TERRESTRIAL
ENERGY USA

Uncertainty Quantification Methodology for
Calculation of IMSR® Off-Gas Source Term
Technical Report

Revision 1

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Abstract

This document presents a methodology for performing uncertainty quantification analysis for the Terrestrial Energy Integral Molten Salt Reactor (IMSR®) source term based on coupled modeling of neutronics, thermal hydraulics, nuclide mass transport, and chemical speciation. The result of this effort will be a general, coupled, modeling capability for MSR systems, including the capability to propagate uncertainties in model input parameters for purposes of mass accountancy and the calculation of source term. The capability will be applicable to the fluoride fuel salt of the IMSR® and will be useful in the analysis of off-gas release during steady-state and accident scenarios. In addition, these tools also provide a high-fidelity benchmarking opportunity for the vendor and the regulator to use as a reference tool for future licensing submittals when appropriate, especially when test and operational data are not abundantly available (e.g., new methods and new design configurations).

Non-Proprietary

ACKNOWLEDGMENTS

CONTENTS

ABBREVIATIONS

CALPHAD	CALculation of PHAse Diagrams
ISMR®	Integral Molten Salt Reactor
MCMC	Markov Chain Monte Carlo
MSBR	molten salt breeder reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
NEAMS	Nuclear Energy Advanced Modeling and Simulations
ORNL	Oak Ridge National Laboratory
QoIs	quantities of interest
UQ	uncertainty quantification

NOTATION

v_l	liquid velocity
v_g	gas velocity
ϕ	neutron flux
σ	microscopic cross section
Σ_f	macroscopic fission cross section
Σ_a	macroscopic absorption cross section
β	delayed neutron fraction
β_p	delayed neutron fraction for precursor group p
ν	neutron per fission
V	neutron speed
k_{eff}	effective multiplication factor
λ_p	decay constant for precursor group p
γ_j	species j yield
λ_j	species j decay constant
D	neutron diffusion coefficient
P	pressure
T	temperature
A	flow area
g	gravitational acceleration
u_l	bulk liquid internal energy
ρ_l	bulk liquid density
F	frictional pressure drop per unit length
P'	pressure increase per unit length due to external sources (i.e., pumps)
q'''	volumetric heat generation rate
q''	wall heat flux
μ_l	dynamic viscosity
Q	volumetric flow rate
L	characteristic length
h_l	convective heat transfer coefficient
k_l	thermal conductivity
c_p	specific heat capacity
C_p	concentration for precursor group p

c_j^l	isotope j concentration in bulk liquid phase
c_j^w	isotope j concentration at wall (deposition and erosion surfaces)
c_j^s	isotope j concentration at leaching surface
c_j^b	isotope j concentration at bubbles
γ_j	fission yield fraction for isotope j
λ_j	decay constant for isotope j
σ_j	absorption cross section for isotope j
$\zeta_{i \to j}$	fraction of isotope i neutron capture producing isotope j
$\xi_{i \to j}$	fraction of isotope i decay producing isotope j
D_j^l	liquid diffusivity
Φ_j	species j, molar flux at gas-liquid interface
a_j	species j, gas-liquid interfacial area
k_j^b	mass transfer coefficient for bubbles
k_j^d	mass transfer coefficient for deposition
$k_j^{\delta\mu}$	mass transfer coefficient for leaching
k_j^e	mass transfer coefficient for erosion
a_b	bubbles interfacial area for bubbles
a_w	wall interfacial area for deposition and erosion
a_s	interfacial area for leaching
c_j^g	bulk gas concentration
D_j^g	gas diffusivity in liquid
p_j^g	bulk gas phase, species j partial pressure
p_j^i	gas phase, species j partial pressure at interface
p_j^*	gas phase, species j theoretical partial pressure in equilibrium with c_j^l
c_j^l	bulk liquid phase, species j concentration
c_j^i	liquid phase, species j concentration at interface
c_j^*	liquid phase, species j theoretical concentration in equilibrium with p_j^g
T_g	vapor temperature
c_j^g	species j, vapor molar density
ρ_g	vapor density
u_g	vapor internal energy
h_g	vapor convective heat transfer
M_j	molecular weight of species j
M_l	molecular weight of bulk liquid
V_j	sum of the diffusion volume, species j
V_l	sum of the diffusion volume, bulk liquid

ABSTRACT

This document presents a methodology for performing uncertainty quantification analysis for the Terrestrial Energy Integral Molten Salt Reactor (IMSR®) source term based on coupled modeling of neutronics, thermal hydraulics, nuclide mass transport, and chemical speciation. The result of this effort will be a general, coupled, modeling capability for MSR systems, including the capability to propagate uncertainties in model input parameters for purposes of mass accountancy and the calculation of source term. The capability will be applicable to the fluoride fuel salt of the IMSR® and will be useful in the analysis of off-gas release during steady-state and accident scenarios. In addition, these tools also provide a high-fidelity benchmarking opportunity for the vendor and the regulator to use as a reference tool for future licensing submittals when appropriate, especially when test and operational data are not abundantly available (e.g., new methods and new design configurations).

