vapor phase as a function of time after a simulated salt spill onto the floor of a confinement building by coupling the *Gibbs Energy Minimization Software (GEMS)* with the severe accident code *MELCOR*. The salt solution was modeled as a mixture of binary fluoride and iodide systems (e.g., CsF-CsI, LiF-ThF₄, LiF-UF₄) using the Redlich-Kister solution model. This model employs a polynomial expansion to account for the excess Gibbs energy of mixing for a binary system:

$$G_{ij}^{xs} = RTx_i x_j \left[a_0 + a_1(x_i - x_j) + a_2(x_i - x_j)^2 + \dots \right] \quad \text{Equation 3}$$

where a_0 , a_1 , and a_2 are dimensionless fitting parameters and x_i and x_j are the mole fraction of species i and j in the binary system, respectively. Ten binary systems (e.g., LiF-ThF₄, LiF-CsF, CsI-LiI) were considered in the solution model and a G^{xs} function was generated for each by fits to empirical thermodynamic information.

The retention of major radionuclide species (CsI, CsF, LiI, ThF₄, UF₃, and UF₄) in the molten salt significantly increased for the simulation that included Gibbs excess energy functions in comparison to the simulation that ran with pure species vapor pressures [62]. In addition, the inclusion of Gibbs excess energy functions in the simulation affected the timing of release for a few radionuclide species (e.g., UF₄) due to the effect on melting temperature.

The salt spill simulation by Kalilainen et al. is one of the first MST trial calculations for an MSR bounding accident [62]. However, the coupling of the Gibbs free energy minimizer *GEMS* with *MELCOR* was done manually, which is not ideal for source term analyses that may require many simulations. Currently, only one Gibbs energy minimizer code - *Thermochimica* - has the capability of directly integrating with other source term codes. *Thermochimica* is an open source software that was developed by Piro et al. with the goal of efficiently coupling to multi-physics codes in mind [73]. It has an enhanced minimization algorithm that increases computing efficiency by using a leveling procedure to estimate the initial phase compositions prior to a

Gibbs energy minimization. This two-step design eliminates unnecessary iterations in the minimization routine by starting much closer to the final result.

FHR Considerations

The thermodynamic modeling approach to determine the speciation and phase distribution of radionuclides for FHRs will likely resemble the MSR approach. Although the molten salt coolant is a major barrier to radionuclide release in both approaches, source term assessments may differ between FHRs and MSRs due to a few unique chemical characteristics of FHR molten salts:

- ***Much lower concentrations of radionuclides in the molten salt coolant.*** The TRISO fuel will retain the majority of radionuclides in an FHR, with only trace amounts present in the coolant salt. The release of fission products from TRISO fuel during normal operation or accident conditions has been extensively discussed in the literature [11, 107, 220], and was discussed in Sections 3.2.2 and 4.2.1.2 for HTGRs and FHRs, respectively. The key fission products in terms of release potential from TRISO fuel have been identified as Ag, Cs, Ce, Eu, I, Kr, Pd, Sr, and Xe [220]. Additionally, many impurities will be present in the coolant salt in ppm levels; experimental measurements for prototypical coolant salt samples have been reported [235].
- ***Molten salt coolant in contact with SiC in fuel matrices.*** Depending on the core design, FHR coolant salt will be in direct contact with SiC layers in certain regions of the fuel matrix holding the TRISO fuel. These carbon-based materials are generally inert towards fluoride salts but can corrode when in contact with metal impurities (e.g., Ni and Cr) that may be present in the coolant salt as well as oxygen [236, 237]. In addition, the molten salt structure at the solid-liquid interface may differ from the bulk. Considering the potentially larger surface area-to-volume ratio of FHRs for carbon-molten salt, surface interactions could affect the overall chemical behavior of the salt. Additionally, SiC has been shown to corrode in the presence of oxygen impurities and higher temperatures, entering the melt as a silicon fluoride complex [236]. Thus, the chemical behavior of FHR molten salts may differ from MSR salts due to interactions with the SiC surface.
- ***Chemical composition that lacks significant redox buffer.*** Historically, the control of redox potential for molten fuel salts was achieved by maintaining the ratio of U^{4+}/U^{3+} [225]. Metal fluoride salts that have two valence states close in energy (e.g., UF_3 and UF_4) can act as a redox buffer in alkali and alkaline earth metal fluoride salts and were used to control redox potential in the MSRE fuel salt [238]. FHR coolant salts lack these metal fluorides at a high enough concentration to act as a significant redox buffer, and thus, controlling the redox potential of FHR coolant salts can be more challenging than MSR fuel salts. Slight changes in the environment to which the FHR coolant salt is exposed during a transient (i.e., exposure to O_2) could lead to large changes in salt redox potential and hence radionuclide behavior. Similar to MSRs, tritium production will have a large impact on redox potential with FHRs [236] and so tritium modeling capability is important [220], as will be discussed further in Section 4.2.7.

4.2.3 Bubble Transport

The transport of gaseous species in molten salts is important for source term modeling, as many radionuclides generated during operation are volatile at some point in their lifetime, whether due to radioactive decay to a different chemical species or reaction to a more volatile chemical form. For example, the noble gases play important roles that both negatively and positively impact the operation of the reactor, not only as fission products contributing to dose and acting as neutron poisons affecting reactivity, but also acting as a sweep gas which may be used to help clean the salt. Therefore, a better understanding of the solubility, diffusivity, and bubble agglomeration of gaseous species is needed to accurately and mechanistically model the reactor's source term.

The following is not meant to be an exhaustive list of volatile components in a salt-fueled or salt-cooled MSR, but rather to showcase the wide range of possible chemicals that need to be better understood for source term models. It should also be reiterated here that many fission products will change chemical class via beta decay during their lifetime in the reactor, as summarized in Figure 4-6. Depending on system parameters and the materials used, volatile classes include:

- the noble gases (Kr, Xe)
- some noble metal fluorides (Ru, Mo, Te, Nb), typically penta- or hexafluorides, as well as uranium hexafluoride, when there is an excess of free fluorine
- some transition metal fluorides (Sn, Sb)
- some alkali and alkaline earth metals and/or their fluorides
- silicon fluoride complexes
- many of the above metals with other fission product halides (Br, I), and/or associated with each other as complex ions

Some experimental work has been published on the solubility of noble gases in molten fluoride mixtures [239-241]. The solubilities of the gases obey Henry's Law and increase with temperature and with decreasing molecular weight of the gas. This is opposite in the trend exhibited by HF [242] and BF₃ [243] in similar salt systems, where solubility decreases with temperature. Although it should be noted that the solubilities for these gaseous fluorides are several orders of magnitude higher than that seen for the noble gases, owing to stronger chemical interactions between the similar ionic fluoride compounds. Therefore, most of the noble gas inventory is still expected to transport out of the salt relatively easily, while only some amount of gaseous fluorides is probably expected to transfer out with the sweep gas. It was reported that at least 95% of the xenon produced was evolved from the fuel salt during the Aircraft Reactor Experiment [240].

Many have studied the effects of the physical properties of molten salts on gas holdup, bubble size, and liquid phase mass transfer coefficients, particularly with chloride and carbonate salts [244-246]. Correlations for mass transfer coefficients of these systems were empirically determined based on dimensionless numbers. Rates of gas absorption in salts with and without chemical reactions were analyzed, providing a set of experimental data for which future numerical models can be validated against. For example, a model for computing noble gas migration in the MSRE was developed [247], and, as mentioned, it was later improved [152]. During MSRE, a brief review and discussion of the possible phenomena of bubbles, drops, and entrainment in molten salts was also reported [248].

Recently, Bajpai et al. developed a multi-physics model in *COMSOL* for reactivity effects of inert bubbles in the MSFR, which is based on a simplified two phase flow coupled to heat transport and neutron transport equations, including delayed neutron drift [249]. Although the model makes several assumptions, the effort is indicative of the possibilities for modeling radionuclide transport in molten salt flows, specifically bubble transport, a very important phenomena in source term modeling. Because certain key radionuclides are expected to stay gaseous from the time they are born as fission products in the salt until they travel to the cover gas system, the ability to effectively simulate bubble agglomeration and transport in the salt is a necessary capability in mechanistic source term modeling.

4.2.4 Corrosion and Deposition Modeling

There is considerable literature on the effect of impurities on corrosion mechanisms for molten salts [250, 251], including fluoride [225-227] and chloride salt systems [252]. Therefore, a discussion on the underlying theory of corrosion is omitted for brevity. It should be noted that many radionuclides formed in the salt via fission or in structural materials via neutron activation may transfer to the other phase. Activation products of the structural materials may corrode into the salt, often becoming salt soluble fluorides. As an example, Table 4-4 displays the concentrations of Cr, Fe, and Ni in many samples taken during the processing of MSRE flush and fuel salts [253]. Activation and fission products in the salt may be reduced and/or deposit on structures such as the heat exchanger tubing or graphite. The permeation of tritium and certain fission products into porous graphite was recently studied [254], but it is also necessary to consider other physical and electrochemical processes relevant to radionuclide deposition in an MSR. Towards the end of the Aircraft Nuclear Propulsion (ANP) Program at ORNL, a thermodynamic interpretation of equilibrium data on the corrosion of iron and chromium by molten salt fuels was published [255]. An analytical and experimental discussion of chromium migration in thermal convection loops was also reported, noting the removal of Cr from alloy and the time- and temperature-dependent mass transfer between hot and cold zones [256]. Further detail and discussion on corrosion behavior relevant to modeling during MSRE can be found in literature [257].

Kedl published a report on the migration of noble metals in MSRE [219], including the development of an analytical model that is based on generation, decay, and deposition on various materials such as graphite, heat exchanger, liquid-gas interfaces. After comparing to experimental data, they conclude that the analytical model, which is only based on simple mass transfer laws, shows that noble metals do migrate and deposit on the heat exchanger. Additionally, comparison of the model to core surveillance samples, fuel salt samples, and gas samples reinforces the conclusion that noble metals and reduced corrosion products in the salt flow adhere to liquid-gas interfaces and remain there with a high degree of stability. Mass transport is dominantly a function of elemental concentration in the salt, although second order effects must also be considered for certain elements, e.g., chemical interactions between Nb and graphite. More recently, noble metal mass transport in MSRs was modeled within the *VERA-CS* suite of reactor physics codes [258].

Recommendations from work during MSRE include constructing a salt loop with tracers to better understand the various behaviors of noble metals including: [219]

- Deposition on solid surfaces
- Deposition on liquid-gas interfaces
- Stabilizing effects of noble metals on bubble and foam interfaces
- Effects of the noble metal chemical species
- Effects of the chemical state of the salt
- Various aspects of circulating bubble mechanics when noble metals are attached to their surfaces
- Prototype testing of bubble sensitive components proposed for a specific MSR

Table 4-4: Structural Metal Fluoride Concentrations in MSRE Flush and Fuel Salts [253]

	Concentration (ppm)		
	Cr $\pm$ 10	Fe $\pm$ 40	Ni $\pm$ 15
Flush salt ^a			
In reactor system	76	150	52
Before fluorination	104	—	—
After fluorination	133	210	516
After 10.8 hr of H ₂ sparging	No sample taken		
After 604 g of Zr and 9 hr of H ₂ sparging	100 ^b	174 ^b	50 ^b
After 1074 g of Zr and 25 hr of H ₂ sparging	No sample taken		
After filtration	76 ^b	141 ^b	26
Fuel salt ^a			
In reactor system	85	130	60
Before fluorination	170	131	36
After fluorination	420	400	840
After 17.1 hr of H ₂ sparging	420 ^b	430 ^b	^c
After 33.5 hr of H ₂ sparging	420 ^b	400 ^b	520 ^b
After 51.1 hr of H ₂ sparging	460 ^b	380 ^b	180 ^b
After 5000 g of Zr and 24 hr of H ₂ sparging	100 ^b	110 ^b	<10 ^b
After 5100 g of Zr and 32 hr of H ₂ sparging	No sample taken		
After filtration	34	110	60

^aH₂ sparging times are cumulative.^bFiltered sample.^cContaminated sample.

Glover et al. developed a multiphysics model which couples neutronics and thermal fluid dynamics to estimate conjugate heat transfer and freezing of the salt on the walls of the MSFR [259]. This can be useful in source term modeling for MSRs because many radionuclides are salt-soluble and will transport with the salt, whether flowing as a liquid or freezing on walls of the reactor as a solid. Neutronics is solved by the nodal diffusion code *DYN3D-MG* [260] with *Serpent*-supplied cross sections, and the open source *Code_Saturne* is used for CFD [261].

Mahmoud et al. performed direct numerical simulations of turbulent nanofluid flow and heat transfer for high Prandtl number fluids and cold and hot walls [262]. They used the open source *Nek5000* due to its efficient parallelization capabilities and validation history, and they were able to model nanoparticle dynamics within the flow due to the effects of wall temperature. This could provide very important information in understanding temperature-based wall deposition of particulates.

Argonne is developing a multiphysics solver for molten salt chemistry and corrosion [263]. It is currently written using the *OpenFOAM* framework, and a *Nek5000* version is also under development. The solver accounts for the combined fluid flow, mass transfer, and chemical/electrochemical reactions occurring in molten salt flow systems. The solver has been applied to a wide variety of cases, including thermal convection flow loops, and validation of the solver is underway. Potential applications include corrosion modeling, mass accountancy, and source term simulations. Figure 4-8 displays an overview of its use in simulating corrosion in a thermal convection loop.

Figure 4-8: Overview and results from early demonstration simulations of corrosion in a thermal convection loop using Argonne's multiphysics solver [263]

4.2.5 Salt Release

Although the fuel salt itself exists as the first barrier to release for radionuclides, the system's primary structural boundary enclosing the fuel salt from the containment cell represents a barrier equally as important. Therefore, a breach of this boundary (i.e., vessel wall, pipe wall, etc.), could result in an event of varying severity and consequence dependent on the size and location of the breach. Although event frequency is dependent on the MSR design (loop, integral/pool type, modular), the consensus among the ORNL MSR LBE workshop participants is that this LBE can probably be classified as a design DBE or lower [126]. For many MSR designs and most locations of potential boundary breaches, the salt will release into a reactor containment cell designed with this scenario in mind. Consequences will be limited based on the implementation of defense-in-depth and the functional containment approach. The containment cell represents an important barrier for release, therefore if the cell has also been breached, is open, or contains air

or water, the consequences of the salt spill will be more severe than expected. For this reason, the consensus of the workshop is that this severity can be anywhere from “low to high” depending on other event sequence safety function successes and failures. Salt spills were also classified as important IEs for FHRs in one of a series of whitepapers summarizing workshops [123].

It is generally accepted that the ability to model salt releases is a necessary capability in any MST strategy for all reactor types with a flowing molten salt. Industry has begun an effort to model salt releases, such as those involving spraying molten salt occurring due to pipe breaks [189]. It should also be noted here that most types of salt releases will result in aerosol formation, which is further discussed in Section 4.2.6. As mentioned previously, Kalilainen et al. have shown how evaporation of a spilled salt can be modeled with modifications to existing models in MELCOR [62]. This is coupled with separate codes for radionuclide inventory calculation and thermochemical modeling.

Due to the lack of experimental data in molten salt spills, and the uncertainties associated with numerical models of such unique liquid systems, a relatively large-scale salt pour/spill test at Argonne is currently being planned by the DOE MSR Campaign to better understand the associated phenomena and inform computational modeling tools. Examples of information that can be elucidated from such a test include the effect of the pour rate and temperature on the spread behavior and morphology of the spill, as well as thermal hydraulic properties. As a reference point, considerable work at Argonne has been completed in computationally modeling the spreading behavior [264] and cooling behavior [265] of corium melts for LWR accident scenarios based on corresponding experimental tests. A large-scale molten salt spill test could help close many knowledge gaps associated with radionuclide releases which might occur during this important LBE.

One important phenomenon that needs further investigation is the volatilization of uranium which occurs as a uranium-bearing fluoride salt cools due to excess fluorine forming, and oxidizing uranium tetrafluoride to the highly volatile hexafluoride. Nearly 25 years after the MSRE was shutdown, a sample was taken from the off-gas system connected to the cover gas of the drain tanks holding fuel salt [266]. Nearly half of this sample was found to be fluorine gas and a considerable fraction was found to be UF_6 gas. Both of which were unexpected, demonstrating the necessity of experimental validation of computational chemistry models. The ease with which uranium hexafluoride and other volatile fluoride compounds can form as fluoride salt systems cool provides further motivation for an integral molten salt spill test to inform modeling tools.

4.2.6 *Aerosol Modeling*

An overview of aerosol phenomena and modeling for LWRs was discussed in Section 2.4 and has been extensively covered in literature. But very little on aerosol phenomena specific to high temperature liquid salts has been published in literature. During a salt spill, aerosolization of the molten salt may occur, but salt aerosols may also form within the reactor due to turbulent flows interacting with the sweep gas, followed by transport to the cover gas and eventually to the off-gas system. Radionuclides can be easily entrained in these salt mists/aerosols, as well as in carbonaceous particles in aerosols such as graphite dust. Because many radionuclides are salt soluble, it is important to consider the impact on the source term that aerosol generation will have inside and outside the reactor. Additionally, some fraction of insoluble noble metals can enter the

gas phase (and off-gas system) via aerosol-forming bubble bursting, supported by the hypothesis that noble metals accumulate at the liquid-gas interface (See Section 4.2.4).

The most widespread mechanism for aerosol generation is evaporation of a liquid followed by condensation into the gas phase, although this is typically accomplished with large variations in the temperatures of the phases. Therefore, condensation aerosols will likely be formed during a severe accident or breach of molten salt beyond the reactor system structure. Buckle and Ubbelohde published considerable research on aerosol formation of molten alkali halides using a cloud chamber technique [267-269]. Rapidly increasing the temperature of a liquid droplet of a pure molten salt will form a supersaturated vapor phase around it, which is followed by fine particles of the salt condensing into the gas phase. This method of aerosol formation may not occur in all regions of the primary system during normal operation, but it should be considered during a severe accident or salt spill. Additionally, these experiments provide a better understanding of aerosol formation rates, particle size distributions, and temperature dependencies. Later, Buckle published experimental data and theory on the nucleation and growth of cesium iodide salt aerosols, specifically as it relates to the application of LWR source term [270]. This is an important reminder that potential CsI formation and release is not unique to MSRs, so there is already considerable literature on CsI characterization in general.

During the normal operation of an MSR, a bigger contribution to aerosol formation might be turbulent flow of the molten salt throughout the reactor system and breakup into the cover gas. Although very little research has been completed in the area of mechanical aerosol formation of molten salts, the Pacific Northwest National Laboratory (PNNL) has published comprehensive reports on large-scale spray release tests for various types of aqueous solution [271, 272]. Tests were performed to quantify the release fraction and aerosol generation rate for a range of orifice sizes, shapes, fluids, and spray pressures. It is left to the modeler to assess the applicability of the data generated for the simulants used in these tests to molten salts. This mechanical aerosol generation can also occur during a severe accident. Ginsberg published a composite flow-regime-dependent model for aerosol generation by gas-flow-induced mechanical breakup of molten corium during core/concrete interactions for an LWR during a meltdown scenario [273]. The correlations and theory used here may be applicable to a pool of molten salt with bubble flow inducing aerosol generation at the surface.

Williams developed a system which aerosolizes molten salts for analytical applications, utilizing laser-induced breakdown spectroscopy (LIBS) to measure elemental concentration in the aerosols [274-276]. A specific nebulizer design is used to generate aerosols of LiCl-KCl salt systems containing Ce and U. The limit of detection was found to be on the order of hundreds of ppm Ce or U for these experiments, indicating its potential use for estimating compositions of aerosols which are generated from salts of radionuclide concentration 1 wt% or more. These studies can be used to better understand salt aerosol generation via the instrument's nebulizer, as well as using LIBS for analyzing salt aerosols generated via other techniques.

