U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Modeling Radionuclide Transport across the Salt-Gas

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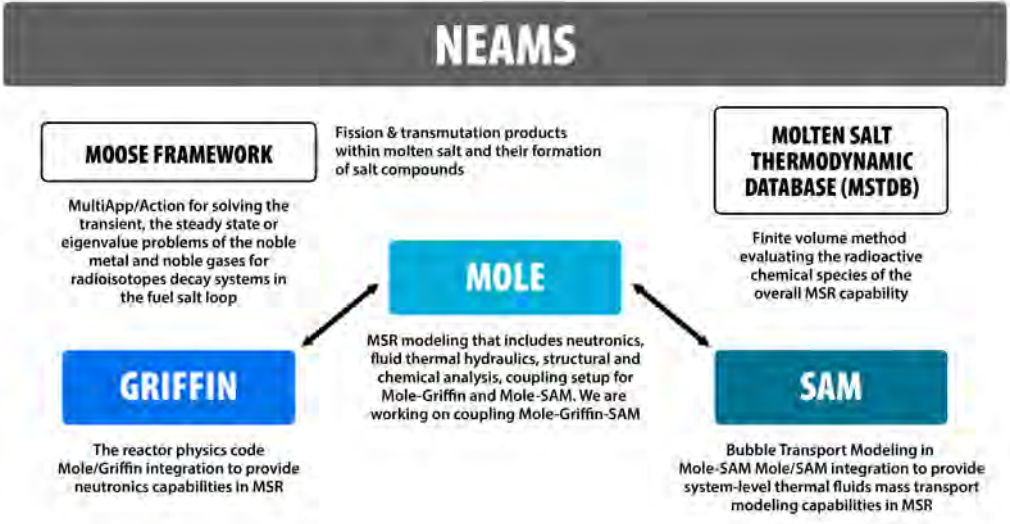
NEAMS
Nuclear Energy Advanced Modeling and Simulation

Nuclear Energy Advanced Modeling and Simulation (NEAMS), DOE-NE



Introduction to Mole Code

- The Mole code is part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program supported by US Department of Energy Office of Nuclear Energy
- The Mole code predicts the formation of fouling, erosion, and corrosion in molten salt reactor (MSR) fuel cycle loops and flow loops with **thermophysical or thermochemical properties and phase equilibrium or transitions**
- The Mole code performs macroscale or mesoscale diffusion or coupling with reactor codes to solve eigenvalue problems and perform transient analysis with improved fidelity for **MSR safety analysis**
- **Decay and the transmutation of nuclides** can be used to calculate the transition of the parent nucleus to a daughter nucleus of the static or dynamic radioisotopes

Introduction to Mole Code (cont.)

Mole Code

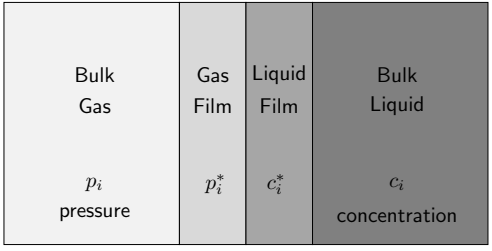
- The Mole code includes the corrosion and leaching of Cr with heat conduction
- The Mole code focuses on **noble metals and noble gases**

Salt Dynamics

- Solid alloy
  - Macroscopic description of diffusion in the solid alloy below the surfaces
- Surface interface in liquid solution
  - Electrochemical surface reaction
  - Solid to liquid solution for convective mass transfer
  - Liquid solution to solid for convective mass transfer
- Salt interface in liquid–gas transition
  - **Liquid solution to gas (vapor)** for convective mass transfer
  - **Gas to liquid** or depositions on wall for convective mass transfer
- Nuclear transmutation and radioactivity
  - Bateman equations with fission fragments and neutron sources

Mass Transfer across a Gas-Liquid Interface

There are two film theories and the overall mass transfer coefficients



$$c_i^* = p_i H$$

$$p_i^* = c_i / H$$

Figure 1: The gas-liquid interface of the film theory expressed in partial pressures and concentrations.