1. PURPOSE

The goal of this project is the development of a broadly applicable modeling and simulation capability and methodology for the propagation of uncertainties in model parameters of importance to molten salt reactor (MSR) source terms and off-gas systems. Oak Ridge National Laboratory (ORNL) will then use details of the specific fuel and coolant conditions of the (IMSR®) being developed by Terrestrial Energy USA (TEUSA) to demonstrate the capabilities of the methodology in calculation source terms that could be supportive of future licensing submittals. Uncertainties to be addressed include those related to nuclear data (e.g., cross sections and yields), the physical properties of the salt (e.g., solubilities and viscosities), thermochemical properties (e.g., vapor pressures), and system design parameters such as power level and fluid flow rates. The developed capability will provide the boundary conditions to off-gas systems and will be used for the determination of the source term for postulated radioactive releases important to the licensing analyses.

Off-gas properties/aspects to investigate include the following: [

.] This work will leverage codes under active development within Nuclear Energy Advanced Modeling and Simulations (NEAMS) Program of the US Department of Energy (DOE) and the NEAMS Molten Salt Thermal Properties Database. The codes that will be leveraged include Shift, for neutronics, nuclide transmutation, and nuclear cross-section uncertainty; the SAM code, for fluid transport; the Mole code, for

time-dependent species transport, and Gibbs Energy Minimizer. A primary goal of this work will be the development and exercise of the integrated capability for determining the composition and activities with uncertainties for source term and the off-gas system. This will include the propagation of uncertainties in model input parameters to establish requirements for a given off-gas system, which will also serve the purpose of providing a bound on source term under postulated accident scenarios.

2. NUCLIDE CONTRIBUTORS TO OFF-GAS SOURCE TERM

The transport and retention of radionuclides within molten salt reactor systems is highly dependent on its chemical form within the fuel salt, otherwise known as its chemical speciation, which may be comprised of gas, liquid and/or solid phases. During postulated accident scenarios, the chemical speciation of a radionuclide, its production/destruction through fission and radioactive decay will determine its transport and potential release to the environment. Chemical speciation for a given radionuclide depends on a number of critical factors including fuel salt chemical composition, fuel salt redox potential, radionuclide concentration, pressure, and temperature. Experience with the Molten Saft Reactor Experiment (MSRE) led to the characterization of fission and transmutation products as noble gases, salt-seeking elements, and noble metals (Kedl 1972). The noble gases of primary interest are Kr and Xe, which are insoluble in salt and readily form a gas phase (bubble formation). In contrast, salt seekers create non-volatile, low-volatile, and redox-dependent, low-volatile fluorides within the fuel salt.

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Table 1 displays an example list of fluoride salts and their boiling temperature that were of interest to the MSRE fuel handling. These are provided for illustrative purposes because of their low boiling temperature relative to the higher operating temperature of the TEUSA IMSR®. [

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**Table 1. Boiling Points of Fluoride Salts (Lindauer 1965)**

Salt	Boiling Temperature (°C)
CF_4	-127.8
TeF_6	-37.6
IF_7	4.8
MoF_6	38.9
UF_6	56.5
Te_2F_{10}	58.9
PuF_6	62.0
IF_5	102.0
CrF_5	117.0
SbF_5	149.5
MoF_5	212.0
NbF_5	236.0
RuF_5	227.0
TeF_4	283.9
SbF_3	376.0
CrF_4	400.0
RuF_4	200.0

2.1 RADIONUCLIDES ASSESSMENT AND MASS ACCOUNTANCY

Figure 1 displays the nuclide chains that need to be assessed for source term development. Arrows from left to right within a row indicate nuclide transmutation with the progression continuing across subsequent rows. For example, the transmutation of Kr will result in Rb that, in turn, transmutes to Sr, and so on. Similarly, I will produce Xe, Cs, and Ba. Nuclides may be produced directly in fission, through radioactive decay, and through neutron capture. As indicated in Figure 1, the chemical behavior for each nuclide is quite different and includes noble gas, nonvolatile metals, and low-volatile fluorides. The chemical speciation depends on the chemical thermodynamic behavior of a given nuclide within the salt. [