Recently, Kairos Power submitted a Mechanistic Source Term Topical Report for their FHR, which includes methodology for characterizing aerosols [189]. The methodology considers mechanical aerosol generation during two scenarios: pipe breaks and transients in the pebble handling and storage system (PHSS). During a pipe break, aerosols can form from the breakup of the jet of liquid, as well as due to salt splashing on a surface. They consider aerosols which

contain graphite dust created from fuel pebble handling, which is also a very important source term phenomenon for HTGRs. Depending on the graphitic materials used in the reactor and the core/flow design, dust-containing aerosols should be considered for most MSRs, not just FHRs. Additionally, Kairos Power plans to use the radioactive material transport code *RADTRAD* to determine exclusion area boundary (EAB) dose estimates based on aerosol formation, transport, and deposition, also accounting for radioactive decay. As will be discussed further in Section 6, *RADTRAD* models transport between control volumes which can represent buildings or smaller spaces such as cover gas regions.

4.2.7 Tritium Modeling

Compared to other aspects of source term analysis, considerable work has been done in creating transport models for tritium in MSRs and FHRs due to potentially high generation rates from neutron interactions with Li-6. Additionally, because certain chemical forms of tritium are highly permeable, releases to the environment and resulting contributions to off-site dose may be important. Tritium generation and transport phenomena are reviewed here, while tritium-specific chemistry phenomena when released to the environment is discussed in Section 6.

A report on tritium assessment in molten salt reactors was recently published [277], but a brief summary on the current state of codes that have been developed for tritium speciation, transport, and other phenomena relevant to its behavior in MSRs is provided here. It is also worth noting that estimating tritium production can be accomplished with certain tools outlined in Sections 4.2.1.1 and 4.2.1.2, corresponding to the modeling of the radionuclide inventory in MSRs and FHRs, respectively. Therefore, this subsection focuses on modeling tritium speciation and transport behavior in the system. Generally speaking, the main pathways for the tritium source term in MSRs and FHRs include dissolution in the salt as TF, permeation through metal as T₂/HT, adsorption in graphite as either chemical form, or a design-dependent controlled removal.

The *Tritium Diffusion Evolution and Transport* code (*TRIDENT*) was developed by Stempien for the FHR as an integrated tritium transport model [220, 278]. The model incorporates tritium production, speciation, solubility, adsorption on graphite, permeation through metal, and corrosion modeling. The model considers tritium production from neutron absorption reactions of Li-6 and Li-7, as well as production of Li-6 from transmutation of Be-9. The model can conceivably be reconfigured to factor in other tritium producing reactions given the one-group cross sections for that reaction. The speciation of tritium to either TF or T₂ is calculated based on the redox potential of the salt, which is related to other redox buffers that can be used in the reactor design and factored into the model, such as the ratio of a uranium-bearing fuel salt U⁴⁺/U³⁺. The ability to model changing redox chemistry allows for a better understanding of the distribution of tritium in the oxidizing TF form vs the gaseous T₂ form, which is more problematic from a source term perspective due to its permeability in metal. Both forms of tritium can be adsorbed onto graphite, which can be modeled by *TRIDENT*. The model estimates the amount of tritium that permeates through metal based on diffusion coefficients. Most notably, metal can be found as the structural material in the heat exchanger, representing an important pathway to release to either a secondary salt or potentially a gaseous coolant, depending on the design. The model also has corrosion modeling capabilities built-in, as the effect of TF on corrosion as well as the production of T₂/HT and metal fluorides from corrosion are intimately

related in modeling the redox chemistry of the coolant. Finally, tritium removal systems can be factored into the model's mass balance.

TRIDENT was designed specifically for tritium behavior in the coolant salt of the solid-fueled FHR but can be used for salt-fueled molten salt reactors also. The validity of *TRIDENT* for MSRs of the thermal spectrum and fluoride salt variety is evidenced by validation studies that were completed for certain parts of the model against experimental data from MSRE or elsewhere. This includes comparing the model against experimental data of hydrogen gas diffusion through two fluoride salts. Additionally, the model was used to estimate corrosion in actinide-bearing fluoride salt loops that were run in support of the MSBR. Modifications should be made to reconfigure the code to fast spectrum and chloride salt MSRs, as certain aspects of the model rely on assumptions specific to FHR or fluoride salts containing a redox buffer of UF_4/UF_3 .

Lam continued validation and development of *TRIDENT*, updating the code with data and models relevant to tritium capture via adsorption by carbon materials [279, 280]. They considered adsorption on materials such as activated carbons, graphene nanoplatelets, and nuclear graphite materials. Comprehensive experimental data is provided, comparing results with respect to the differing adsorptive properties of the materials. *TRIDENT* is used to model an FHR with such a high-performance adsorbent, and they note the impact a potential tritium adsorption column in a reactor system's design will have on reducing the source term during a severe accident.

The *Transient Simulation Framework of Reconfigurable Models (TRANSFORM)* is a *Modelica*-based simulation environment that has been developed at ORNL for advanced reactor modeling, and has been used to model tritium diffusion [281]. The researchers derive analytical solutions for tritium transport and diffusion in vacuum permeators. They consider two different materials for the vacuum permeator, which is used to capture or allow transport of tritium in an advanced energy system such as a fission or fusion reactor. The permeators were modeled in the *TRANSFORM* framework using components in the library and the steady-state results were compared against the analytical solutions with excellent agreement. This tracking of a single substance demonstrates the capability of *TRANSFORM* for use in other aspects of tritium transport in MSRs.

The *SAM* code is currently being developed for tritium transport modeling based on its passive scalar mass transport model which can be used to track any number of species in the fluid flow in conjunction with its capabilities in whole plant transient analysis [33]. Using the mass transport model, preliminary capabilities have been implemented in *SAM* for tritium production within the core, transport, and diffusion through the primary loop in the FHR. The tritium transport modeling capability in *SAM* will be further enhanced under the support of a recent DOE-NE industry FOA award led by Kairos Power [282].

Various modeling tools have been developed for specific tritium mitigation and control systems. Wu developed a tritium mitigation and control system for a representative FHR heat exchanger, discussing much of the necessary tritium transport theory including diffusion, mass transport, and dissolution [283]. They use the *COMSOL* Multiphysics platform to simulate the crossflow of the tritium removal facility, which is discussed in detail. They also analyze potential materials for use as a tritium permeation barrier coating, considering tritium permeability, diffusivity, and solubility. Similarly, Zhang et al. designed and modeled a double-walled heat exchanger for use

in an FHR or MSR, coupling a tritium transport model to a heat transport model [284]. Although an experimental setup has never been completed before, it is worth noting the theoretical and computational models that have been utilized in modeling these important auxiliary systems for both FHRs and MSRs.

As mentioned previously, tritium behavior in molten salt-cooled nuclear reactor systems is relatively well documented compared to that of other radionuclides. The reader is directed to the following studies which summarize some of these important characteristics of tritium, including tritium control and capture [285], as well as the effect of impurities (such as tritium) on the primary coolant salt of FHRs [226].

Internationally, several tritium transport codes for MSR variants have recently been developed in China. Many of the models utilized in these codes are documented in the open literature. The *Tritium Transport Characteristics Analysis* code (*TAPAS*) was developed by researchers at Xi'an Jiaotong University in China as part of the development of the prismatic-core transportable FHR (TFHR) design [286]. The same researchers also used *TAPAS* for the solid-fueled pebble bed Thorium-based Molten Salt Reactor (TMSR-SF), showing the integrated theoretical model can be used for various fluoride salt-cooled, solid-fueled designs [287]. They provide details on the models built into the code as well as results from the code on the effect of Li-7 enrichment on tritium production and the effect of redox potential on T₂ permeation and Cr corrosion [286].

The *TAPAS* code was updated and used for transient analysis of the FHR for an unprotected reactivity insertion accident (URIA) and an unprotected overcooling accident (UOCA) [288]. For a URIA, the varying tritium concentration in the salt is plotted alongside the amount of Cr corroded off and deposited onto the structural material. An overcooling accident will cause a response in tritium transport characteristics much different of that seen for the URIA. For example, a decrease in temperature affects the mass transfer coefficient in salt and diffusion coefficient in metal of TF and Cr, respectively, in addition to speciation and adsorption. Overall, the results show that both transients have a minor effect on graphite adsorption rate, an opposite effect on tritium production rate (increase in tritium production for UOCA), and a considerable decrease in T₂ permeation for UOCA.

The *Tritium Transport Analysis Code* (*TTAC*) was developed to model tritium speciation, transport, and distribution in MSRs, specifically the Thorium-based Molten Salt Reactor (TMSR) [289]. There are two designs of the TMSR: a salt-cooled, solid fuel reactor similar to FHR, and a salt-fueled molten salt reactor, but *TTAC* is applicable to both due to similarities in tritium modeling characteristics in the coolant for both. Built using *MATLAB/Simulink*, the code was also validated against MSRE data. The code consists of mass balances for both hydrogen and tritium, tritium production via transmutation of coolant nuclei as well as ternary fission, tritium adsorption on core graphite, tritium permeation through metal, the removal of tritium via a purification term, and chemical speciation of tritium.

Finally, the *Tritium Migration Analysis Program* (*TMAP*) code, which was developed originally for fusion energy systems [290], can also be used for MSR applications with minor modifications. Recent developments have shown that it can be integrated with the severe accident modeling code *MELCOR*, which provides additional capabilities in source term modeling [291].

4.2.8 Salt Processing System

If a salt-fueled MSR developer chooses to design a fuel processing system to be used in conjunction with the reactor, then a corresponding source term analysis must be completed for the auxiliary system. The same applies for a salt processing system, or salt clean-up system, that is utilized in a salt-cooled MSR, such as the FHR. A brief discussion is given of studies completed and models developed in support of such auxiliary systems.

Because there is very little public information on these design-dependent chemical systems for the reactors that are currently under development by industry, the fuel processing system of the single-fluid MSBR was previously used as an example when analyzing for important phenomena and functional requirements for its simulation [2]. There is considerable information in open literature on this system (Figure 4-9) which relies on successive steps of fluorination (to volatilize uranium) and reductive extraction (for other metal separations) [292-294]. A successful source term model should provide an understanding of the distribution coefficients and separation factors at each stage, allowing for material tracking not only relevant to MST development, but also safeguards concerns and reactivity control. For example, the buildup of plutonium needs to be analyzed and accounted for, as it is a radiotoxicity hazard as well as a proliferation concern, even at the relatively low concentrations of around 100-200 ppm found during MSRE [257].

Figure 4-9: Conceptual Flow Diagram for Processing a Single-Fluid MSBR by the Fluorination-Reductive Extraction-Metal Transfer Process [292]

As discussed previously, in their analysis of the MSBR fuel cycle, Nuttin et al. built a model of the core in *MCNP* and utilized an in-house code to iteratively calculate depletion and actinide inventory [199]. Although they use assumptions about the removal and separation efficiencies of the chemical processing system, they provide discussion relevant to source term inventory tracking. They provide comparison calculations of varying removal rates and the resulting effect

on breeding ratio and core neutronics. An updated version of the coupled codes (*MCNP-REM*) was used by the same research group to assess various thorium fuel cycle characteristics such as actinide inventory for the MSFR [202, 204].

Heuer et al. used the *MCNP-REM* tool to couple neutronics with the reprocessing system, accounting for isotope removal due to chemical extraction using estimated elemental extraction efficiencies [201]. They correctly point out that a successful model of the reprocessing system must account for the competition between decay and chemical extraction at each step. They also provide preliminary results of residual heat and neutron flux for materials in various portions of the system. Doligez et al. continued work on the reprocessing system numerical model, which that takes various chemical separations parameters as input and provides time-dependent isotope removal from the reactor as well as tracks radionuclide inventories at the various locations [203]. The additional work in this study includes a modeling of the sweep gas bubbling in the reactor core, lanthanide extraction via reductive extraction, and a discussion on the impact of reprocessing on neutronic, thermal, and chemical properties of the core. The authors address the important knowledge gap associated with having accurate partition coefficients for these separation processes by completing an uncertainty analysis on the effect they have on extraction efficiency. It is worth reiterating the importance of a tool that characterizes and models the reprocessing system and is also coupled to a neutronics tool as well as a sensitivity analysis tool.

Rykhlevskii et al. developed the open source tool *SaltProc* which expands the capabilities of *Serpent-2* for analyzing fuel cycle performance of MSRs [295, 296]. It allows for generic geometry modeling, multi-flow capabilities, time-dependent feed and removal, and user-specified separation efficiencies. This batch-wise tool was developed to account for the effect of the processing system on the fuel equilibrium composition and breeding, as well as operational safety. They provide an overview of the chemical processing system for the single-fluid MSBR and analyze the MSBR's fuel cycle performance while determining the necessary thorium feed rate. The tool can conceivably be used for high-level source term modeling of the processing system while still accounting for radionuclide compositions in the primary system.

Many other fuel cycle analyses have been completed for various types of MSRs with on-line reprocessing. A custom *SCALE* sequence has been used for MOSART [196], *Serpent-2* has been used for MSFR [209], a custom *MCNP-CINDER90* sequence has been used for MSBR [215, 216], and *ChemTriton-ORIGEN* has been used with the systems dynamics fuel cycles tool *ORION* for a fast spectrum MSR [297]. Similarly, recent updates to *ORIGEN* allow for addition and removal streams in the point depletion solver [53]. Although these studies provide good discussion on actinide inventories in MSRs over time and especially at equilibrium, the modeling of radionuclide addition or removal is usually accomplished with decay constants in the transport equations representing constant transfer rates. A deeper understanding of actinide and fission product transport in molten salt separations processes is needed for modeling source term in these design-specific auxiliary systems. For example, Figure 4-10 displays the calculated volatilization (e.g., separation from the salt) of some fission products and uranium dependent on the chosen fuel salt fluorination process [253]. Theoretical data such as this needs to be experimentally validated in order to influence computational models. A review of MSR waste and effluent management strategies has been published and provides discussion on all chemical streams, not only those relevant to salt processing systems, but also for the off-gas system and other potential separations technologies [298].

Figure 4-10: Calculated Fission Product Volatilization During MSRE Fuel Salt Fluorination [253]

4.2.9 Off-Gas System

Just as with the salt processing system and other auxiliary reactor systems, the off-gas system will need to be characterized for potential source term releases and material tracking. Some research has been performed in off-gas system behavior and chemistry during MSRE. For example, analysis of particle trap filters in the off-gas line for the MSRE resulted in some preliminary conclusions [257]. Very small concentrations of the non-volatile salt components Be and Zr were found in this filter, indicating the amount of entrained salt mist carried to the filter was negligible. Despite this, it can't be safely assumed that salt aerosols don't necessarily enter the off-gas system. But it can be hypothesized that certain filters or flow geometries can be very effective at stripping aerosols from the off-gas feed, which is probably why very little was found in the filter further downstream of the off-gas line. Similarly, many solid fission products were found to be removed by mesh filters earlier on in the off-gas lines. Additionally, the high activities of Ba and Sr (which are the daughters of Kr and Xe) found in the particle filter indicate that a large fraction

of these solid elements still travel down the off-gas lines even after decay from their noble gas precursors.

More recently, McFarlane et al. have continued research and development in potential off-gas system technologies for the separation, sequestration, and characterization of volatile components in molten salt reactors [299, 300]. An important consideration for off-gas systems is the decay of fission products to often very different chemical classes, as discussed previously and better illustrated in Figure 4-6. An MST model of an off-gas system would have to account for the appropriate chemistry at each separation stage in addition to the time-dependent chemistry of the fission product, accounting for decay to a different chemistry either during transport or separations. For example, a fission product can decay from a noble metal to a halide to a noble gas to an alkali metal all within a short period of time, all exhibiting different levels of volatility.

McFarlane et al. provide thorough discussion on relevant fission product chemistry, transport properties needed to model the inventory, and potential accident scenarios allowing release [300]. Therefore, the reader is directed to these reports for a more complete discussion on modeling the chemical and physical phenomena of an off-gas system. An overview of such a system that could be used by a commercial MSR is provided in Figure 4-11.

Figure 4-11: Schematic of a Potential Off-Gas System for a Commercial MSR [299]

4.3 Capability Assessment

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 [301]. This information is used to help inform the importance of the identified gaps noted below. The study 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 those functional needs were salt thermophysical properties, salt constituent volatilities, salt constituent solubilities, mass inventories and corresponding distributions, and chemical interactions. Not surprisingly, it can be seen that many of these inputs and functional needs are chemistry phenomena that are not yet well understood for molten salt systems. These chemical and physical

phenomena that affect mass transport and the chemical interactions of radionuclides, along with fuel depletion, make up the bulk of the source term development.

Considerable MSR system and reactor physics modeling capabilities have been developed, including high fidelity neutronics and computational fluid dynamics, and system-level thermal hydraulics. Dedicated MSR source term modeling and simulation tools are largely immature. Based on the NRC development pathway, the reactor physics suite of codes *SCALE* will be utilized for MSR radionuclide inventory estimation with the integral accident analysis code *MELCOR* being further developed for MSR source term modeling. Salt thermochemistry is very important to MSR source term modeling; computational tools exist, but much of the experimental thermodynamic data necessary for the tools is lacking. Important chemical and physical phenomena in MSR source term modeling include, but are not limited to, bubble transport, aerosols, corrosion, and deposition. A salt spill scenario is potentially a high-priority LBE that will require modeling capabilities for the prospective design. Finally, successful MST strategies will likely also need to assess radionuclide transport throughout all auxiliary systems such as off-gas and salt processing systems.

Based on this evaluation, several key gaps were identified regarding the current capabilities of modeling and simulation tools for salt-fueled and salt-cooled MSRs, described below:

- **Core Neutronics and Fuel Depletion:**
 - There have been many efforts in modeling core neutronics and fuel depletion for salt-fueled MSRs but gaps still exist in the integration of neutronic and depletion solvers for some standard geometries, the level of validation against experimental data (or benchmarks with high-fidelity codes), the impact of delayed neutrons outside the core region, and the ability to conduct uncertainty/sensitivity analyses.
 - For solid-fueled MSRs, neutronics and fuel depletion modeling more closely follows that of HTGRs than salt-fueled MSRs, and is therefore more mature, depending on the fuel geometry
- **Salt Chemistry:**
 - Much research and development in salt thermochemistry is still needed, especially related to the vapor pressures and speciation of fission products as a function of system temperature and pressure. Computational modeling tools exist and are being developed but the underlying thermodynamic data and theory that the tools rely on is still incomplete.
- **Bubble Transport:**
 - Bubble transport experimental work has been completed for some molten salt systems, including noble gases and some volatile fluorides, but a deeper understanding based on experimental work is lacking. Some numerical models have been developed for noble gas migration but there is a gap in modeling other types of volatiles.
- **Corrosion Modeling:**
 - Development has started in corrosion and deposition modeling but a deeper understanding of the chemistry that controls the underlying theory is still needed for many radionuclides, salt systems, and structural materials. A more complete understanding of the electrochemistry of fission products and activated corrosion products is needed in order to validate time-dependent models for electrochemical

reactions coupled with flow modeling. Additionally, further work is needed to elucidate additional phenomena and confirm hypotheses related to the migration of radionuclides such as noble metals.