Liquid transport:

$$\frac{\partial c_l}{\partial t} + \nabla \cdot \mathbf{J}_l = K_L a \left(c_l^* - c_l\right)$$

$$\mathbf{J}_l = -D_l \nabla c_l - \mathbf{u} c_l$$

Gas transport:

$$\frac{\partial c_g}{\partial t} + \nabla \cdot \mathbf{J}_g = K_G a \left(p_g - p_g^*\right)$$

$$\mathbf{J}_g = -D_g \nabla c_g - \mathbf{v} c_g$$

K_G is the overall gas-phase mass transfer coefficient, K_L is the overall liquid-phase mass transfer coefficient, $\mathbf{u}$ is liquid velocity, and $\mathbf{v}$ is gas velocity, H is Henry's gas constant. a is gas-liquid interfacial area per unit volume, and D is diffusivity.

Mass Transfer across a Gas-Liquid Interface (cont.)

The overall mass transfer coefficients, K_G - gas phase and K_L - liquid phase are defined as:

$$\frac{1}{K_G} = \frac{1}{k_l H} + \frac{1}{k_g} \qquad \frac{1}{K_L} = \frac{1}{k_l} + \frac{H}{k_g}$$

where k_l is the liquid phase mass transfer coefficient and k_g is the gas phase mass transfer coefficient. H is Henry's Law constant and defined by $H = c_l/p_g$.
The entropy change for an equilibrium process can be explained by the Gibbs free energy.

$$\Delta G = \Delta \mathcal{H} - T\Delta S$$

where $\Delta\mathcal{H}$ is Enthalpy change, and ΔS is Entropy change, and T is temperature in K. When the temperature of a system changes, the Henry's constant changes and is related to the Van 't Hoff equation. The least squares regression can find the arbitrary number, α and β.

$$\Delta G = RT \ln\left(H/H^\circ\right) = 4\pi\alpha r^2 \gamma(T) + \frac{4}{3}\pi RT\beta r^3$$

where R is the ideal gas constant, r is Van der Waals radius, and γ is the surface tension.

$$H(r, T; \gamma(T)) = H^\circ \exp\left(\frac{\Delta G}{RT}\right)$$

Henry's gas constant

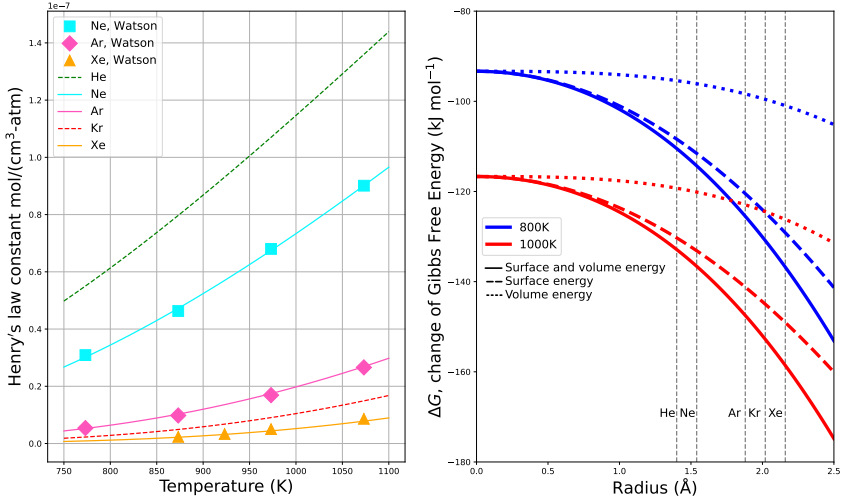


Figure 2: Henry's gas constant dependant on temperature and the change of Gibbs free energy ($LiF\text{-}BeF_2$)