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Figure 1. Low-volatile fluoride (brown), Low-volatile fluoride with redox (orange), Nonvolatile fluoride (green), Nonvolatile metal (blue), and Noble gas (red)
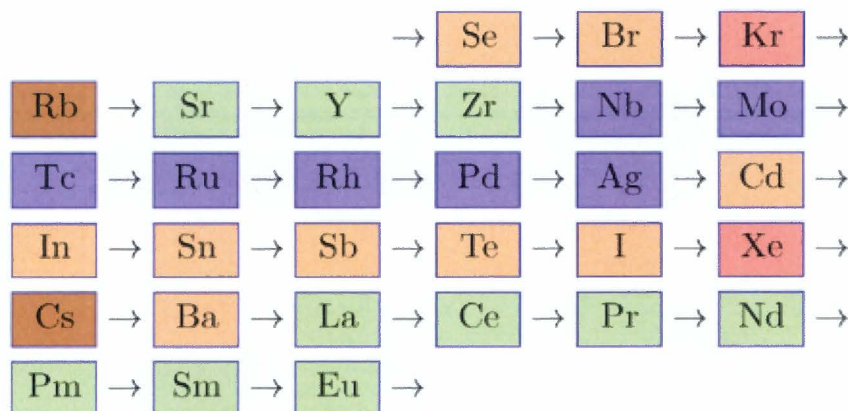
Radionuclides are color coded by noble gas, noble metal, and the affinity for a given nuclide to create fluoride compounds (non-volatile and low-volatile fluorides). As shown, [

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Table 2 displays the radionuclides requiring assessment for source term ranked by radiotoxicity. Dose factor is provided in units of millirems/millicurie. Also shown are the half-life for each nuclide (days), the estimated yield from ^{235}U fission, and an initial characterization of the nuclide within a fluoride salt system based on MSRE experience. The initial characterization shows nuclides that may be categorized as noble gas, nonvolatile fluoride, low-volatile fluoride, and non-volatile (noble) metal using the same color coding as Figure 1. The low-volatile fluoride (redox dep) indicates nuclides with a dependence on redox potential. The column, label "MSRE system state", shows an expected ordering from highest to lowest concentration for the given nuclide within the system state (salt, off-gas, and/or graphite/metal). Parentheses indicate a trace amount found in within the MSRE system. For example, [

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Table 2. Fission product radionuclides ranked in terms of radiotoxicity^{a,b}

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As shown in Table 2, [

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2.2   VAPOR THERMODYNAMICS WITHIN SALT

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3. GOVERNING EQUATIONS FOR MOLTEN SALT THERMAL FLUID

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3.1 FRICTIONAL LOSSES

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3.2 HEAT GENERATION AND REMOVAL

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3.3   SALT THERMOPHYSICAL PROPERTIES AND FLUID CLOSURES

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4. GOVERNING EQUATIONS FOR MOLTEN SALT NEUTRONICS

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5. GOVERNING EQUATIONS FOR MOLTEN SALT SPECIES TRANSPORT

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5.2   GAS-PHASE SPECIES TRANSPORT

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6.   INTERPHASE MASS TRANSFER

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6.1 CORRELATION FOR MASS TRANSFER COEFFICIENT

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6.2 LIQUID DIFFUSIVITY

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6.3 GAS DIFFUSIVITY (IN LIQUID)

[—   ]   (41)

6.4 HENRY'S GAS LAW CONSTANT

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6.5 GAS VELOCITY

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6.6 GAS-LIQUID INTERFACIAL AREA

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7. GOVERNING EQUATIONS FOR PLENUM GAS TRANSPORT

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7.1 GAS DIFFUSIVITY (PLENUM)

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8. UNCERTAINTY QUANTIFICATION METHODOLOGY BASED ON GOVERNING EQUATIONS

8.1 NEUTRONICS ANALYSIS

The nuclide inventory for the actinides (U, Np, Pu, Cm, and Am) and fission products of Table 1 is determined by the solution of the steady-state eigenvalue equation for k_{eff} and the neutron flux ϕ of Eqs. 21 and 22, followed by solution of the coupled set of nuclide transmutation equations represented by Eq. 20 (Hightower 1975).

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8.2 NEUTRONICS AND THERMAL-HYDRAULICS UNCERTAINTY PROPAGATION

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8.3 CHEMICAL SPECIATION UNCERTAINTY PROPAGATION

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8.4 UNCERTAINTY PROPAGATION IN BULK LIQUID- AND VAPOR-PHASE SPECIES TRANSPORT

Equations 24 and 25 represent species transport in the bulk liquid phase and gas phase, respectively, for nuclides defined in Table 1. For the bulk liquid, [

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8.5   DAKOTA UQ ANALYSIS

Table 3 summarizes the physics, model parameter input, and calculated QoIs based on the governing equations. [

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The Dakota software from Sandia National Laboratories (Adams et al. 2021) provides a toolbox of advanced parametric analysis techniques enabling quantification of margins and uncertainty, risk analysis, model calibration, and design exploration with computational models. Dakota includes methods for optimization, UQ, parameter estimation, and sensitivity analysis, [

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Table 3. UQ analysis model input and output QoIs

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Random sampling methods are the "black-box" UQ work horse provided in Dakota (as well as the Sampler code used with Shift). Essentially, [

] Dakota has several sampling methods, including Monte Carlo and Latin hypercube, that are easy to understand.

9. CONCLUSIONS

The analyses and UQ methodology for determining off-gas nuclide composition based on the governing equations for coupled neutronics, thermal fluids, mass transfer, and chemical speciation has been established. A number of closure relations outlined in the document will be subject to change but will generally not impact the approach outlined .

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