- **Salt Spill and Aerosol Modeling:**

- Understanding the behavior and impact of a salt spill scenario is a high priority gap, as boundary breaches for fuel-salt represent a high-priority LBE among subject matter experts. A relatively large-scale molten salt pour/spill test is currently being planned to aid in the development of modeling tools.
- Aerosol modeling of molten salts is relatively immature, except for some experimental studies in salt aerosol formation via evaporation and condensation, and studies on supercooling. Fortunately, existing knowledge from testing of mechanically generated aerosols of other materials (e.g., aqueous solutions) is potentially applicable to molten salts.

- **Fuel Processing & Off-Gas Systems:**

- Simulation tools have been developed for radionuclide tracking in on-line fuel processing systems of MSRs from a high-level, including those with coupled neutronic tools. However, very little has been performed in model development from a chemical phenomenon standpoint, evidenced by constant separation efficiencies being used in the aforementioned simulation tools. The largest knowledge gaps associated with these models are related to the lack of experiments that have been completed on the generally immature chemical processing methods. The understanding of the underlying chemistry and chemical separations phenomena may be insufficient to adequately model source term distribution in prospective fuel processing systems.
- At a system-level, the same simulation tools used for fuel processing systems can conceivably be used for off-gas system material tracking with modifications in the separation efficiencies. Also similar to processing systems, knowledge gaps seem to exist in modeling the actual chemistry of off-gas systems, but dedicated work in this area is ongoing. Fortunately, some aspects of off-gas system engineering and design can also be informed by similar technologies in other industries, as gas phase chemistry is not unique to MSRs.

5 Sodium-Cooled Fast Reactors

Compared to other advanced reactor technologies, there has been relatively extensive work performed concerning the development of MST assessments for SFRs. As will be detailed in this section, this is partly a result of the considerable sodium reactor experience in the U.S. and also recent research motivated by substantial U.S. industry interest in SFR development. The focus of the following assessment is metal fuel pool-type SFR designs, which is the variation proposed by U.S. industry.

Licensing Basis Events

At a high level, there are two main types of radionuclide sources in a typical pool-type SFR design: the reactor fuel and the auxiliary systems (such as the primary sodium or cover gas clean-up systems). Therefore, the central LBEs of interest for MST analysis can also be segregated on these lines.

For potential fuel damage sequences, progress in inherent and passive safety measures has generally resulted in the practical elimination of hypothetical core disruptive accidents (HCDAs), which historically included phenomena such as fuel melting, re-criticality, and subsequent core energetics. Such events were theorized to result in large-scale fuel vaporization, extreme vessel loading, and potentially large sodium fires. Instead, the current focus is typically on low-frequency LBEs involving limited core damage. As ref [60] explored, such LBEs could involve partial fuel damage due to localized core blockages, overpower events, or loss of long-term decay heat removal. These events generally do not result in energetic consequences or core relocation, and therefore do not challenge vessel integrity.

For LBEs associated with auxiliary system releases, the central focus is on the potential release of radionuclides contained within filtration systems, decay beds, or accompanying piping. For primary sodium cleanup systems, this could include leaks resulting in sodium fires and volatilization of any contained radionuclides. Typically, such systems are contained within vaults or other structures with containment-like properties to limit the release associated with such potential events.

5.1 System Modeling Capabilities

In the U.S., modeling of SFR reactor transients and the evaluation of potential fuel damage has historically been performed using the *SAS4A/SASSYS-1* code [302] or its predecessors. *SAS4A/SASSYS-1* is a computer code developed by Argonne for thermal, hydraulic, and neutronic analysis of power and flow transients in SFRs and lead-cooled fast reactors (LFRs). In addition to thermohydraulic modeling, *SAS4A/SASSYS-1* includes severe accident models for both oxide and metal fuel³, which predict the timing, location, and magnitude of fuel failure and melting. However, the severe accident models do not currently track fission product migration or release for the purpose of source term modeling. As will be discussed in the following subsection,

³ The metal fuel severe accident models are currently under development, see ref [303].

SAS4A/SASSYS-1 simulations have been used as an input or basis to both SFR and LFR MST analyses.

Within the NRC code development vision and strategy, *SAM* has been proposed as the system analysis tool for SFR DBEs [87]. Several recent studies have utilized *SAM* for the assessment of SFR behavior during both steady-state operation [304] and transients [305], including validation utilizing tests from EBR-II and FFTF [306-308].

Recent efforts have also explored the application of high-fidelity tools for the evaluation of SFR and LFR hot channel factors, which are typically utilized within system codes to address uncertainties related to core performance [45]. These analyses utilized the *SHARP* toolkit, developed under NEAMS, that is comprised of *PROTEUS*, *Nek5000*, and *Diablo*. The derived hot channel factors could aid in the determination of the possibility of core damage, and its magnitude, during transient scenarios modeled with system codes.

5.2 Source Term Modeling Capabilities

In regard to source term modeling, the central LBEs of interest (fuel damage events and auxiliary system releases) share some aspects of radionuclide release and transport phenomena, while differing in other areas. As shown in Figure 5-1, the assessment of potential radionuclide release and transport from core fuel involves a variety of physical and chemical phenomena and multiple radionuclide barriers. Beginning with the generation and migration within the fuel pin, radionuclides released from the fuel would then enter the sodium pool. Through bubble transport or vaporization, radionuclides could enter the cover gas region. From there, leakage pathways could result in transport to the containment volume and eventually to the environment.

In contrast, radionuclide releases from auxiliary systems would likely result in radionuclides entering a vault or similar volume directly, as shown in Figure 5-2 for a primary sodium cleanup system. This could include the release of radioactive noble gases from decay beds or cryogenic distillation system, or a release of liquid sodium containing activated sodium and other radioactive impurities. In general, the main concern is the vaporization or aerosolization of radionuclides, which could then leak to the environment.

In terms of modeling and simulation tools, several integral SFR source term analysis codes have been developed or are currently under development. Recently, Argonne released the *Simplified Radionuclide Transport (SRT)* code, which focuses on radionuclide release and transport for pool-type, metal-fuel SFRs [309]. As shown in Figure 5-3, *SRT* does not perform reactor transient analysis but instead utilizes the output of system-level codes (such as *SAS4A/SASSYS-1* described in the previous subsection) or user-developed reactor conditions as inputs to the model. From there, *SRT* assesses the release of radionuclides from the fuel, the transport of radionuclides through the sodium pool, behavior in the cover gas and containment regions, and finally dispersion offsite. The models used for this analysis will be described in further detail in the following subsections but in general *SRT* offers a mix of simplified and detailed phenomena models with a high level of user flexibility. This framework is used to facilitate the use of the code in sensitivity and uncertainty analyses. The focus of *SRT* is radionuclide release from fuel within the reactor vessel and the code does not currently evaluate non-core releases without modification by the user.

Figure 5-1: Summary Diagram for SFR Radionuclide Retention Phenomena [310]

Figure 5-2: Example Radionuclide Barrier Arrangement for an SFR Sodium Cleanup System [310]

Figure 5-3: SRT SFR Model Code Overview [309]

MELCOR

The DOE funded efforts to enhance *MELCOR*'s ability to model sodium reactors by incorporating models that can be used to simulate containment accidents involving sodium. These models were previously developed for the *CONTAIN/LMR* code [311] and have received validation against experiments and have been used for more than a decade by modelers. Since *CONTAIN/LMR* is no longer actively developed, its models were implemented into *MELCOR*. Implemented models include those for sodium fires (pool fires and spray fires), aerosol interactions and chemical reactions. Sodium has also been added as a working fluid to *MELCOR*. Additionally, a heat pipe model has been incorporated into the software, a key system for many micro-reactors. These models have been incorporated in a managing "NAC" physics package [58].

The proposed NRC evaluation model for sodium fast reactor systems can be seen in Figure 5-4. Within this model, *AMPX* is to be used for cross section generation, *SCALE* is to be used to reactor physics simulations, *MELCOR* is to be used for system behavior and fission product transport within the system, and *MACCS* is to be used for consequence analysis [58].

Key accident phenomena to take into account for SFR accidents and associated reactor events are covered in Table 5-1, which was developed by the NRC as part of its non-LWR code development plan. Of particular note is the necessity for *MELCOR* to implement an SFR-specific core module that would capture the behavior of fuel degradation during postulated events [58].

Figure 5-4: Proposed NRC Evaluation Model for SFRs [58]

Internationally, the *Transport Phenomena of Radionuclides for Accident Consequence Evaluation of Reactor (TRACER)* code was developed in Japan to evaluate in-vessel radionuclide transport during fuel pin failure transients in SFRs [312]. *TRACER* contains models for radionuclide release from the fuel, transport through the sodium, and release to the cover gas region from bubbles or vaporization, with a focus on oxide fuel loop-type SFR designs. Ongoing efforts as part of the Civil Nuclear Energy Research and Development Working Group (CNWG) are currently performing code comparisons between *TRACER* and *SRT*.

In addition, a sodium-specific version of *ASTEC*, *ASTEC-Na*, was developed in Europe for the analysis of severe accidents in SFRs [313]. Point kinetic models have been added for the assessment of unprotected transients [314], in addition to sodium fire models for the evaluation of volume temperature and pressure loading [313].

Table 5-1: Key Accident Progression Phenomena for SFRs [58]

Key Phenomenon	Importance	Existing Capabilities	Modeling Gaps
Liquid Metal to be used as a working fluid	Modeling the liquid metal coolant heat transfer properties is essential in simulating the reactor response to accident conditions	No equation of state libraries already available to MELCOR.	<ul style="list-style-type: none"> Ability to model sodium as the working fluid in some control volumes and water in others will be added (development item M1.7) Addition of Pb and Pb/Bi EOS/Properties (infrastructure developed under development item M1.7)
Fission Product Speciation	Affects the release, vapor pressure, and chemical interactions of fission products.	MELCOR utilizes radionuclide classes organized by chemical similarities that can be easily adapted for reactor application	<ul style="list-style-type: none"> Determination of MELCOR class structures (development item M1.3)
Fission Product Release Model	Determines distribution of fission products between the fuel and fission gas plenum.	MELCOR has a generic release model easily adapted for metallic fuel.	<ul style="list-style-type: none"> Extension of existing modeling for FP release for metallic fuel (development item M1.4)
Fuel degradation model.	Degraded fuel components lead to release of fission products from the fission gas plenum as well as some fuel/clad material.	MELCOR has models for fuel components that can be extended to SFP application	<ul style="list-style-type: none"> Extend MELCOR fuel component to capture melting fuel in fuel matrix Model for cladding failure from eutectic penetration or molten fuel contact Ejection of fuel/sodium from failed rod. (development item M1.2)
Sodium fire modeling	Sodium fires provide a source of heat to the containment and also provide a path for transport of sodium and fission products to the atmosphere.	Sodium pool fire and spray fire models, as well as atmospheric chemistry models have already been added to the code.	<ul style="list-style-type: none"> Addition of a hot gas layer model during sodium fires (development item M1.6)
Sodium concrete interactions	Important source of aerosols and possible combustible gases		<ul style="list-style-type: none"> Add sodium concrete interactions (development item M1.5)
Key Phenomenon	Importance	Existing Capabilities	Modeling Gaps
Dissolution of RN and vaporization of dissolved species	Transport of radionuclides to and from the sodium pool and into the cover gas		<ul style="list-style-type: none"> Add models for dissolution and vaporization of dissolved species (development item M1.3)
Bubble Transport/partitioning between bubble & sodium pool	Transport of radionuclides directly to the atmosphere.	MELCOR's SPARC model might be leveraged, though modified significantly for this application	<ul style="list-style-type: none"> Development of bubble transport model (development item M1.3)
Heat Pipe Thermal Hydraulics	The heat pipe is the primary means of heat removal from fuel.		<ul style="list-style-type: none"> MELCOR does not currently have a heat pipe model. Code modifications have been proposed to remove this gap (see Appendix B) (development item M1.1)
Reactor kinetics	Calculate transient power feedback	Existing point kinetics and reactivity feedback model	<ul style="list-style-type: none"> Evaluate neutronics parameters in the existing point kinetics model (development item M1.9)
Failure of Individual heat pipes and propagation of failure to adjacent fuel elements	Determines the extent of core degradation and source term released from fuel.	Existing multi-rod model can be leveraged in calculating propagation of local heat pipe failure (development item 1.8)	<ul style="list-style-type: none"> Development of heat pipe models Development item M1.8.

5.2.1 Radionuclide Inventory

For the assessment of the source term associated with an SFR core damage transient, the first step is the determination of the initial fuel radionuclide inventory, which typically is an input to radionuclide transport codes, such as *SRT*. In general, the codes and process for estimating the radionuclide inventory are well established, with computational tools such as *SCALE* and

ORIGEN used in past analyses [60, 315]. Table 5-2 displays the current maturity level of the *SCALE* suite of codes for neutronics and fuel depletion modeling for SFRs. In general, burnup dependence is used to develop the radionuclide inventory for different batches of fuel within the core. Note that this could include spent fuel that is removed from the core but remains within the vessel in a storage location.

Table 5-2: SCALE Maturity for SFR Analysis [58]

Element	Maturity Level ¹	Comments
Representation and Geometric Fidelity	3	Capability to model reactor designs for the needs of MELCOR/MACCS is available. However there needs to be further assessment work performed
Physics and Model Fidelity	3	Capability exists to model SFR designs to support MELCOR calculations. However there needs to be further assessment work performed
Code Verification	2	Internal SQA program provides coverage for code verification
Solution Verification	2	SCALE has been used for the SFR UAM and been provided with some formal review. Assessments continue.
Model Validation	1	Assessments continue with EBR2
Uncertainty Quantification and Sensitivity Analysis	2	Uncertainties and numerical propagation of errors have been examined extensively for LWR applications though not for Na application

¹Maturity Levels

- level 0, little or no assessment of accuracy and completeness and highly reliant on personal judgment and experience;
- level 1, some informal assessment of accuracy and completeness, and some assessment has been made by an internal peer review group;
- level 2, some formal assessment of accuracy and completeness, and some assessments have been made by an external peer review group; and
- level 3, formal assessment of accuracy and completeness, and essentially all assessments have been made by an independent, external peer review group.

For potential radionuclide releases associated with support systems, such as cold traps or cover gas cleanup systems, the initial radionuclide inventory can typically be derived based on system design parameters. For example, cesium traps that are part of the primary sodium cleanup system are typically sized to accommodate a specified amount of Cs-137 (in curies). In addition, experience with past U.S. SFRs can serve as the basis of estimates of the activity contained within the cover gas system and primary sodium. See Section 3 of ref [310] for additional detail on the radionuclide inventory associated with non-core SFR sources.

5.2.2 Fuel Migration and Release

For core damage transients, in addition to the initial radionuclide inventory, the location and chemical form of the radionuclides within the fuel pin is also an influential factor on potential fuel releases. For metal fuel SFR cores, a recent study [316] explored radionuclide migration and potential fuel pin radionuclide release based on past SFR experience and experimentation. Simplified, burnup-dependent functions were derived for different classes of radionuclides. This approach was adopted for the modeling of radionuclide migration and release within the *SRT* code.

In contrast to this data-driven, simplified approach, the development of mechanistic models regarding metal fuel radionuclide migration may be possible. Work is ongoing to update the metal fuel failure models within *SAS4A/SASSYS-I*, include modules that characterize the migration and release of fuel components, include U/Pu and fission products, as these are important factors in the determination of cladding failure and fuel melting [303]. However, the models do not yet track individual fission product elements to the level of detailed necessary to resolve release fractions during fuel damage events.

Similarly, the NEAMS code *BISON* has also be utilized to characterize metal fuel behavior, including the migration of certain fission products during irradiation, for fuel performance modeling [22, 317] *BISON* has not yet been utilized to examine fission product relocation as part of a source term analysis.

5.2.3 Sodium Pool

Radionuclides released during fuel damage transients will first enter the sodium pool, either directly or as part of fission gas bubbles. In general, separate modeling approaches are necessary to capture these two potential outcomes.

For radionuclides that enter the sodium pool directly, the pool can represent a significant retention mechanism, preventing further release to the cover gas region and containment. For these radionuclides, the resulting chemical form will dictate the mobility of the radionuclide and the potential to vaporize from the pool surface. Thermodynamic equilibrium codes have been utilized previously for this assessment, including *HSC Chemistry* [60] and *ChemSage* [318]. However, custom thermodynamic databases were developed for these efforts, based on past experimentation. In general, any thermodynamic equilibrium code, such as those discussed in Section 2.3, could be utilized for the analysis. A similar approach is used in the *SRT* code, where *HSC Chemistry* was utilized to develop response surfaces that are used in the code to calculate potential vaporization from the pool surface. The weakness of the thermodynamic equilibrium approach is that it ignores chemical reaction kinetics and potential heterogeneity within the sodium pool and is also directly dependent on the quality of the associated database. In addition, physical phenomena, such as plate-out on structures is also not considered.

In contrast, radionuclide bubble transport through the sodium pool has previously been noted as a high priority for SFR MST calculations, given that it has the potential to bypass the sodium pool and its radionuclide retention capabilities [60]. A bubble transport code utilizing classical theories for pool scrubbing was developed by Argonne during the Integral Fast Reactor (IFR) project in the 1980s [319]. The code included models for Brownian diffusion, inertial deposition, gravitational sedimentation, and condensation, but does not capture thermophoresis or diffusiophoresis. Recently, this code was updated and integrated into the *SRT* code for the analysis of bubble transport. Lack of validation data for radionuclide bubble transport is a known gap [60] and in response, DOE is funding ongoing experimental efforts to produce validation data for codes such as *SRT* [320].

5.2.4 Aerosol Modeling

As highlighted previously, radionuclide aerosol behavior is important for both fuel damage accidents and also potential radionuclide releases from auxiliary systems. For fuel damage accidents, radionuclides may exist in the cover gas region due to bubble transport or vaporization from the pool (and subsequent condensation to an aerosol form). Since inert gases are used in the cover gas region, the main phenomena of interest are agglomeration and deposition. The cover gas region will also contain a significant amount of sodium aerosols and vapor. For auxiliary systems, the initial aerosol assessment may be similar if they are placed within an inerted vault. If not, the aerosol analysis more closely resembles the containment assessment outlined below.

A potentially important phenomenon for radionuclide transport from the cover gas region to containment⁴ is leak plugging due to the formation of sodium oxides. If a leakage path is available for aerosols to move from the inert cover gas region to containment, the aerosols will likely encounter oxygen along the pathway. With the high concentration of sodium vapor and aerosols, the formation of sodium oxide, which has a melting temperature of $>1,100^{\circ}\text{C}$, will cause solid deposits, restricting further transport. The degree and rate at which a leakage pathway may be restricted is highly dependent on the available flow area and the orientation of the pathway.

For aerosols within containment, the presence of oxygen and water will result in chemical reactions with sodium. The formation of sodium oxides has both chemical and physical consequences. Sodium in combination with iodine (NaI) may dissociate in reaction with oxygen to form sodium oxide and gaseous iodine, which could promote further iodine transport [321]. Sodium reactions with oxygen will also change the physical form of the aerosols, potentially increasing agglomeration and deposition.

Within *SRT*, several simplified aerosol models are available, including the use of user-specified, element-specific removal rates due to deposition and a correlation developed by Fauske and Associates (FAI) based on past sodium aerosol experimentation [322]. These models are considered simplified as they do not mechanistically model the physics of aerosol behavior, such as diffusion, thermophoresis, etc., described in Section 2.4.

5.2.5 Sodium Fires

Historically, a variety of sodium fire codes have been utilized in the U.S. for the assessment of room temperatures and pressures associated with sodium leaks or spills. These include *SOFIRE-II* [323] and *NACOM* [324], which are no longer supported. The models within these codes were later integrated in *CONTAIN-LMR 1.0* [325] and have now also been added to *MELCOR 2.2* [83]. Although these codes include the formation of sodium oxide aerosols, they do not model the release of any radionuclides contained with the sodium itself or the associated structure (such as a cesium trap). Previous experimentation has explored the release of radionuclides during sodium fires (see Section 5 of ref [310]) but such information has not yet been included in any sodium fire modeling and simulation tools. The inclusion of separate models for the release of radionuclides from the burning sodium is important, as the release fraction of radionuclides can differ substantially from those of the sodium itself (see ref [326] for examples).

⁴ This phenomenon would also be important for leakage from an inerted vault containing auxiliary systems.

5.3 Lead-Cooled Fast Reactors

For pool-type lead-cooled fast reactors (LFRs), the source term phenomena may be sufficiently similar to those for SFRs that most models which have been developed for one reactor concept could be applicable to the other. Therefore, only a brief summary of key differences and LFR-specific code development is provided here.

Although SFR and LFR source term assessment have many similarities, there are several key differences. First, while U.S. SFR vendors typically utilize sodium-bonded metal fuel, for LFR designers, oxide, nitride, and bondless metal fuels have all been proposed. This has major implications on the failure models and radionuclide release. While radionuclide transport within oxide fuel is well understood from the LWR industry, radionuclide transport as a result of boundary breaches in oxide fuels is less understood. Additionally, the use of novel nitride and bondless metal fuel concepts may require new model development. Next, the production of Po-210 from Bi-209 within the lead coolant is another potential source of radioactive material unique to LFRs. This is most common in reactors cooled with a lead-bismuth eutectic, but is also present in other LFRs from Bi-209 impurities or Bi-209 generation from the isotope Pb-208 [327]. As with SFRs, the lead pool offers a significant radionuclide retention pathway, but the difference in chemical forms of some radionuclides will require analysis to understand the impact. While thermodynamic equilibrium codes could again be used for this task, it would require the development of a lead-specific database⁵. Lastly, energetic reactions between lead and oxygen are not a concern at the normal operating temperatures of LFRs.

Recent efforts have pursued the expansion of *SRT* to include LFR-specific modeling capabilities [328]. *HSC Chemistry* was used to develop new response surfaces for radionuclide vaporization from the lead pool, which are incorporated into *SRT*. This task necessitated the initial development of a lead-specific thermodynamic database for the analysis. In addition, the bubble transport model in *SRT* was also modified to account for lead properties. Other models within the code were considered to be equally applicable to LFR designs.

Hua et al. reported on the development of a MST analysis tool called *SAS4A-FATE* for LFRs and SFRs [329]. The safety analysis code *SAS4A/SASSYS-1* was coupled to the facility analysis *Facility flow, Aerosol, Thermal, and Explosion (FATE)* code, with a *Radionuclide Release Module (RRM)* developed to link the codes together. *FATE* uses information from *SAS4A/SASSYS-1* to determine radionuclide releases and tracks transport through the coolant pool, cover gas region, and the containment system. *SAS4A/SASSYS-1* calculates fuel temperature and fuel rod failure, feeding as inputs to *RRM* which calculates the radionuclide release rate from the fuel, the transport and retention in the primary liquid pool, and the release to the cover gas region. The initial radionuclide inventory in the fuel is user-specified, typically calculated by a code such as *ORIGEN* based on fuel loading, assembly power, and burnup histories. Of this inventory, fractions of radionuclides which have accumulated in the fuel-cladding gap and in the fission gas plenum are calculated and are immediately released to the coolant pool upon fuel cladding failure. The remaining inventory in the fuel matrix is released based on a diffusional release model dependent on time, the grain radius, and a diffusion coefficient, which is calculated

⁵ DOE is currently funding experimental work to examine the behavior of radionuclides in liquid lead under NEUP project 18-15471.

from temperature and pre-determined multipliers pertaining to each class of radionuclides. The model is largely based on empirical data for oxide fuels in LWRs.

Once released to the coolant pool in *RRM*, radionuclides are transported to the cover gas either as gas bubbles or aerosols within bubbles. A fraction can be deposited in the coolant due to either forming a stable compound with the lead coolant, dissolving into the coolant pool, or having a low vapor pressure at the temperature of the pool. Scrubbing is handled in the model by user-input decontamination factors for each radionuclide class. The user is left to consider the chemical equilibrium and dynamics of aerosol containing gas bubbles for a particular coolant and chemical species of radionuclide formed in the fuel. *FATE* assesses radionuclide vaporization from the pool based on available vapor pressure information.

5.4 Capability Assessment

Recent studies have evaluated the current modeling and simulations capabilities for SFR source term assessments, including the available validation data [60, 330]. Ref [60] identified gaps in modeling capabilities and determined their importance through sensitivity studies. The results of this analysis are presented in Table 5-3, with a ranking of gap-groups by priority in Table 5-4. Based on this evaluation, the study found that gaps regarding radionuclide bubble transport and in-pin radionuclide migration and release were of highest priority for resolution. The results of this study were one of the motivators of the sodium bubble transport experiments highlighted in Section 5.2.3 and have guided subsequent development efforts for the *SRT* code.

Table 5-3: Grouping of SFR MST Gaps from ANL-ART-49⁶ [60]

Group	Phenomena	Gap	Importance¹
<i>In-Pin Migration and Release</i>	<i>Radionuclide migration within the fuel pin during irradiation</i>	Partial	<i>H</i>
	<i>Radionuclide release fractions from failed fuel pins</i>	Partial	<i>H</i>
	<i>Radionuclide partitioning between gas and liquid/solid in plenum region</i>	Partial	<i>H</i>
	<i>Timing of radionuclide release from failed fuel pins</i>	Partial	<i>L</i>
<i>Bubble Transport</i>	<i>Partitioning of released radionuclides into bubbles or sodium pool</i>	Yes	<i>VH</i>
	<i>Bubble diameter</i>	Yes	<i>VH</i>
	<i>Aerosol particle size distribution</i>	Yes	<i>VH</i>
	<i>Aerosol particle density</i>	Partial	<i>VH</i>
	<i>Scrubbing of vapors/aerosols in bubble passing through sodium pool</i>	Partial	<i>VH</i>
<i>Vaporization</i>	<i>Vaporization from Sodium Pool (Thermodynamic Effects)</i>	Partial	<i>L</i>
	<i>Timing of Vaporization</i>	Yes	<i>L</i>
	<i>Kinetic Effects</i>	Yes	<i>L</i>
	<i>Non-homogenous Radionuclide Mixing (Surface-Boundary Concentration)</i>	Yes	<i>L</i>
	<i>Plate-out (Adsorption) and Settling in Sodium Pool</i>	Yes	<i>L</i>
	<i>Mechanical Forces (Liftoff due to Bubble Pop, Turbulence, or Convection)</i>	Yes	<i>L</i>
<i>Aerosol Behavior</i>	<i>Radionuclide Decay and Daughter Products</i>	Partial	<i>L</i>
	<i>Aerosol Agglomeration and Deposition</i>	Partial	<i>M</i>
	<i>Resuspension and Re-vaporization</i>	Yes	<i>M</i>
	<i>Aerosol Deposition in Flow Paths</i>	Yes	<i>M</i>
<i>Hold-up/Leakage</i>	<i>Cover Gas Temperature and Pressure Response</i>	Partial	<i>M</i>
	<i>Seal Leakage</i>	Yes	<i>M</i>
	<i>Containment Temperature and Pressure Response</i>	Yes	<i>M</i>
<i>Dispersion</i>	<i>Radionuclide Characteristics</i>	Partial	<i>L</i>
	<i>Chemical Interactions</i>	Yes	<i>L</i>
	<i>Dose Conversion Factors</i>	Partial	<i>L</i>

¹VH – Very High, H – High, M – Medium, L – Low

Table 5-4: Prioritization of SFR MST Gap Groups from ANL-ART-49 [60]

Ranking	Group	Notes
1	<i>Bubble Transport</i>	<i>Represents a potential bypass mechanism of a major radionuclide barrier (the sodium pool). Very high importance in sensitivity calculations, and direct impact on non-noble gas radionuclide transport. Difficult to determine if analysis assumptions are realistic, and significant impact on subsequent analysis steps.</i>
2	<i>In-Pin Migration and Release</i>	<i>Determines initial radionuclide release fractions. High importance in sensitivity calculations. Conservative assumptions are straightforward to apply, but assumptions propagate through subsequent analysis steps, resulting in potentially unrealistic releases of transuranics.</i>
3	<i>Aerosol Behavior</i>	<i>Aerosol deposition/condensation is a significant retention mechanism, with medium importance in sensitivity calculations. Data regarding deposition, condensation, and chemical interactions are not as well-established as LWR data. Codes, such as MELCOR, have ability to model phenomena, but data are a necessary input.</i>
4	<i>Hold-up/Leakage</i>	<i>The delay in radionuclide releases due to hold-up in the cover gas region and containment. Medium importance in sensitivity calculations. Assumptions are straightforward to apply and characterize, as shown in trial MST calculation where multiple leakage values were assumed.</i>
5	<i>Vaporization</i>	<i>Vaporization of radionuclides from the sodium pool. Many gaps in the modeling of phenomena, but was low importance in sensitivity calculations (and in trial MST calculation). However, the relative importance of vaporization could increase, if bubble transport calculations can be shown to be overly conservative.</i>
6	<i>Dispersion</i>	<i>Radionuclide characteristics and dose conversion factors. Although these factors have a direct impact on offsite consequence, the magnitude of their influence is likely lower than the factors related to radionuclide release fractions from the plant.</i>

⁶ Those phenomena that were not associated with a gap, and those considered part of “accident modeling” rather than source term assessment, are not included in the table.

Based on the results of past studies and the evaluation performed as part of the current work, several key gaps were identified regarding the current capabilities of modeling and simulation tools for metal fuel pool-type SFRs, described below:

- **Radionuclide in-pin migration and fuel release:**
 - Available modeling tools primarily use data-driven correlations regarding radionuclide migration within the fuel pin during irradiation and potential release during fuel pin failure events. High-fidelity fuel performance models, while available, have not been used to assess radionuclide behavior in the context of source term analyses.
- **Radionuclide bubble transport:**
 - Ongoing experimentation is examining the behavior of radionuclide bubbles in both sodium and lead. The available modeling tools may require refinement based on the experimental results.
- **Radionuclide aerosol behavior:**
 - Current modeling tools do not mechanistically account for chemical reactions associated with sodium aerosols, including the possible dissociation of radionuclide species and the potential for sodium oxide plugging of leakage pathways.
- **Sodium fires:**
 - Current sodium fire analysis tools do not assess the release of radionuclides entrained within the burning sodium, although some experimental data regarding the phenomenon is available.

In addition to those gaps listed above, deficiencies were also identified regarding the use of thermodynamic equilibrium models for the assessment of radionuclide vaporization from the sodium pool, as do not capture kinetics or potentially important non-homogeneities. However, ref [60] placed low importance on vaporization phenomena in comparison to those outlined above due to its small impact in sensitivity studies.

6 Consequence Modeling

6.1 Overview of Consequence Modeling

After an MST is created by either an integral code such as *MELCOR* or an analogous code suite, the consequences of the source term need to be assessed. This is done both onsite, for workers, and offsite, for the public. Both health and economic consequences may be of interest, depending on the application. The consequence of the radionuclide source term is assessed by modeling the behavior of local weather conditions and site characteristics to include population data and local land use data. Radionuclides are tracked as they exit the containment or reactor facility into ancillary systems or the environment and are transported and dispersed within the atmosphere [3].

Important information for consequence assessment includes, but is not limited to: [3]

- Atmospheric transport and dispersion
- Wet and dry deposition
- Meteorology
- Exposure pathways
- Dose to workers and the public
- Health effects
- Economic losses from events
- Consequence assessment provides guidance to regulators, governments and other local authorities as to what emergency phase, intermediate phase, and long-term phase protective actions need to be implemented to ensure public safety [3].

As highlighted in Section 2.6, the RAMP code *RADTRAD* is used within the NRC framework to calculate three doses, (1) those in the control room; (2) at the edge of the exclusion area boundary (EAB); and (3) in the low-population zone (LPZ) [91]. *RADTRAD* uses a combination of (1) realistic system behavior and (2) atmospheric dispersion characteristics to model radionuclides as they move from the primary containment to elsewhere onsite [3].

For calculations of consequence offsite, such as dose to the public resultant from an event, the *MACCS* software is primarily used. It accounts for atmospheric transport, dispersion and deposition of radionuclides. From those results, it estimates doses, health effects, land contamination areas, and economic consequences [3, 331].

The *RASCAL* software, a RAMP code, is a response tool used to make recommendations regarding emergency response decisions. During a radiological release event, it is used by the NRC, state governments and other local responders to provide guidance on evacuation decisions [3, 332].

All three of these software programs are funded primarily by the NRC and differ significantly in terms of the level of physical representation involved and applicable use cases. The responsibility to develop and maintain such software traditionally falls to regulatory agencies worldwide. This is especially the case for emergency response software, such as *RASCAL*. Both *RADTRAD* and *RASCAL* are distributed through the RAMP program, along with several other atmospheric modeling and consequence software programs, all of which can be used to support different parts

of the regulatory process [3, 333]. *MACCS* is distributed through the *CSARP* along with *MELCOR*, *MelMACCS*, *SCALE*, and several other software tools. *MACCS* is used for licensing and regulatory analyses.

This section highlights atmospheric transport and dispersion, as well as radiological event consequence assessment software that may be used to license advanced reactors. It also highlights phenomena that need to be addressed within any atmospheric transport or consequence software before it can be used for advanced reactors. Relevant assessments of software for near-field and consequence modeling capability are addressed.

6.2 Offsite Dispersion Modeling Software and Associated Capabilities

There are many offsite dispersion codes currently available. This subsection highlights the high-level capabilities of radionuclide transport and dispersion models, as well as consequence assessment software. It is possible to use a subset of these software programs to develop the licensing case for an advanced reactor, provided the appropriate adjustments for non-LWR phenomena were added, as described in the following subsection.

1. *AERMOD* [334]
 - Developed by EPA
 - Gaussian plume model
 - Designed for short-range (up to 50 km) impacts from a variety of industrial source types
2. *ARCON96* [335]
 - Developed by Pacific Northwest National Laboratory (PNNL)
 - Code system to calculate atmospheric relative concentrations in and downwind of building wakes for nearfield doses
 - Modified Gaussian plume model
 - Developed to calculate relative concentrations in plumes from nuclear power plants at control room air intakes in the vicinity of the release point
 - May be used by Kairos Power (FHR) for atmospheric dispersion modeling [189]
3. *ARGOS* [336]
 - Developed by PDC-ARGOS
 - Primarily used by European countries without operating nuclear reactors
 - Supports nuclear, radiological, chemical and biological incidents
 - Uses *RIMPUFF* atmospheric dispersions model for short/medium range
 - Supports multiple long-range atmospheric dispersion models
4. *CALPUFF* [337]
 - Developed by Exponent, Inc., for the EPA
 - Main components of the modeling system are *CALMET* (a diagnostic meteorological model), *CALPUFF* (air quality dispersion model), and *CALPOST* (post-processing package).
 - Gaussian puff model
 - Applications
 - Near-field impacts in complex flow or dispersions situations
 - Long-range transport

- Criteria and secondary pollutant modeling
 - Visibility assessments for Class I areas
5. *EPIcode* [338]
 - Developed by National Atmospheric Release Advisory Center (NARAC) at LLNL
 - Analyzes atmospheric dispersion hazards from toxic chemicals
 - Used for emergency response by DOE
 6. *FLEXPART* [339]
 - Open-source, developed by an international consortium
 - Lagrangian transport dispersion model
 - Higher fidelity code that requires significant knowledge of the system
 - Designed for calculating long-range and mesoscale dispersion of air pollutants from point sources
 - Can be run in either forward or backward mode
 - Forward mode determines downwind concentrations or mixing ratio of pollutants
 - Backward mode estimates origin of observed emissions
 7. *GENII Version 2* [340]
 - Developed by PNNL
 - Estimates radionuclide concentrations in the environment and doses to humans from acute or chronic exposures from radiological releases to the environment or initial contamination conditions
 - Air transport options include both puff and plume models
 - Radionuclide transport via air, water, or biological activity may be considered
 8. *MACCS* [331]
 - Developed by SNL for NRC
 - Calculates the individual and population doses from radiological exposures, health effects, land contamination areas, and economic consequences
 - Used by NRC to model source terms from nuclear power plants
 - Primary tool used to perform atmospheric transport and consequence modeling as part of Level 3 PRA and SOARCA
 - Used by NRC and licensees for NEPA calculations
 - Used by NRC for regulatory and backfit analyses and for research
 - Validated for use with both near-field and far-field ATD problems
 9. *MILDOS* [341]
 - Developed by ANL for the NRC
 - Calculates the radiological dose commitments received by individuals and the general population within an 80-km radius of an operating uranium recovery facility
 - Air and ground concentrations of radionuclides are estimated for individual locations, as well as for a generalized population grid
 - Extra-regional population doses resulting from transport of radon and export of agricultural produce are also estimated
 10. *HOTSPOT* [342]
 - Developed by LLNL for the DOE
 - Gaussian plume model

- Provides emergency response personnel and planners with a fast and portable set of software tools for evaluating incidents involving radioactive material
- Also used for safety analyses of facilities handling nuclear material
- Designed for near-surface releases, short-range (less than 10 km), and short-term (less than 24 hours)
- Unobstructed terrain and simple meteorological conditions

11. *OpenFOAM* [343]

- Developed by CFD Direct Ltd.
- Open source computational fluid dynamics software
- Applicable to a wide range complex fluid flows involving:
 - Chemical reactions
 - Turbulence
 - Heat transfer
- Actively used for aviation and atmospheric modeling

12. *RADTRAD*

- *RADTRAD-AC* – Developed by NRC [90, 344]
- *RADTRAD-NAI* - Developed by Zachry Nuclear Engineering [91]
- *RADTRAD* is a licensing analysis code used to show compliance with nuclear plant siting criteria for:
 - Exclusion Area Boundary (EAB) and
 - Low Population Zone (LPZ)
- Assesses occupational radiation doses in the control room (CR) and /or Emergency Offsite Facility for various loss-of-coolant accidents (LOCA) and non-LOCA design basis accidents (DBAs)
- *RADTRAD* uses a combination of tables and numerical models of source term reduction phenomena to determine the time-dependent dose at the CR, EAB and LPZ for given DBA scenarios
- Will be modified and used by Kairos Power (FHR) for EAB dose estimates [189]

13. *RASCAL* [332]

- Developed by SNL, PNNL and Athey Consulting for the NRC
- Emergency response software that is used to assess dose to the public at the site boundary
- Contains an atmospheric transport and dispersion model as well as a reduced order representation model of the nuclear power plant system, including the containment and filtration systems
- Considers a variety of different reactor types and accident scenarios, accounting for safety systems

14. *RODOS* [345]

- Developed by Karlsruhe Institute of Technology
- Decision support system for assessing, presenting and evaluating nuclear accident consequences
- Can be applied at any distance from the release site
- Short range model utilizes *RIMPUFF* and *ASTEP*

15. *SCIPUFF* [346]

- Developed by Environmental Sciences group of Sage Management
- Gaussian puff model

- Used for short range (up to 50 km) and long range (up to 3000 km)
- Appropriate for modeling steady or non-steady state emissions of primary pollutants with buoyant or neutral sources using time-dependent meteorological data

16. *QUIC* [347]

- *Quick Urban & Industrial Complex Dispersion (QUIC)*
- Developed by Los Alamos National Laboratory
- Chemical, biological, and radiological agent dispersion modeling capability
- Building to neighborhood scales
- Accounts for the effects of buildings and local terrain

6.3 Regulatory Efforts to Improve Non-LWR Modeling Capability of ATD and Consequence Assessment Software

The NRC is currently assessing the capabilities of *MACCS* for use with non-LWRs and other advanced reactor types. The assessments of the software are guiding plans to improve the *MACCS* software to better capture non-LWR specific behavior, as outlined in Volume 3 of the NRC vision and strategy document [58].

Given that many advanced reactor vendors are seeking to reduce the size of the plant site and EAB, the first step of enacting this plan was to perform a near-field assessment of the NRC-standard *MACCS* software against *ARCON*, *AERMOD* and *QUIC*. These three software programs have a widely accepted capability to model near-field atmospheric transport and dispersion (ATD). Evaluations were performed for a wide range of weather conditions, building dimensions, and plume buoyancy that are representative of potential accident conditions. This analysis indicated that *MACCS* provides a bounding result when an appropriate set of input choices are used. The specific parameters that are required for adequate representation of nearfield modeling in *MACCS* are: [348]

- The parameterization of Eimutis and Konicek [349] for the dispersion model.
- The plume meander model based on NUREG/CR-2260 [350] and Regulatory Guide 1.145 [351].
- The release modeled as a ground-level, nonbuoyant plume.
- The inclusion of the effects of the building wake (area source).

These results are summarized in in ref [348]. This report demonstrates that *MACCS* can be used at distances significantly shorter than 500 m downwind from a containment or reactor building [348].

6.4 Phenomenological and Modeling Gaps

Phenomenological gaps in existing ATD codes to model advanced reactor source terms were identified by SNL at the request of the NRC. SNL focused on gaps that exist within the *MACCS* software suite. However, the phenomena that need to be represented in order to adequately model the behavior of non-LWR source terms as they are transported and deposited on the ground are generally the same for any ATD code.

Categorically, any software would need to model the following in order to capture radionuclide transport, dispersion and associated consequences: [351, 352]

- Characteristics of the released radionuclides
- Atmospheric transport and dispersion
- Meteorological data
- Protective action and other site data
- Dosimetry
- Health effects
- Economic factors

The released radionuclides have several characteristics that need to be captured, these include: [351, 352]

- Magnitude and timing of releases
- Aerosol particle size distribution
- Release elevation
- Buoyancy of released plume
- Chemical composition of release radionuclides
- Isotopic inventory

Of these parameters, the release elevation and buoyancy of the release plume of advanced reactor source terms is very similar to those that existing software are already capable of modeling. The magnitude and timing of releases is something that would be taken as an input for an ATD code, not something that an ATD code would determine for itself. However, significant differences may be seen in the chemical composition of released radionuclides and the isotopic inventory, when compared to historic LWR models. The chemical composition of released radionuclides might impact the hygroscopic nature of the aerosols, which may influence deposition once aerosols are released into the atmosphere. Chemical form may also affect dose conversion factors (DCFs). It is possible for released radionuclides to change chemical composition following their release into the environment. This is already an issue for LWRs and might need to be considered for non-LWRs [351, 352].

The majority of ATD parameters are independent of the type of reactor built. For example, meteorological data are fully independent of reactor type, therefore current site weather characterization methods are sufficient. There are two key exceptions to this: released gases and aerosols that evolve in their deposition characteristics; and the necessity to evaluate radionuclide concentrations and doses at short distances. Given the reduced size and passive safety systems of many non-LWRs, the anticipated size of the EAB will likely be smaller for non-LWR systems. Currently, the majority of consequence assessment software does not need to have atmospheric transport and dispersion models that can capture effects, particularly buildings and their wakes, within 0.5 km of the reactor. This has driven the models that have been chosen for ATD and consequence assessment software. This deficiency needs to be addressed through requalification of existing models or new models in existing software [351, 352].

While dosimetry modeling is essentially the same as for existing reactor types, there may be some assumptions related to the absorption type that need to be reevaluated for non-LWR advanced reactors. Absorption type is dependent on the chemical form. Additional research may be needed

to determine if the chemical forms currently assumed for radionuclides are valid for non-LWRs [351, 352].

Like dosimetry, the cost of decontamination may be different if the chemical forms of the radionuclides present are different than those currently released by LWRs. If the chemical forms are different, additional research may be needed to determine the cost of cleanup of the new radionuclide forms [351, 352].

6.5 Capability Assessment

Based on a review of several recent studies of modeling and simulation needs for the assessment of non-LWR offsite consequence [351, 352], the most salient phenomenological gaps are summarized as follows:

- **Near-Field Dispersion:**
 - Modeling near-field dispersion may be required to estimate doses and other consequences at or just beyond the EAB, which may be very close to the reactor location. New models may need to be implemented in existing software to perform this calculation. Enhanced near-field capabilities are already planned and ongoing for some codes (e.g., MACCS).
- **Isotopic Inventory:**
 - If very different than that of an LWR, may need to be reevaluated to ensure that all important isotopes are included in the analysis.
- **Chemical Form:**
 - Differences in chemical form are possible when the oxygen potential within the RCS is substantially different than that of an LWR, where steam is usually the dominant gas-phase component. More important, fission products may interact with coolant and other materials, e.g., sodium or molten salt, that are not present in an LWR, to form new chemical forms.
- **Deposition Behavior:**
 - Evolution of deposition behavior may occur either because aerosols are hygroscopic or because some of the radionuclides are chemically reactive and change chemical form.
- **Dose Conversion Factors:**
 - Depending on the chemical and physical form of the released radionuclides, the applicability of existing DCFs for the internal exposure pathway may require additional evaluation.
- **Decontamination Costs:**
 - Cost of decontamination could be different for advanced reactors if the released isotopes and their chemical forms are substantially different than for existing LWRs.

7 Conclusion

A survey was performed of available modeling and simulation tools for advanced (non-LWR) reactor MST assessments. Functional requirements documented in ref [2] and potential event sequences described in ref [3] were used as the foundation of the assessment of current modeling and simulation capabilities. While the reactor-specific capability gaps are documented within their respective sections, several overarching issues were identified, which are highlighted below:

- **Auxiliary System Modeling:**

Advanced reactors may utilize auxiliary or peripheral systems, such as coolant cleanup systems or off-gas systems, that contain considerable quantities of radionuclides. Given that advanced reactor designs utilize passive and inherent features to reduce the likelihood of severe accidents and radionuclide release from the fuel, event sequences associated with radionuclide releases from these auxiliary systems may be the dominant risk contributors. The source term analysis for the auxiliary systems may require additional model development or data collection, given possible differences in phenomena compared to historical severe accident analysis.

- **Radionuclide Inventory Estimation and Fuel Depletion:**

Many reactor physics tools have been developed for advanced reactors. Code development for the solid-fueled advanced reactors is more mature than that for salt-fueled MSRs, but multiple efforts and developments have been published in modeling the neutronics of flowing fuels.

- **Fuel Performance and Radionuclide Behavior:**

High-fidelity fuel performance codes are available and have been utilized to assess most advanced reactor types. However, they have generally not been used for the assessment of fission product relocation or release in terms of source term analysis. Instead, current source term analysis tools rely primarily on empirical correlations or simplified assumptions.

- **Chemistry Modeling:**

Chemical speciation can have a major impact on the transport properties of radionuclides. Thermodynamic modeling tools, such as Gibbs free energy minimization solvers, have been utilized to inform source term analyses but the tools are generally not integrated directly into the calculation. In addition, the underlying thermochemical data that the solvers use to perform speciation (e.g., phase diagram, and vapor pressure calculations) are incomplete for many systems. Thermodynamic equilibrium codes also do not account for phenomena such as kinetics or inhomogeneity, which will be important as they are known drivers of behavior in LWRs.

- **Aerosol Behavior:**

There is significant knowledge in aerosol modeling from decades of experience and research for LWRs and SFRs, as well as from other industries. While the fundamental phenomena and models are well established, the radionuclide-specific factors, such as chemical form, aerosol size, etc., are not yet known for some event sequences of interest, such as a fuel-salt spill for an MSR. It is likely that codes used for aerosol modeling in LWRs could be modified for use with advanced

reactors with modifications mostly related to the physical and chemical properties of the aerosols based on experimental data.

- **Consequence Modeling:**

Many codes have been developed for dose consequence and off-site dispersion modeling for LWRs as well as other types of nuclear facilities. However, due to a desire to reduce advanced reactor plant sites, an increased focus on near-field dispersion requires model development or modification. In addition, assumptions regarding radionuclide chemical form (and associated dose conversion factors) must also be reviewed for applicability of the selected advanced reactor.

The identified gaps will be utilized in FY21 to develop a modeling and simulation development pathway, which will establish a course for gap resolution. In general, the pathway will attempt to leverage existing tools to rapidly expand and develop advanced reactor MST capabilities. In addition, the development pathway will be generated in coordination with other DOE-NE programs and industry feedback, with a focus on prioritizing gaps that cross-cut multiple reactor types.

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] S. Shahbazi and D. Grabaskas, "Functional Requirements for the Modeling and Simulation of Advanced (Non-LWR) Reactor Mechanistic Source Term," Argonne National Laboratory, ANL/NSE-20/17, June 2020.
- [3] N. C. Andrews, A. J. Clark, T. M. Nenoff, J. E. Leute, and D. L. Luxat, "Mechanistic Source Term Considerations for Advanced Non-LWRs," Sandia National Laboratories, SAND2020-6730, July 2020.
- [4] J. J. DiNunno, R. E. Baker, F. D. Anderson, and R. L. Waterfield, "Calculation of Distance Factors for Power and Test Reactors," Atomic Energy Commission, Division of Licensing and Regulation, TID-14844, March 23 1962.
- [5] L. Soffer, S. B. Burson, C. M. Ferrell, R. Y. Lee, and J. N. Ridgely, "Accident Source Terms for Light-Water Nuclear Power Plants," U.S. Nuclear Regulatory Commission, NUREG-1465, February 1995.
- [6] U.S. Nuclear Regulatory Commission. Office of Nuclear Regulatory Research., Regulatory guide 1.183 alternative radiological source terms for evaluating design basis accidents at nuclear power reactors, [Washington, D.C.]: U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, 2000, p. [62] p. [Online]. Available: <http://purl.access.gpo.gov/GPO/LPS107166>.
- [7] 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.
- [8] W. D. Travers, "Policy Issues Related to Licensing Non-Water Reactor Designs," U.S. Nuclear Regulatory Commission, SECY-03-0047, March 28 2003.
- [9] "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.
- [10] "Risk-Informed Performance-Based Technology Inclusive Guidance for non-LWR Licensing Basis Development, Revision 1," Nuclear Energy Institute, 18-04, August 2019.
- [11] D. L. Luxat, "Simplified Approach for Scoping Assessment of Non-LWR Source Terms," Sandia National Laboratories, SAND2020-0402, January 2020.
- [12] A. Alfonsi, P. Humrickhouse, K. Vedros, and H. Zhang, "Risk Informed, Performance-Based, Technology-Inclusive Regulatory Infrastructure: Technology-Inclusive Determination of Mechanistic Source Terms for Offsite Dose-Related Assessments for Advanced Nuclear Reactor Facilities," Idaho National Laboratory, INL/EXT-20-58717, June 2020.

- [13] "Probabilistic Risk Assessment Standard for Advanced Non-LWR Nuclear Power Plants," ASME/ANS, ASME/ANS RA-S-1.4-2013, December 9 2013.
- [14] "State-of-the-Art Reactor Consequence Analyses (SOARCA) Report: Part 1," NRC, Office of Nuclear Regulatory Research, NUREG-1935, November 2012.
- [15] C. Stanek, "Overview of DOE-NE NEAMS Program," Los Alamos National Laboratory, LA-UR-19-22247, 2019.
- [16] D. Gaston, C. Newman, G. Hansen, and D. Lebrun-Grandié, "MOOSE: A parallel computational framework for coupled systems of nonlinear equations," *Nuclear Engineering and Design*, vol. 239, no. 10, pp. 1768-1778, 2009.
- [17] C. J. Permann *et al.*, "MOOSE: Enabling massively parallel multiphysics simulation," *SoftwareX*, vol. 11, 2020.
- [18] R. L. Williamson *et al.*, "Multidimensional multiphysics simulation of nuclear fuel behavior," *Journal of Nuclear Materials*, vol. 423, no. 1-3, pp. 149-163, 2012.
- [19] M. R. Tonks, D. Gaston, P. C. Millett, D. Andrs, and P. Talbot, "An object-oriented finite element framework for multiphysics phase field simulations," *Computational Materials Science*, vol. 51, no. 1, pp. 20-29, 2012.
- [20] J. Hales, "BISON TRISO Training Material," Idaho National Laboratory, INL/MIS-20-57334-Revision-0, March 2020.
- [21] J. Galloway, N. Carlson, and C. Unal, "BISON framework for the analysis of performance of metallic fuels," Los Alamos National Laboratory, LA-UR-12-26297, 2012.
- [22] J. Galloway, C. Unal, N. Carlson, D. Porter, and S. Hayes, "Modeling constituent redistribution in U–Pu–Zr metallic fuel using the advanced fuel performance code BISON," *Nuclear Engineering and Design*, vol. 286, pp. 1-17, 2015.
- [23] M. R. Tonks *et al.*, "Mechanistic materials modeling for nuclear fuel performance," *Annals of Nuclear Energy*, vol. 105, pp. 11-24, 2017.
- [24] F. N. Gleicher *et al.*, "The coupling of the neutron transport application RATTLESNAKE to the nuclear fuels performance application BISON under the MOOSE framework," presented at the PHYSOR 2014 - The Role of Reactor Physics toward a Sustainable Future, The Westin Miyako, Kyoto, Japan, September 28 - October 3, 2014.
- [25] C. Lee and W. S. Yang, "MC2-3: Multigroup Cross Section Generation Code for Fast Reactor Analysis," *Nuclear Science and Engineering*, vol. 187, no. 3, pp. 268-290, 2017.
- [26] E. R. Shemon, C. Lee, and M. A. Smith, "PROTEUS-SN User Manual, Revision 3.0," Argonne National Laboratory, ANL/NE-14/6 Rev3.0, February 2016.
- [27] E. R. Shemon, C. Lee, and M. A. Smith, "Proteus-SN Methodology Manual," Argonne National Laboratory, ANL/NE-14/5, June 2014.

- [28] Y. S. Jung, C. Lee, and M. A. Smith, "PROTEUS-MOC User Manual," Argonne National Laboratory, ANL/NSE-18/10, September 2018.
- [29] Y. S. Jung, C. Lee, and M. A. Smith, "PROTEUS-NODAL User Manual," Argonne National Laboratory, ANL/NSE-18/4, September 2018.
- [30] G. Aliberti and M. A. Smith, "PERSENT: Need of a deterministic code for sensitivity analysis in 3D geometry and transport theory," presented at the PHYSOR 2014 – The Role of Reactor Physics Toward a Sustainable Future, The Westin Miyako, Kyoto, Japan, September 28 - October 3, 2014.
- [31] R. Hu, L. Zou, and G. Hu, "SAM User's Guide," Argonne National Laboratory, ANL/NSE-19/18, August 2019.
- [32] R. Hu, "SAM Theory Manual," Argonne National Laboratory, ANL/NE-17/4, March 2017.
- [33] R. Hu, G. Hu, L. Zou, G. Zhang, B. Hollrah, and M. Gorman, "SAM Developments to Support Transient Safety Analysis of Advanced Non-LWRs," Argonne National Laboratory, ANL/NSE-19/31, September 2019.
- [34] A. J. Novak *et al.*, "Pronghorn Theory Manual," Idaho National Laboratory, INL/EXT-18-44453-Rev001, May 2020.
- [35] L. Zou, J. W. Peterson, R. C. Martineau, R. N. Slaybaugh, and A. J. Novak, "Pronghorn: a porous media thermal-hydraulics core simulator and its validation with the SANA experiments," Idaho National Laboratory, INL/CON-18-44290-Revision-0, April 2018.
- [36] P. F. Fischer, J. W. Lottes, and S. G. Kerkemeier. (2008). *nek5000 Web page*. Available: <https://nek5000.mcs.anl.gov/>
- [37] E. Merzari, A. Obabko, P. Fischer, and M. Aufiero, "Wall resolved large eddy simulation of reactor core flows with the spectral element method," *Nuclear Engineering and Design*, vol. 364, 2020.
- [38] E. Merzari *et al.*, "Toward Exascale: Overview of Large Eddy Simulations and Direct Numerical Simulations of Nuclear Reactor Flows with the Spectral Element Method in Nek5000," *Nuclear Technology*, pp. 1-17, 2020.
- [39] R. Martineau *et al.*, "Multiphysics for nuclear energy applications using a cohesive computational framework," *Nuclear Engineering and Design*, vol. 367, 2020.
- [40] B. W. Spencer *et al.*, "Grizzly Usage and Theory Manual," Idaho National Laboratory, INL/EXT-16-38310, March 2016.
- [41] B. W. Spencer *et al.*, "Summary of Grizzly Development for Advanced Reactor Structural Materials," Idaho National Laboratory, INL/EXT-19-55958, September 2019.

- [42] D. Parsons, J. M. Solberg, R. M. Ferencz, M. A. Havstad, N. E. Hodge, and A. P. Wemhoff, "Diablo User Manual," Lawrence Livermore National Laboratory, UCRL-SM-234927, 2007.
- [43] Y. Q. Yu, E. R. Shemon, J. W. Thomas, V. Mahadevan, R. Rahaman, and J. M. Solberg, "SHARP User Manual," Argonne National Laboratory, ANL/NE-16/6, March 2016.
- [44] J. M. Solberg, "LLNL contributions to ANL Report ANL/NE-16/6 "Sharp User Manual", " Lawrence Livermore National Laboratory, LLNL-TR-691097, May 2016.
- [45] E. R. Shemon, Y. Yu, Y. S. Jung, B. Feng, and T. K. Kim, "Extension and Demonstration of NEAMS Multiphysics Tools to Lead-Cooled, Sodium-Cooled, and Molten Salt Fast Reactor Applications," Argonne National Laboratory, ANL/NSE-19/30, September 2019.
- [46] M. H. A. Piro, S. Simunovic, and T. M. Besmann, "Thermochimica User Manual v1.0," Oak Ridge National Laboratory, ORNL/TM-2012/576, December 2012.
- [47] M. H. A. Piro, S. Simunovic, T. M. Besmann, and M. Poschmann, "Thermochimica User Manual," 2019, Available: <https://github.com/ORNL-CEES/thermochimica>, Accessed on: September 1, 2020.
- [48] M. Poschmann, B. W. N. Fitzpatrick, S. Simunovic, and M. H. A. Piro, "Recent Development of Thermochimica for Simulations of Nuclear Materials," in *TMS 2020 149th Annual Meeting & Exhibition Supplemental Proceedings*, 2020, pp. 1003-1012.
- [49] M. Poschmann, M. H. A. Piro, and S. Simunovic, "Acceleration of Thermochimica Calculations in Bison," Oak Ridge National Laboratory, ORNL/TM-2020/1473, January 2020.
- [50] T. M. Besmann, J. C. Ard, K. E. Johnson, M. S. Christian, and J. A. Yingling, "The Molten Salt Thermodynamic Database (MSTDB): A Resource for MSR Design, Development, and Regulation," presented at the American Nuclear Society, Virtual Conference, June 8-11, 2020.
- [51] J. W. McMurray, T. M. Besmann, J. Ard, S. Utlak, and R. A. Lefebvre, "Status of the molten salt thermodynamic database, MSTDB," Oak Ridge National Laboratory, ORNL/SPR-2019/1208, August 2019.
- [52] P. Bajpai, M. Poschmann, D. Andrš, C. Bhave, M. Tonks, and M. Piro, "Development of a New Thermochemistry Solver for Multiphysics Simulations of Nuclear Materials," in *TMS 2020 149th Annual Meeting & Exhibition Supplemental Proceedings*(The Minerals, Metals & Materials Series, 2020, pp. 1013-1025.
- [53] W. A. Wieselquist, R. A. Lefebvre, and M. A. Jessee, "SCALE Code System, Version 6.2.4," Oak Ridge National Laboratory, ORNL/TM-2005/39, April 2020.
- [54] S. M. Bowman, "SCALE 6: Comprehensive Nuclear Safety Analysis Code System," *Nuclear Technology*, vol. 174, no. 2, pp. 126-148, 2011.

- [55] M. D. DeHart and S. M. Bowman, "Reactor Physics Methods and Analysis Capabilities in SCALE," *Nuclear Technology*, vol. 174, no. 2, pp. 196-213, 2011.
- [56] I. C. Gauld, G. Radulescu, G. Ilas, B. D. Murphy, M. L. Williams, and D. Wiarda, "Isotopic Depletion and Decay Methods and Analysis Capabilities in SCALE," *Nuclear Technology*, vol. 174, no. 2, pp. 169-195, 2011.
- [57] S. Goluoglu, L. M. Petrie, M. E. Dunn, D. F. Hollenbach, and B. T. Rearden, "Monte Carlo Criticality Methods and Analysis Capabilities in SCALE," *Nuclear Technology*, vol. 174, no. 2, pp. 214-235, 2011.
- [58] "NRC Non-Light Water Reactor (Non-LWR) Vision and Strategy, Volume 3 - Computer Code Development Plans for Severe Accident Progression, Source Term, and Consequence Analysis, Draft Rev.1," U.S. Nuclear Regulatory Commission, ML20030A178, January 31 2020.
- [59] M. J. Bell, "ORIGEN - the ORNL isotope generation and depletion code," Oak Ridge National Laboratory, ORNL-4628, May 1973.
- [60] D. Grabaskas *et al.*, "Regulatory Technology Development Plan - SFR Mechanistic Source Term Trial Calculation," Argonne National Laboratory, ANL-ART-49, October 2016.
- [61] P. R. Patel and A. John Arul, "In-vessel source term calculation using chemical equilibrium approach for a medium sized sodium cooled fast reactor," *Nuclear Engineering and Design*, vol. 362, 2020.
- [62] J. Kalilainen, S. Nichenko, and J. Krepel, "Evaporation of materials from the molten salt reactor fuel under elevated temperatures," *Journal of Nuclear Materials*, vol. 533, 2020.
- [63] C. W. Bale *et al.*, "FactSage thermochemical software and databases, 2010–2016," *Calphad*, vol. 54, pp. 35-53, 2016/09/01/ 2016.
- [64] G. Eriksson and K. Hack, "ChemSage—A computer program for the calculation of complex chemical equilibria," *Metallurgical Transactions B*, vol. 21, no. 6, pp. 1013-1023, 1990/12/01 1990.
- [65] D. A. Kulik *et al.*, "GEM-Selektor geochemical modeling package: revised algorithm and GEMS3K numerical kernel for coupled simulation codes," *Computational Geosciences*, vol. 17, no. 1, pp. 1-24, 2013/02/01 2013.
- [66] I. K. Karpov, K. V. Chudnenko, D. A. Kulik, O. V. Avchenko, and V. A. Bychinskii, "Minimization of Gibbs free energy in geochemical systems by convex programming," *Geochemistry International*, vol. 39, pp. 1108 - 1119, 2001.
- [67] (2020). *GEMS specific HERACLES v.0.2. database for U, TRU and FP speciation*. Available: <https://www.psi.ch/en/heracles/gems-specific-heracles-database>
- [68] E. B. White, S. M. Johnson, and G. B. Dantzig, "Chemical Equilibrium in Complex Mixtures," *The Journal of Chemical Physics*, vol. 28, pp. 751 - 755, 1958.

- [69] Outotec. (2019). *HSC - Equilibrium Module*. Available: <https://www.outotec.com/globalassets/products/digital-solutions/hsc/13-Equilibrium-Module.pdf>
- [70] B. Sundman *et al.*, "The OpenCalphad thermodynamic software interface," (in eng), *Computational materials science*, vol. 125, pp. 188-196, 2016.
- [71] M. Hillert, "Some viewpoints on the use of a computer for calculating phase diagrams," *Physica B+C*, vol. 103, no. 1, pp. 31-40, 1981/01/01/ 1981.
- [72] J. O. Andersson, T. Helander, L. Höglund, P. Shi, and B. Sundman, "Thermo-Calc & DICTRA, computational tools for materials science," *Calphad*, vol. 26, no. 2, pp. 273-312, 2002/06/01/ 2002.
- [73] 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.
- [74] E. C. Beahm, C. F. Weber, T. S. Kress, and G. W. Parker, "Iodine chemical forms in LWR severe accidents. Final report," U.S. Nuclear Regulatory Commission, Oak Ridge National Laboratory, NUREG/CR-5732, ORNL/TM-11861, April 1992.
- [75] D. J. Wren, "Kinetics of iodine and cesium reactions in the CANDU reactor primary heat transport system under accident conditions," Atomic Energy of Canada Ltd., AECL-7781, April 1983.
- [76] V. M. McCree, "Accident Source Terms and Siting for Small Modular Reactors and Non-Light Water Reactors," U.S. Nuclear Regulatory Commission, SECY-16-0012, February 7 2016.
- [77] "State-of-the-art report on nuclear aerosols for LWRs," OECD Nuclear Energy Agency, Committee on the Safety of Nuclear Installations, NEA/CSNI/R(2009)5, 2009.
- [78] N. Andrews, C. Faucett, S. Belon, C. Bouillet, and H. Bonneville, "MELCOR-ASTEC Crosswalk of the Accident at Fukushima-Daiichi Unit 1: Phase I Analysis," U.S. Nuclear Regulatory Commission, NUREG/IA-0510, June 2019.
- [79] D. Luxat, J. Hanophy, and D. Kalinich, "Modular Accident Analysis Program (MAAP)-MELCOR Crosswalk Study Phase 1," EPRI Technical Report, Report 3002004449, 2014.
- [80] F. Gelbard, "MAEROS User Manual," U.S. Nuclear Regulatory Commission, Sandia National Laboratories, NUREG/CR-1391, SAND80-0822, December 1982.
- [81] J. W. Lane, T. L. George, S. W. Claybrook, J. A. Zankowski, and T. Kindred, "Applicability of Gothic 8.3 (QA) for Non-LWR Simulation, Aerosol Modeling and Hydrogen Management," presented at the NURETH-18, Portland, OR, August 18-22, 2019.
- [82] "MELCOR Computer Code Manuals, Vol. 1: Primer and Users' Guide, Version 2.2.9541," Sandia National Laboratories, SAND2017-0455 O / ML17040A429, January 2017.

- [83] "MELCOR Computer Code Manuals, Vol. 2: Reference Manual, Version 2.2.9541," Sandia National Laboratories, SAND2017-0876 O / ML17040A420, January 2017.
- [84] (2020, August 31, 2020). *MELCOR: A Computer Code for Analyzing Severe Accidents in Nuclear Power Plants and the Design Basis Accidents for Advanced Plant Applications*. Available: <https://melcor.sandia.gov/about.html>
- [85] D. L. Luxat, D. A. Kalanich, J. T. Hanophy, R. O. Gauntt, and R. M. Wachowiak, "MAAP-MELCOR Crosswalk Phase 1 Study," *Nuclear Technology*, vol. 196, no. 3, pp. 684-697, 2016.
- [86] P. Hofmann, "Current knowledge on core degradation phenomena, a review," *Journal of Nuclear Materials*, vol. 270, no. 1-2, pp. 194-211, 1999.
- [87] U.S. Nuclear Regulatory Commission, "NRC Non-Light Water Reactor (Non-LWR) Vision and Strategy, Volume 1 – Computer Code Suite for Non-LWR Plant Systems Analysis," Rev 1, 2020.
- [88] U.S. Nuclear Regulatory Commission, "NRC Non-Light Water Reactor (Non-LWR) Vision and Strategy, Volume 2 – Fuel Performance Analysis for Non-LWRs," 2020.
- [89] J. Tomon, "Non-LWR Radiological Protection Efforts," Presentation at the 2019 Fall RAMP User's Meeting, 2019.
- [90] U.S. Nuclear Regulatory Commission, "SNAP/RADTRAD 4.0: Description of Models and Methods," NUREG/CR-7220, 2016.
- [91] (June 26, 2020). *RADTRAD-NAI: RADionuclide Transport Removal and Dose Estimation*. Available: <https://www.numerical.com/software/radtrad/index.php>
- [92] J. M. Beck and L. F. Pincock, "High Temperature Gas-cooled Reactors Lessons Learned Applicable to the Next Generation Nuclear Plant," Idaho National Laboratory, INL/EXT-10-19329 Revision 1, April 2011.
- [93] 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.
- [94] P. Lowry, "Scoping Analysis of Source Term and Functional Containment Attenuation Factors," Idaho National Laboratory, INL/EXT-11-24034 Revision 2, October 2012.
- [95] 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.
- [96] M. Holbrook, "Next Generation Nuclear Plant Licensing Basis Event Selection White Paper," Idaho National Laboratory, INL/EXT-10-19521, September 2010.

- [97] "X-energy Xe-100 Reactor Initial NRC Meeting," ed. Greenbelt, MD: X-Energy, LLC, 2018.
- [98] B. Beeny, "MELCOR Workshop: High Temperature Gas Cooled Reactor Modeling in MELCOR," Sandia National Laboratories, Albuquerque, NM, SAND2018-6112PE, June 2018.
- [99] P. Vegendla, R. Hu, and L. Zou, "Multi-Scale Modeling of Thermal-Fluid Phenomena Related to Loss of Forced Circulation Transient in HTGRs," Argonne National Laboratory, ANL-19/35, 2019.
- [100] M. F. Young, R. O. Gauntt, H. Esmaili, S. Basu, R. Lee, and S. Rubin, "MELCOR Fission Product Release Model for HTGRs," Sandia National Laboratories, Albuquerque, NM, SAND2010-0800C, February 2010.
- [101] B. Beeny, "Thermal Hydraulic Analysis of a Reduced Scale High Temperature Gas-Cooled Reactor Test Facility and its Prototype with MELCOR," M.S. Thesis, Texas A&M University, 2012.
- [102] J. Corson, "Development of MELCOR Input Techniques for High Temperature Gas-Cooled Reactor Analysis," M.S. Thesis, Texas A&M University, 2010.
- [103] C. Jin and C. Huh, "MELCOR Model Development of High Temperature Gas-cooled Reactor," presented at the 2013 Spring Meeting of the KNS, Kwangju, Republic of Korea, 2013.
- [104] J. Kalilainen, "HTGR simulations in PSI using MELCOR 2.2," presented at the 10th Meeting of the European MELCOR User Group (EMUG), Zagreb, Croatia, 2018.
- [105] F. Bostelmann, C. Celik, M. L. Williams, R. J. Ellis, G. Ilas, and W. A. Wieselquist, "SCALE capabilities for high temperature gas-cooled reactor analysis," *Annals of Nuclear Energy*, vol. 147, 2020.
- [106] M. D. DeHart and A. P. Ulses, "A Code-to-Code Benchmark for High-Temperature Gas-Cooled Reactor Fuel Element Depletion," Organisation for Economic Co-Operation and Development, No. NEA-NSC-R--2019-1, February 2019.
- [107] D. Schappel, K. Terrani, L. L. Snead, and B. D. Wirth, "Modeling radionuclide release of TRISO bearing fuel compacts during post-irradiation annealing tests," *Nuclear Engineering and Design*, vol. 357, 2020.
- [108] H. Nabielek, W. Kühnlein, W. Schenk, W. Heit, A. Christ, and H. Ragoss, "Development of advanced HTR fuel elements," *Nuclear Engineering and Design*, vol. 121, no. 2, pp. 199-210, 1990.
- [109] D. A. Petti, J. Buongiorno, J. T. Maki, R. R. Hobbins, and G. K. Miller, "Key differences in the fabrication, irradiation and high temperature accident testing of US and German TRISO-coated particle fuel, and their implications on fuel performance," *Nuclear Engineering and Design*, vol. 222, no. 2-3, pp. 281-297, 2003.

- [110] J. J. Powers and B. D. Wirth, "A review of TRISO fuel performance models," *Journal of Nuclear Materials*, vol. 405, no. 1, pp. 74-82, 2010.
- [111] H. Nickel, H. Nabielek, G. Pott, and A. W. Mehner, "Long time experience with the development of HTR fuel elements in Germany," *Nuclear Engineering and Design*, vol. 217, no. 1-2, pp. 141-151, 2002.
- [112] J. T. Maki, D. A. Petti, D. L. Knudson, and G. K. Miller, "The challenges associated with high burnup, high temperature and accelerated irradiation for TRISO-coated particle fuel," *Journal of Nuclear Materials*, vol. 371, no. 1-3, pp. 270-280, 2007.
- [113] "TRISO-Coated Particle Fuel Phenomenon Identification and Ranking Tables (PIRTs) for Fission Product Transport Due to Manufacturing, Operations, and Accidents, Vol. 1," U.S. Nuclear Regulatory Commission, NUREG/CR-6844, ML042300668, 2004.
- [114] (Sep 3). *Materials challenges for nuclear systems / Elsevier Enhanced Reader*. Available: <https://reader.elsevier.com/reader/sd/pii/S1369702110702200?token=B0890AAF75B648EAABA44FD30F9998138E7C5BD037FDD8BE84CC3C2681404811690D74DB52590144FD1FA50FDBE11D25>
- [115] P. Demkowicz *et al.*, "Preliminary results of post-irradiation examination of the AGR-1 TRISO fuel compacts," presented at the 6th International Topical Meeting on High Temperature Reactor Technology HTR2012, Tokyo, Japan, October 28 - November 1, 2012.
- [116] N. I. Tak, J. H. Lee, S. N. Lee, and C. K. Jo, "POSCA: A computer code for fission product plateout and circulating coolant activities within the primary circuit of a high temperature gas-cooled reactor," *Nuclear Engineering and Technology*, vol. 52, no. 9, pp. 1974-1982, Sep 2020.
- [117] 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.
- [118] C. W. Forsberg, P. F. Peterson, and P. S. Pickard, "Molten-Salt-Cooled Advanced High-Temperature Reactor for Production of Hydrogen and Electricity," *Nuclear Technology*, vol. 144, no. 3, pp. 289-302, 2003.
- [119] D. T. Ingersoll *et al.*, "Status of Preconceptual Design of the Advanced High-Temperature Reactor (AHTR)," Oak Ridge National Laboratory, ORNL/TM-2004/104, May 2004.
- [120] 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.
- [121] 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.

- [122] 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.
- [123] 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.
- [124] 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.
- [125] C. Andreades *et al.*, "Design Summary of the Mark-I Pebble-Bed, Fluoride Salt–Cooled, High-Temperature Reactor Commercial Power Plant," *Nuclear Technology*, vol. 195, no. 3, pp. 223-238, 2016.
- [126] 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.
- [127] 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.
- [128] C. W. Forsberg, J. D. Stempfen, M. J. Minck, and R. G. Ballinger, "Understanding and Pathways to Avoid Major Fuel Failures and Radionuclide Releases in Fluoride Salt–Cooled High-Temperature Reactor Severe Accidents," *Nuclear Technology*, vol. 194, no. 3, pp. 295-313, 2016.
- [129] M. J. Minck, "Preventing fuel failure for a beyond design basis accident in a fluoride salt cooled high temperature reactor," MS Thesis, Department of Nuclear Science & Engineering, Massachusetts Institute of Technology, 2013.
- [130] D. J. Diamond, "Phenomena Important in Molten Salt Reactor Simulations," Brookhaven National Laboratory, BNL-114869-2017-INRE, April 2018.
- [131] A. Cammi, V. Di Marcello, L. Luzzi, V. Memoli, and M. E. Ricotti, "A multi-physics modelling approach to the dynamics of Molten Salt Reactors," *Annals of Nuclear Energy*, vol. 38, no. 6, pp. 1356-1372, 2011.
- [132] 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.
- [133] Z. Guo *et al.*, "Coupled neutronics/thermal-hydraulics for analysis of molten salt reactor," *Nuclear Engineering and Design*, vol. 258, pp. 144-156, 2013.

- [134] C. Shi, M. Cheng, and G. Liu, "Development and application of a system analysis code for liquid fueled molten salt reactors based on RELAP5 code," *Nuclear Engineering and Design*, vol. 305, pp. 378-388, 2016.
- [135] M. Brovchenko *et al.*, "Design-Related Studies for the Preliminary Safety Assessment of the Molten Salt Fast Reactor," *Nuclear Science and Engineering*, vol. 175, no. 3, pp. 329-339, 2013.
- [136] 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.
- [137] 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.
- [138] G. Zhang and R. Hu, "Development of MSR Transient Safety Analysis Capability in SAM," presented at the 2018 ANS Annual Meeting, Philadelphia, PA, 2018.
- [139] 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.
- [140] 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.
- [141] 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.
- [142] Z. Li and J. Cai, "Dynamics analysis of the graphite-moderated channel-type molten salt reactors based on Serpent/NTH3D-MSR," *Annals of Nuclear Energy*, vol. 121, pp. 128-134, 2018.
- [143] B. Collins, C. Gentry, A. J. Wysocki, and R. Salko, "Molten Salt Reactor Simulations using MPACT-CTF," presented at the American Nuclear Society Annual Meeting, San Francisco, CA, June 11-15, 2017.
- [144] 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.
- [145] B. R. Betzler *et al.*, "Fuel cycle and neutronic performance of a spectral shift molten salt reactor design," *Annals of Nuclear Energy*, vol. 119, pp. 396-410, 2018.
- [146] C. Gentry, B. R. Betzler, and B. Collins, "Initial Benchmarking of ChemTriton and MPACT MSR Modeling Capabilities," presented at the American Nuclear Society Winter Meeting, Washington D.C., October 29 - November 2, 2017.

- [147] B. Collins, A. Graham, W. Dawn, R. Salko, N. Smith, and C. Gentry, "Coupled Simulations of Fast-Spectrum Molten Salt Reactors," presented at the PHYSOR 2018 Conference, Cancun, Mexico, April 22-26, 2018.
- [148] A. Graham, B. Collins, R. Salko, Z. Taylor, and C. Gentry, "Development of Molten Salt Reactor Modeling and Simulation Capabilities in VERA," presented at the Global/Top Fuel 2019, Seattle, WA, September 22-27, 2019.
- [149] R. Salko, A. Graham, R. A. Lefebvre, and B. Langley, "Demonstration of MSR Salt Property Database Operation with a Reactor Analysis Tool," Oak Ridge National Laboratory, ORNL/TM-2019/1362, September 2019.
- [150] Z. Taylor, "Implementation of Multi-phase Species Transport into VERA-CS for Molten Salt Reactor Analysis," M.S. Thesis, Department of Nuclear Engineering, University of Tennessee, Knoxville, 2019.
- [151] 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.
- [152] P. J. Vicente-Valdez, B. R. Betzler, W. A. Wieselquist, and M. Fratoni, "Modeling Molten Salt Reactor Fission Product Removal with SCALE," Oak Ridge National Laboratory, ORNL/TM-2019/1418, February 2020.
- [153] 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*, 2020.
- [154] T. Price, O. Chvala, and G. Bereznai, "A dynamic model of xenon behavior in the Molten Salt Reactor Experiment," *Annals of Nuclear Energy*, vol. 144, 2020.
- [155] V. Singh, "Study of the Dynamic Behavior of Molten-Salt Reactors," M.S. Thesis, Department of Nuclear Engineering, University of Tennessee, Knoxville, 2019.
- [156] V. Singh, A. M. Wheeler, M. R. Lish, O. Chvála, and B. R. Upadhyaya, "Nonlinear dynamic model of Molten-Salt Reactor Experiment – Validation and operational analysis," *Annals of Nuclear Energy*, vol. 113, pp. 177-193, 2018.
- [157] V. Singh, M. R. Lish, A. M. Wheeler, O. Chvála, and B. R. Upadhyaya, "Dynamic Modeling and Performance Analysis of a Two-Fluid Molten-Salt Breeder Reactor System," *Nuclear Technology*, vol. 202, no. 1, pp. 15-38, 2018.
- [158] V. Singh, A. M. Wheeler, B. R. Upadhyaya, O. Chvála, and M. S. Greenwood, "Plant-level dynamic modeling of a commercial-scale molten salt reactor system," *Nuclear Engineering and Design*, vol. 360, 2020.
- [159] M. S. Greenwood, B. Betzler, A. L. Qualls, J. W. Bae, and L. G. Worrall, "Dynamic System Models for Informing Licensing and Safeguards Investigations of Molten Salt Reactors - Rev1," Oak Ridge National Laboratory, ORNL/TM-2018/876, Rev.1, July 2019.

- [160] 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, 2019.
- [161] 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.
- [162] A. Alfonsi, C. Rabiti, D. Mandelli, J. Cogliati, R. Kinoshita, and A. Naviglio, "RAVEN and Dynamic Probabilistic Risk Assessment: Software overview," presented at the European Safety and Reliability Conference ESREL, Wroclaw, Poland, September 14-18, 2014.
- [163] M. Fratoni, D. Shen, G. Ilas, and J. Powers, "Molten Salt Reactor Experiment Benchmark Evaluation (Project 16-10240)," University of California, Berkeley, 2020.
- [164] D. Shen, "Validations of Computational Codes of Molten Salt Reactors," PhD Thesis, Department of Nuclear Engineering, University of California, Berkeley, 2019.
- [165] T. Fei, B. Feng, and F. Heidet, "Molten salt reactor core simulation with PROTEUS," *Annals of Nuclear Energy*, vol. 140, 2020.
- [166] J. P. Carter and R. A. Borrelli, "Integral molten salt reactor neutron physics study using Monte Carlo N-particle code," *Nuclear Engineering and Design*, vol. 365, 2020.
- [167] M. Tiberge *et al.*, "Results from a multi-physics numerical benchmark for codes dedicated to molten salt fast reactors," *Annals of Nuclear Energy*, vol. 142, 2020.
- [168] M. Tiberge, D. Lathouwers, and J. L. Kloosterman, "A multi-physics solver for liquid-fueled fast systems based on the discontinuous Galerkin FEM discretization," *Progress in Nuclear Energy*, vol. 127, 2020.
- [169] M. Santanoceto, M. Tiberge, Z. Perko, S. Dulla, and D. Lathouwers, "Uncertainty Quantification in Steady State Simulations of a Molten Salt System Using Polynomial Chaos Expansion Analysis," presented at the PHYSOR 2020: Transition to a Scalable Nuclear Future, Cambridge, U.K., March 29 - April 2, 2020.
- [170] 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.
- [171] A. M. Pauzi, A. Cioncolini, and H. Iacovides, "Parametric study of natural circulation flow in molten salt fuel in molten salt reactor," in *AIP Conference Proceedings*, vol. 1659, no. 1, p. 030004. AIP Publishing LLC, 2015.
- [172] K. Podila, Q. Chen, and Y. Rao, "CFD Simulations of Molten Salt Reactor Experiment Core," *Nuclear Science and Engineering*, vol. 193, no. 12, pp. 1379-1393, 2019.

- [173] (Sep 4, 2020). *Engineer innovation with CFD-focused multiphysics simulation: Simcenter STAR-CCM+*. Available:
<https://www.plm.automation.siemens.com/global/en/products/simcenter/STAR-CCM.html>
- [174] 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.
- [175] 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.
- [176] 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.
- [177] 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.
- [178] 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.
- [179] J. P. van Ravenswaay, G. P. Greyvenstein, W. M. K. van Niekerk, and J. T. Labuschagne, "Verification and validation of the HTGR systems CFD code Flownex," *Nuclear Engineering and Design*, vol. 236, no. 5-6, pp. 491-501, 2006.
- [180] Y. Yang, F. Yao, Z. Yang, S. Qiang, and Z. Jie, "Reactivity-insertion-transient analysis of a Fluoride Salt Cooled High Temperature Reactor," presented at the PHYSOR 2014 - The Role of Reactor Physics Toward a Sustainable Future, The Westin Miyako, Kyoto, Japan, September 28 - October 3, 2014.
- [181] 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.
- [182] V. Ugaz, Y. Hassan, T. Nguyen, and E. Merzari, "Final NEUP Report: Experimental and Computational Analysis of NEAMS Pebble Bed Reactors," Texas A&M University, Project 16 – 10603, December 2019.
- [183] 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.
- [184] K. Ahmed, R. O. Scarlat, and R. Hu, "Benchmark Simulation of Natural Circulation Cooling System with Salt Working Fluid Using SAM," presented at the 17th International Topical Meeting on Nuclear Reactor Thermal Hydraulics NURETH-17, Xi'an, Shaanxi, China, September 3-8, 2017.

- [185] A. J. Novak, J. W. Peterson, L. Zou, D. Andrš, R. N. Slaybaugh, and R. C. Martineau, "Validation of Pronghorn friction-dominated porous media thermal-hydraulics model with the SANA experiments," *Nuclear Engineering and Design*, vol. 350, pp. 182-194, 2019.
- [186] 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.
- [187] T. Downar, Y. Xu, T. Kozlowski, and D. Carlson, "PARCS v2.7 US NRC Core Neutronics Simulator User Manual," Purdue University, 2006.
- [188] X. Wang, "Coupled neutronics and thermal-hydraulics modeling for pebble-bed Fluoride-Salt-Cooled, High-Temperature Reactor (FHR)," PhD Thesis, Department of Nuclear Engineering, University of California, Berkeley, 2018.
- [189] "KP-FHR Mechanistic Source Term Topical Report Application (Non-Proprietary)," Kairos Power, KP-TR-012-NP, June 2020.
- [190] J. Jerden, Personal Communication, Argonne National Laboratory, 2020.
- [191] 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.
- [192] R. O. Scarlat *et al.*, "Design and licensing strategies for the fluoride-salt-cooled, high-temperature reactor (FHR) technology," *Progress in Nuclear Energy*, vol. 77, pp. 406-420, 2014.
- [193] 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.
- [194] J. W. Bae, B. R. Betzler, and A. Worrall, "Molten Salt Reactor Neutronic and Fuel Cycle Sensitivity and Uncertainty Analysis," presented at the American Nuclear Society Winter Meeting, Washington, D.C., November 17-19, 2019.
- [195] A. Rykhlevskii, B. R. Betzler, A. Worrall, and K. D. Huff, "Fuel Cycle Performance of Fast Spectrum Molten Salt Reactor Designs," presented at the International Conference on Mathematics and Computational Methods applied to Nuclear Science and Engineering (M&C 2019), Portland, OR, August 25-29, 2019.
- [196] R. J. Sheu, C. H. Chang, C. C. Chao, and Y. W. H. Liu, "Depletion analysis on long-term operation of the conceptual Molten Salt Actinide Recycler & Transmuter (MOSART) by using a special sequence based on SCALE6/TRITON," *Annals of Nuclear Energy*, vol. 53, pp. 1-8, 2013.
- [197] C. Y. Zou *et al.*, "Optimization of temperature coefficient and breeding ratio for a graphite-moderated molten salt reactor," *Nuclear Engineering and Design*, vol. 281, pp. 114-120, 2015.

- [198] C. Yu *et al.*, "Analysis of minor actinides transmutation for a Molten Salt Fast Reactor," *Annals of Nuclear Energy*, vol. 85, pp. 597-604, 2015.
- [199] A. Nuttin *et al.*, "Potential of thorium molten salt reactors: detailed calculations and concept evolution with a view to large scale energy production," *Progress in Nuclear Energy*, vol. 46, pp. 77-99, 2005.
- [200] X. Doligez, D. Heuer, E. Merle-Lucotte, M. Allibert, and V. Ghetta, "Numerical tools for Molten salt reactor simulation," presented at the GLOBAL 2009 Congress: The Nuclear Fuel Cycle: Sustainable Options and Industrial Perspectives, Paris, France, September 6-11, 2009.
- [201] D. Heuer, E. Merle-Lucotte, M. Allibert, X. Doligez, and V. Ghetta, "Simulation Tools and New Developments of the Molten Salt Fast Reactor," *Revue Générale Nucléaire*, no. 6, pp. 95-100, 2010.
- [202] E. Merle-Lucotte, D. Heuer, M. Allibert, M. Brovchenko, N. Capellan, and V. Ghetta, "Launching the thorium fuel cycle with the molten salt fast reactor," presented at the ICAPP 2011, Nice, France, May 2-5, 2011.
- [203] X. Doligez, D. Heuer, E. Merle-Lucotte, M. Allibert, and V. Ghetta, "Coupled study of the Molten Salt Fast Reactor core physics and its associated reprocessing unit," *Annals of Nuclear Energy*, vol. 64, pp. 430-440, 2014.
- [204] D. Heuer, E. Merle-Lucotte, M. Allibert, M. Brovchenko, V. Ghetta, and P. Rubiolo, "Towards the thorium fuel cycle with molten salt fast reactors," *Annals of Nuclear Energy*, vol. 64, pp. 421-429, 2014.
- [205] J. Krepel, S. Pelloni, K. Mikityuk, and P. Coddington, "EQL3D: ERANOS based equilibrium fuel cycle procedure for fast reactors," *Annals of Nuclear Energy*, vol. 36, no. 5, pp. 550-561, 2009.
- [206] C. Fiorina *et al.*, "Investigation of the MSFR core physics and fuel cycle characteristics," *Progress in Nuclear Energy*, vol. 68, pp. 153-168, 2013.
- [207] C. Fiorina, "The Molten Salt Fast Reactor as a Fast-Spectrum Candidate for Thorium Implementation," PhD Thesis, Energy and Nuclear Science and Technology, Politecnico Di Milano, 2013.
- [208] M. Aufiero, "Development of Advanced Simulation Tools for Circulating-Fuel Reactors," PhD Thesis, Energy and Nuclear Science and Technology, Politecnico Di Milano, 2014.
- [209] M. Aufiero, A. Cammi, C. Fiorina, J. Leppänen, L. Luzzi, and M. E. Ricotti, "An extended version of the Serpent-2 code to investigate fuel burn-up and core material evolution of the Molten Salt Fast Reactor," *Journal of Nuclear Materials*, vol. 441, no. 1-3, pp. 473-486, 2013.

- [210] B. Hombourger, J. Křepel, and A. Pautz, "The EQL0D fuel cycle procedure and its application to the transition to equilibrium of selected molten salt reactor designs," *Annals of Nuclear Energy*, vol. 144, 2020.
- [211] A. A. Kasam, "Conceptual design of a breed & burn molten salt reactor," PhD Thesis, Department of Engineering, University of Cambridge, 2018.
- [212] (2020). *WIMS - A General Purpose Reactor Physics Program for Core Physics Calculations*. Available: <https://www.answerssoftwareservice.com/wims/>
- [213] B. Lindley *et al.*, "Developments within the WIMS Reactor Physics Code for Whole Core Calculations," presented at the M&C 17: International Conference on Mathematics & Computational Methods Applied to Nuclear Science & Engineering, Jeju, Korea, April 16-20, 2017.
- [214] A. Ahmad, E. B. McClamrock, and A. Glaser, "Neutronics calculations for denatured molten salt reactors: Assessing resource requirements and proliferation-risk attributes," *Annals of Nuclear Energy*, vol. 75, pp. 261-267, 2015.
- [215] J. Park, Y. Jeong, H. C. Lee, and D. Lee, "Whole core analysis of molten salt breeder reactor with online fuel reprocessing," *International Journal of Energy Research*, pp. n/a-n/a, 2015.
- [216] Y. Jeong, J. Park, H. C. Lee, and D. Lee, "Equilibrium core design methods for molten salt breeder reactor based on two-cell model," *Journal of Nuclear Science and Technology*, vol. 53, no. 4, pp. 529-536, 2016.
- [217] A. G. Nelson, J. A. Stillman, F. Heidet, and E. H. Wilson, "The ADDER Software for Reactor Depletion and Fuel Management Analysis," presented at the 2020 American Nuclear Society Winter Meeting (accepted), Virtual, November, 2020.
- [218] 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.
- [219] R. J. Kedl, "Migration of a class of fission products (noble metals) in the Molten-Salt Reactor Experiment," Oak Ridge National Laboratory, ORNL-TM-3884, December 1972.
- [220] J. D. Stempien, "Tritium transport, corrosion, and Fuel performance modeling in the Fluoride Salt-Cooled High-Temperature Reactor," PhD Thesis, Nuclear Science & Engineering, Massachusetts Institute of Technology, 2015.
- [221] D. E. Holcomb, W. P. Poore, and G. F. Flanagan, "MSR Fuel Salt Qualification Methodology," Oak Ridge National Laboratory, ORNL/TM-2020/1576, July 2020.
- [222] E. Capelli, "Thermodynamic Characterization of Salt Components for Molten Salt Reactor Fuel," PhD Thesis, Delft University of Technology, 2016.

- [223] 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.
- [224] E. L. Compere, E. G. Bohlmann, S. S. Kirslis, F. F. Blankenship, and G. W. R., "Fission Product Behavior in the Molten Salt Reactor Experiment," Oak Ridge National Laboratory, ORNL-4865 October 1975.
- [225] J. Zhang *et al.*, "Redox potential control in molten salt systems for corrosion mitigation," *Corrosion Science*, vol. 144, pp. 44-53, 2018.
- [226] J. Zhang, "Impurities in Primary Coolant Salt of FHRs: Chemistry, Impact, and Removal Methods," *Energy Technology*, vol. 7, no. 10, 2019.
- [227] 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.
- [228] C. Robelin, "Models for the thermodynamic properties of molten salt systems: Perspectives for ionic liquids," *Fluid Phase Equilibria*, vol. 409, pp. 482-494, 2016.
- [229] M. Blander, "Contributions to molten salt chemistry by Ole J. Kleppa," *Journal of Alloys and Compounds*, vol. 321, no. 2, pp. 164-168, 2001.
- [230] J. W. McMurray and T. M. Besmann, "Thermodynamic Modeling of Nuclear Fuel Materials," in *Handbook of Materials Modeling*, 2020, pp. 2335-2363.
- [231] J. W. McMurray *et al.*, "Multi-Physics Simulations for Molten Salt Reactor Evaluation: Chemistry Modeling and Database Development," ORNL/SPR-2018/864, June 2018.
- [232] "Aircraft Nuclear Propulsion Project Quarterly Progress Report for Period Ending September 10, 1956," Oak Ridge National Laboratory, ORNL-2157, 1956.
- [233] 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.
- [234] E. Capelli, O. Beneš, and R. J. M. Konings, "Thermodynamics of soluble fission products cesium and iodine in the Molten Salt Reactor," *Journal of Nuclear Materials*, vol. 501, pp. 238-252, 2018.
- [235] 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.
- [236] 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.

- [237] J. W. Geringer, T. Koyanagi, Y. Lee, and Y. Katoh, "SiC/SiC Composites Technology Gap Analysis for Molten Salt Reactors," ORNL/TM-2018/842, June 2018.
- [238] D. R. Olander, "Redox condition in molten fluoride salts - Definition and control," *Journal of Nuclear Materials*, vol. 300, pp. 270-272, 2002.
- [239] M. Blander, W. R. Grimes, N. V. Smith, and G. M. Watson, "Solubility of Noble Gases in Molten Fluorides. II. In the LiF–NaF–LF Eutectic Mixtures. ," *The Journal of Physical Chemistry*, vol. 63, no. 7, pp. 1164-1167, 1959.
- [240] W. R. Grimes, N. V. Smith, and G. M. Watson, "Solubility of Noble Gases in Molten Fluorides. I. In Mixtures of NaF–ZrF₄ (53–47 Mole%) and NaF–ZrF₄–UF₄ (50–46–4 Mole%)." *The Journal of Physical Chemistry*, vol. 62, no. 7, pp. 862-866, 1958.
- [241] G. M. Watson, R. B. Evans III, W. R. Grimes, and N. V. Smith, "Solubility of Noble Gases in Molten Fluorides," *Journal of Chemical and Engineering Data*, vol. 7, no. 2, pp. 285-287, 1962.
- [242] J. H. Shaffer, W. R. Grimes, and G. M. Watson, "Solubility of HF in Molten Fluorides. I. In Mixtures of NaF–ZrF₄," *The Journal of Physical Chemistry*, vol. 63, no. 12, pp. 1999-2002, 1959.
- [243] J. H. Shaffer, W. R. Grimes, and G. M. Watson, "Boron Trifluoride as a Soluble Poison in Molten Salt Reactor Fuels," *Nuclear Science and Engineering*, vol. 12, no. 3, pp. 337-340, 1962.
- [244] A. Borodziński, A. Sokołowski, and L. Suski, "Solubility of carbon dioxide, nitrogen, and argon in molten zinc chloride. ," *The Journal of Chemical Thermodynamics*, vol. 7, no. 7, pp. 655-660, 1975.
- [245] E. Sada, S. Katoh, H. Yoshii, T. Yamanishi, and A. Nakanishi, "Performance of the gas bubble column in molten salt systems. ," *Industrial & Engineering Chemistry Process Design and Development*, vol. 23, no. 1, pp. 151-154, 1984.
- [246] Y. Kanai, K.-i. Fukunaga, K. Terasaka, and S. Fujioka, "Mass transfer in molten salt and suspended molten salt in bubble column," *Chemical Engineering Science*, vol. 100, pp. 153-159, 2013.
- [247] R. J. Kedl and A. Houtzeel, "Development of a Model for Computing ¹³⁵Xe Migration in the MSRE," Oak Ridge National Laboratory, ORNL-4069, June 1967.
- [248] H. W. Kohn, "Bubbles, Drops, and Entrainment in Molten Salts," Oak Ridge National Laboratory, ORNL-TM-2373, December 1968.
- [249] P. Bajpai, S. Lorenzi, and A. Cammi, "A multiphysics model for analysis of inert gas bubbles in Molten Salt Fast Reactor," *The European Physical Journal Plus*, vol. 135, no. 6, 2020.
- [250] K. Sridharan and T. R. Allen, "Corrosion in molten salts," in *Molten salts chemistry*: Elsevier, 2013, pp. 241-267.

- [251] S. S. Raiman and S. Lee, "Aggregation and data analysis of corrosion studies in molten chloride and fluoride salts," *Journal of Nuclear Materials*, vol. 511, pp. 523-535, 2018.
- [252] A. M. Kruizenga, "Corrosion Mechanisms in Chloride and Carbonate Salts," Sandia National Laboratories, SAND2012-7594, September 2012.
- [253] R. B. Lindauer, "Processing of the MSRE Flush and Fuel Salts," Oak Ridge National Laboratory, ORNL-TM-2578, August 1969.
- [254] J. J. Lee, J. D. Arregui-Mena, C. I. Contescu, T. D. Burchell, Y. Katoh, and S. K. Loyalka, "Protection of graphite from salt and gas permeation in molten salt reactors," *Journal of Nuclear Materials*, vol. 534, 2020.
- [255] M. Blander, "Aircraft Nuclear Propulsion Program Quarterly Progress Report for Period Ending December 31, 1957," Oak Ridge National Laboratory, ORNL-2440, p. 87, 1958.
- [256] R. B. Evans, "Aircraft Nuclear Propulsion Program Quarterly Progress Report for Period Ending December 31, 1957," Oak Ridge National Laboratory, ORNL-2440, p. 104, 1958.
- [257] R. E. Thoma, "Chemical Aspects of MSRE Operations," Oak Ridge National Laboratory, ORNL-4658, December 1971.
- [258] S. A. Walker, Z. Taylor, R. Salko, B. Collins, and W. Ji, "Noble Metal Mass Transport Model for Molten Salt Reactor Analysis in VERA-CS," presented at the M&C 2019, Portland, OR, August 25-29, 2019.
- [259] G. Cartland Glover *et al.*, "On the numerical modelling of frozen walls in a molten salt fast reactor," *Nuclear Engineering and Design*, vol. 355, 2019.
- [260] U. Rohde *et al.*, "The reactor dynamics code DYN3D – models, validation and applications," *Progress in Nuclear Energy*, vol. 89, pp. 170-190, 2016.
- [261] F. Archambeau, N. Méchitoua, and M. Sakiz, "Code Saturne: A Finite Volume Code for the computation of turbulent incompressible flows - Industrial Applications," *International Journal on Finite Volumes*, Institut de Mathématiques de Marseille, AMU, 2004, Available: <https://hal.archives-ouvertes.fr/hal-01115371/>, Accessed on.
- [262] B. H. Mahmoud *et al.*, "Turbulent heat transfer in nanoparticulate multiphase channel flows with a high Prandtl number molten salt fluid," presented at the International Conference on Particle-Based Methods. Fundamentals and Applications Particles 2019, Barcelona, Spain, Oct 28-30, 2019.
- [263] N. C. Hoyt and H. Yuan, "Thermogalvanic Modeling Capabilities to Support Engineering-Scale Corrosion Simulations," Argonne National Laboratory, ANL/CFCT-20/33, 2020.

- [264] M. T. Farmer, "The MELTSPREAD Code for Modeling of Ex-Vessel Core Debris Spreading Behavior Code Manual – Version3-beta," Argonne National Laboratory, ANL/NE-17/20, September 2017.
- [265] M. T. Farmer, "The CORQUENCH Code for Modeling of Ex-Vessel Corium Coolability under Top Flooding Conditions Code Manual – Version4.1-beta," Argonne National Laboratory, ANL-18/22, August 2018.
- [266] F. J. Peretz, "Identification and Evaluation of Alternatives for the Disposition of Fluoride Fuel and Flush Salts from the Molten Salt Reactor Experiment at ORNL," ORNL/ER-380, August 1996.
- [267] E. R. Buckle, "Studies on the freezing of pure liquids II. The kinetics of homogeneous nucleation in supercooled liquids," *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences*, vol. 261, no. 1305, pp. 189-196, 1961.
- [268] E. R. Buckle and A. R. J. P. Ubbelohde, "Studies on the freezing of pure liquids I. Critical supercooling in molten alkali halides," *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences*, vol. 259, no. 1298, pp. 325-340, 1960.
- [269] E. R. Buckle and A. R. J. P. Ubbelohde, "Studies on the freezing of pure liquids III. Homogeneous nucleation in molten alkali halides," *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences*, vol. 261, no. 1305, pp. 197-206, 1961.
- [270] E. R. Buckle, "Nucleation and growth of caesium iodide aerosols," *Journal of Aerosol Science*, vol. 22, no. 2, pp. 135-147, 1991.
- [271] R. C. Daniel *et al.*, "Large-Scale Spray Releases: Additional Aerosol Test Results," Pacific Northwest National Laboratory, PNNL-22415, WTP-RPT-221 Rev. 0, August 2013.
- [272] P. P. Schonewill *et al.*, "Large-Scale Spray Releases: Initial Aerosol Test Results," Pacific Northwest National Laboratory, PNNL-21333, WTP-RPT-217 Rev. 0, December 2012.
- [273] T. Ginsberg, "Aerosol Generation by Liquid Breakup Resulting from Sparging of Molten Pools of Corium by Gases Released During Core/Concrete Interactions," *Nuclear Science and Engineering*, vol. 89, no. 1, pp. 36-48, 1985.
- [274] A. N. Williams, "Measurement of Rare Earth and Uranium Elements Using Laser-Induced Breakdown Spectroscopy in an Aerosol System for Nuclear Safeguards Applications," PhD Thesis, Mechanical and Nuclear Engineering, Virginia Commonwealth University, 2016.
- [275] A. N. Williams and S. Phongikaroon, "Laser-Induced Breakdown Spectroscopy (LIBS) in a Novel Molten Salt Aerosol System," *Appl Spectrosc*, vol. 71, no. 4, pp. 744-749, Apr 2016.

- [276] A. N. Williams and S. Phongikaroon, "Laser-Induced Breakdown Spectroscopy (LIBS) Measurement of Uranium in Molten Salt," *Appl Spectrosc*, vol. 72, no. 7, pp. 1029-1039, Jul 2018.
- [277] 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.
- [278] J. D. Stempien, R. G. Ballinger, and C. W. Forsberg, "An integrated model of tritium transport and corrosion in Fluoride Salt-Cooled High-Temperature Reactors (FHRs) – Part I: Theory and benchmarking," *Nuclear Engineering and Design*, vol. 310, pp. 258-272, 2016.
- [279] S. Lam, "Managing tritium inventory and release with carbon materials in a fluoride salt-cooled high-temperature reactor," M.S. Thesis, Nuclear Science & Engineering, Massachusetts Institute of Technology, 2017.
- [280] 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.
- [281] J. D. Rader, M. S. Greenwood, and P. W. Humrickhouse, "Verification of Modelica-Based Models with Analytical Solutions for Tritium Diffusion," *Nuclear Technology*, vol. 203, no. 1, pp. 58-65, 2018.
- [282] U. S. DOE. (2019). *FY19 ARD-19-1-17578 Summary Abstract: Modeling and Simulation Development Pathways to Accelerating KP-FHR Licensing*. Available: <https://www.energy.gov/sites/prod/files/2019/04/f61/Kairos%20FY2019-1%20Summary-Abstract%20ARD-19-1-17578.pdf>
- [283] X. Wu, "Design of a Tritium Mitigation and Control System for Fluoride-salt-cooled High-temperature Reactor Systems," M.S. Thesis, The Ohio State University, 2016.
- [284] S. Zhang, X. Wu, S. Shi, X. Sun, R. Christensen, and G. Yoder, "A coupled heat transfer and tritium mass transport model for a double-wall heat exchanger design for FHRs," *Annals of Nuclear Energy*, vol. 122, pp. 328-339, 2018.
- [285] 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.
- [286] H. Qin, C. Wang, S. Qiu, D. Zhang, W. Tian, and G. H. Su, "Study of tritium transport characteristics in a transportable fluoride-salt-cooled high-temperature reactor," *International Journal of Energy Research*, vol. 42, no. 4, pp. 1536-1550, 2018.
- [287] C. Wang, H. Qin, D. Zhang, W. Tian, S. Qiu, and G. H. Su, "Numerical study of tritium transport characteristics in Thorium Molten Salt Reactor with Solid Fuel (TMSR-SF)," *Nuclear Engineering and Design*, vol. 335, pp. 391-399, 2018.

- [288] H. Qin, C. Wang, D. Zhang, W. Tian, S. Qiu, and G. H. Su, "Transient analysis of tritium transport characteristics in fluoride-salt-cooled high-temperature reactor," *Progress in Nuclear Energy*, vol. 117, 2019.
- [289] Y. Zeng *et al.*, "Development of the Tritium Transport Analysis Code for the Thorium-Based Molten Salt Reactor," *Nuclear Technology*, vol. 203, no. 1, pp. 48-57, 2018.
- [290] B. J. Merrill, J. L. Jones, and D. F. Holland, "TMAP/Mod 1: Tritium Migration Analysis Program code description and user's manual," EG and G Idaho, Inc, (No. EGG-EP--7407), 1986.
- [291] B. J. Merrill, P. W. Humrickhouse, and M. Shimada, "Recent development and application of a new safety analysis code for fusion reactors," *Fusion Engineering and Design*, vol. 109-111, pp. 970-974, 2016.
- [292] 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.
- [293] R. C. Robertson, "Conceptual Design Study of a Single-Fluid Molten-Salt Breeder Reactor," Oak Ridge National Laboratory, ORNL-4541, June 1971.
- [294] 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.
- [295] A. Rykhlevskii, "Advanced online fuel reprocessing simulation for thorium-fueled molten salt breeder reactor," M.S. Thesis, Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign, 2018.
- [296] A. Rykhlevskii, J. W. Bae, and K. D. Huff, "Modeling and simulation of online reprocessing in the thorium-fueled molten salt breeder reactor," *Annals of Nuclear Energy*, vol. 128, pp. 366-379, 2019.
- [297] E. Davidson, B. Betzler, R. Gregg, and A. Worrall, "Modeling a fast spectrum molten salt reactor in a systems dynamics fuel cycles code," *Annals of Nuclear Energy*, vol. 133, pp. 409-424, 2019.
- [298] B. J. Riley, J. McFarlane, G. D. Del Cul, J. D. Vienna, C. I. Contescu, and C. W. Forsberg, "Molten salt reactor waste and effluent management strategies: A review," *Nuclear Engineering and Design*, vol. 345, pp. 94-109, 2019.
- [299] 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.
- [300] 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.

- [301] 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.
- [302] T. H. Fanning, A. J. Brunett, and T. Sumner, eds., "The SAS4A/SASSYS-1 Safety Analysis Code System: User's Guide," Argonne National Laboratory, ANL/NE-16/19, 2017.
- [303] A. Tentner, A. Karahan, and S. H. Kang, "Overview of the SAS4A Metallic Fuel Models and Extended Analysis of a Postulated Severe Accident in the Prototype Gen-IV Sodium-Cooled Fast Reactor," *Nuclear Technology*, vol. 206, pp. 242-254, 2019.
- [304] R. Hu and Y. Yu, "A Computationally Efficient Method for Full-Core Conjugate Heat Transfer Modeling of Sodium Fast Reactors," *Nuclear Engineering and Design*, vol. 308, pp. 182-193, 2016.
- [305] R. Hu, Y. Zhu, and A. Kraus, "Advanced Model Developments in SAM for Thermal Stratification Analysis during Reactor Transients," ANL/NSE-18/7, 2018.
- [306] M. Travis, R. Hu, and G. Zhang, "Uncertainty Quantification on SAM Simulations of EBR-II Loss-of-Flow Tests," *Proceedings of NURETH-18*, 2019.
- [307] R. Hu and T. Sumner, "Benchmark Simulations of the Thermal-Hydraulic Response during EBR-II Inherent Safety Tests using SAM," *Proceedings of ICAPP 16*, 2016.
- [308] L. Yang and R. Hu, "Benchmark Modeling and Simulation of the FFTF LOFWOS Test #13 using SAM," ANL/NSE-20/8, 2020.
- [309] D. Grabaskas, M. Bucknor, and J. Jerden, "Simplified Radionuclide Transport (SRT) Code: Users Manual," ANL-SRT-4, Rev 2.0, 2020.
- [310] D. 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.
- [311] K. K. Murata, D. E. Carroll, K. D. Bergeron, and G. D. Valdez, "CONTAIN LMR/1B-Mod. 1, A Computer Code for Containment Analysis of Accidents in Liquid-Metal-Cooled Nuclear Reactors," Sandia National Laboratories, SAND91-1490, 1993.
- [312] D. Toyohara, S. Ohno, T. Matsuki, H. Hamada, and S. Miyahara, "In-Vessel Source Term Analysis Code TRACER Version 2.3, User's Manual," JNC TN9520 2004-004, 2005.
- [313] L. E. Herranz, L. Lebel, F. Mascari, and C. Spengler, "Progress in Modeling In-containment Source Term with ASTEC-Na," *Annals of Nuclear Energy*, vol. 112, pp. 84-93, 2018.
- [314] V. Matuzas, L. Ammirabile, L. Cloarec, D. Lemasson, P.-M. S., and A. Ponomarev, "Extension of ASTEC-Na Capabilities for Simulation Reactivity Effects in Sodium Cooled Fast Reactor," *Annals of Nuclear Energy*, vol. 119, pp. 440-453, 2018.

- [315] K. A. El-Sheikh and D. Hahn, "Effectiveness of natural phenomena in reducing the source term and consequences of severe accidents in the ALMR," in *Proc. 3rd Int. Conf. Nuclear Engineering (ICONE-3)*, 1995, pp. 17-23.
- [316] D. 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.
- [317] C. Arnold, J. Galloway, and C. Unal, "Modeling Lanthanide Transport in Metal Fuels with BISON," *Proceedings of GLOBAL 2015*, 2015.
- [318] R. Schram, E. Cordfunke, and M. Huntelaar, "Source Term Calculations of the ALMR," ECN-R-95-021, 1995.
- [319] M. Bucknor, M. T. Farmer, and D. Grabaskas, "An Assessment of Fission Product Scrubbing in Sodium Pools Following a Core Damage Event in a Sodium Cooled Fast Reactor," in *International Conference on Fast Reactors and Related Fuel Cycles: Next Generation Nuclear Systems for Sustainable Development (FR 17)*, Yekaterinburg, RU, 2017.
- [320] University of Wisconsin and Argonne National Laboratory, "NEUP Project 18-14908: Experimental Measurements of Fission Product Retention in Liquid Sodium," 2018.
- [321] M. Naritomi, M. Murata, and Y. Yoshida, "Monitoring Technique for Radioiodine in Sodium Aerosol," *Proceedings of the 3rd International Conference on Radiation Protection*, vol. 2:388, 1974.
- [322] Fauske & Associates LLC, "IDCOR Technical Report 11.7: FAI Aerosol Correlation," 1985.
- [323] P. Beiriger, J. Hopenfeld, M. Silberberg, J. Baurmash, and R. Koontz, "SOFIRE II User Report," AI-AEC-13055, 1973.
- [324] S. Tsai, "The NACOM Code for Analysis of Postulated Sodium Spray Fires in LMFBRs," NUREG/CR-1405, BNL-NUREG-51180, 1980.
- [325] K. K. Murata *et al.*, "User's Manual for CONTAIN 1.1: A Computer Code for Severe Nuclear Reactor Accident Containment Analysis," SAND87-2309, 1989.
- [326] S. Kawahara, T. Murata, H. Yusa, and N. Sagawa, "Release of Sodium and Radioisotopes into Air Associated with Sodium Combustion," *Journal of Nuclear Science and Technology*, vol. 13, no. 7, pp. 382-388, 1976.
- [327] "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.
- [328] A. J. Brunett, M. Bucknor, A. Dix, D. Grabaskas, J. Jerden, and D. O'Grady, "Development of Integrated Mechanistic Source Term Assessment Software for Liquid Metal Reactors," *Proceedings of the ANS Winter Conference 2019*, 2019.

- [329] T. Q. Hua *et al.*, "Development of Mechanistic Source Term Analysis Tool SAS4A-FATE for Lead- and Sodium-Cooled Fast Reactors," *Nuclear Technology*, vol. 206, no. 2, pp. 206-217, 2020.
- [330] B. D. Middleton, E. J. Parma Jr, T. J. Olivier, J. Phillips, and J. L. LaChance, "The development of a realistic source term for sodium-cooled fast reactors: Assessment of current status and future needs," Sandia National Laboratories, SAND2011-3404, June 2011.
- [331] N. E. Bixler. (2020, June 26). *About MACCS*. Available: <https://maccs.sandia.gov/maccs.aspx>
- [332] (June 26). *RASCAL Overview*. Available: <https://ramp.nrc-gateway.gov/RASCAL%20Overview>
- [333] (September 14). *Radiation Protection Computer Code Analysis and Maintenance program (RAMP)*. Available: <https://ramp.nrc-gateway.gov/>
- [334] USEPA, "User's Guide for the AMS/EPA Regulatory Model—AERMOD, EPA-454/B-19-027," ed: Office of Air Quality Planning and Standards Washington, DC, USA, 2004.
- [335] J. Ramsdell Jr and C. Simonen, "Atmospheric relative concentrations in building wakes," Nuclear Regulatory Commission, Washington, DC (United States), NUREG/CR-6331, PNNL-10521, 1997.
- [336] S. Hoe *et al.*, "ARGOS decision support system for emergency management," in *12th International Congress of the International Radiation Protection Association*, 2009: Argentine Radiation Protection Society.
- [337] (2018, September 14). *CALPUFF Modeling System, 2014-2018*. Available: <https://www.src.com/>
- [338] (September 14). *HotSpot and EPIcode*. Available: <https://narac.llnl.gov/tools/hotspot-epicode>
- [339] I. Pisso *et al.*, "The Lagrangian particle dispersion model FLEXPART version 10.4," *Geoscientific Model Development*, vol. 12, no. 12, pp. 4955-4997, 2019.
- [340] B. Napier, "GENII Version 2 Users' Guide," Pacific Northwest National Laboratory, Richland, WA, PNNL-14583 Rev. 4, 2012.
- [341] B. Biwer, D. LePoire, S. Kamboj, Y.-S. Chang, and C. Sun, "Technical Manual and User's Guide for MILDOS, Version 4.1," Argonne National Laboratory/U.S. NRC, Argonne, IL, ANL/EVS-18/5, NUREG/CR-7258, 2018.
- [342] S. G. Homann and F. Aluzzi, "HotSpot health physics codes version 3.0 user's guide," *Lawrence Livermore National Laboratory, CA, USA*, 2013.
- [343] (2019, September 14). *OpenFOAM: Then open source CFD toolbox*. Available: <https://www.openfoam.com/>

- [344] (2020, September 14). *SNAP/RADTRAD Overview*. Available: <https://ramp.nrc-gateway.gov/content/snpradtrad-overview>
- [345] W. Raskob, "Brief description of RODOS installation PV6. 0," *Forschungszentrum PO Box*, vol. 3640, 2004.
- [346] R. Sykes, S. Parker, D. Henn, and R. Gabruk, "SCIPUFF—A Generalized Dispersion Model," in *Air Pollution Modeling and Its Application XI*: Springer, 1996, pp. 425-432.
- [347] M. Brown. (September 14, 2020). *QUIC Atmospheric Dispersion Modeling System, 2010-2011*. Available: <https://www.lanl.gov/projects/quic/>
- [348] D. Clayton and N. E. Bixler, "Assessment of the MACCS Code Applicability for Nearfield Consequence Analysis," Sandia National Laboratories, Albuquerque, NM, SAND2020-2609, 2020.
- [349] E. Eimutis and M. Konicek, "Derivations of continuous functions for the lateral and vertical atmospheric dispersion coefficients," *Atmospheric Environment (1967)*, vol. 6, no. 11, pp. 859-863, 1972.
- [350] W. Snell and R. Jubach, "Technical Basis for Regulatory Guide 1.145," Atmospheric Dispersion Models for Potential Accident Consequence Assessments at Nuclear Power Plants," Division of Health, Siting and Waste Management, U.S. Nuclear Regulatory Commission, NUREG/CR-2260, NUS-3854, 1981.
- [351] "Atmospheric dispersion models for potential accident consequence assessment at nuclear power plants," US Nuclear Regulatory Commission, Regulatory Guide 1.145 Rev. 1, 1983.
- [352] N. E. Bixler, "Evaluation of MACCS Model/Data Improvements Needed to Support Consequence Analyses of Advanced Reactors," Sandia National Laboratories, Albuquerque, NM, Internal Report, 2017.

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