Liquid Salt Thermophysical and Thermochemical Properties

- $LiF\text{-}BeF_2$ (64.0-36) %aw.
  $\gamma(T) = 235.5 - 0.09T(^\circ\mathrm{C})[\mathrm{erg/cm^2}]$
  $\rho_f(T) = 2.09 - 2.7 \times 10^{-4} T(^\circ\mathrm{C})[\mathrm{g/cm^3}]$
  $\mu_f(T) = 0.0594 \exp\left[4605/T(^\circ\mathrm{K})\right][\mathrm{mPa-s}]$
- LiF-NaF-KF (46.5-11.5-42.0)%aw.
  $\gamma(T) = 237.0 - 0.0788T(^\circ\mathrm{C})[\mathrm{erg/cm^2}]$
  $\rho_f(T) = 2.47 - 0.68 \times 10^{-3} T(^\circ\mathrm{C})[\mathrm{g/cm^3}]$
  $\mu_f(T) = 0.04 \exp\left[4170/T(^\circ\mathrm{K})\right][\mathrm{mPa-s}]$
- Liquid-Mass Diffusivity: Stokes-Einstein and the Wilke-Chang equations, and Hayduk-Minhas Correlation

$$D = \frac{k_B T(^\circ\mathrm{K})}{6\pi\mu_f r_0} \quad \text{or} \quad \frac{7.4 \times 10^{-8}(\phi M)^{0.5} T(^\circ\mathrm{K})}{\mu_f V_B^{0.6}}[\mathrm{m^2/sec}] \quad \text{or} \quad 13.3 \times 10^{-8} \frac{T^{1.47}\mu_f^{\epsilon}}{V_b^{0.71}}$$

$$\epsilon = 10.2/V_b - 0.791$$

where k_b is the Boltzmann constant, r_0 is the solute radius, M is Molecular weight solvent, V_B is Molar volume at boiling point, and ϕ is factor for solute.

Dittus-Boelter Equation: Turbulent Flow

The Dittus-Boelter equation in circular tubes is:

$$\mathrm{Sh} \ = 0.023 \mathrm{Re}^{0.8} \mathrm{Sc}^{n} = \frac{h_c L}{D},$$

where:
$n = 0.4$ for heating, $n = 0.3$ for cooling,
L is a cylinder diameter,
D is the diffusivity, and
Turbulent flow, $\mathrm{Re} >$ 10,000

$$\mathrm{Re} = \frac{\rho_f \left|\mathbf{u}\right| L}{\mu_f} = \frac{\rho_f Q L}{\mu_f A} \qquad \text{Reynolds number}$$

$$\mathrm{Sc} = \frac{\mu_f}{\rho_f D} \qquad \text{Schmidt numbers}$$

$$\mathrm{Sh} = \frac{h_c L}{D} \qquad \text{Sherwood number}$$

where: ρ_f is the density of the fluid,
μ_f is the dynamic viscosity of the fluid, and
$\mathbf{u}$ is the velocity of the fluid

- The mass transfer coefficient h_c is:

$$h_c = 0.023 \frac{D}{L} \mathrm{Re}^{0.8} \mathrm{Sc}^{n}.$$

Reynolds number, Diffusivity, and Schmidt number

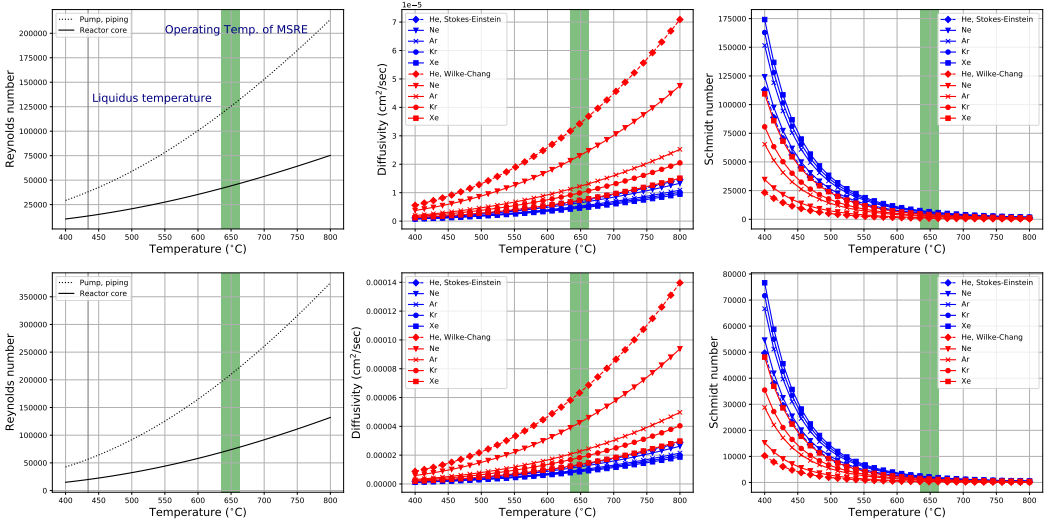


Figure 3: Top: $\mathrm{LiF\text{-}BeF_2}$, Bottom: $\mathrm{LiF\text{-}NaF\text{-}KF}$

Liquid Mass Transfer Coefficients

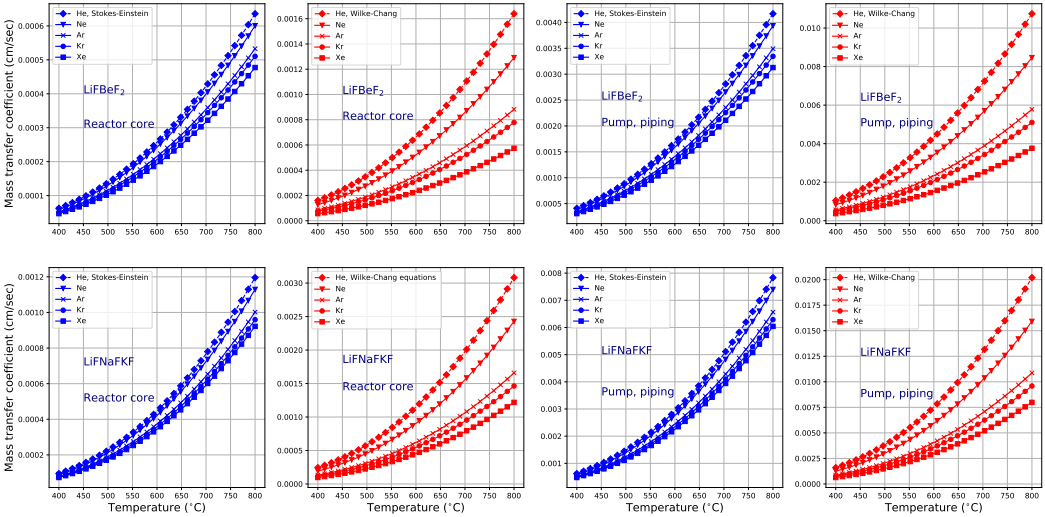


Figure 4: Top: $LiF\text{-}BeF_2$, Bottom: LiF-NaF-KF

Gas-Mass Diffusivity

- The diffusion coefficient for gases can be expressed by using Fuller's equation:

$$D_g = \frac{10^{-3} T^{1.75} \left(\frac{1}{M_A} + \frac{1}{M_B} \right)^{0.5}}{P \left(V_A^{1/3} + V_B^{1/3} \right)}$$

where P is the total system pressure [atm] M_i is molecular weight and V_i is sum of the diffusion volume, index i; A is solvent, B is solute, D_g is gas diffusivity, [cm^2/s], and T is temperature, ($^\circ\mathrm{K}$).

- Gas diffusivity makes it difficult to determine which gas solute is the mixture from salt, air or vacuum involving flow through the pipes.
- Gas velocity is complex to define the system because Reynolds number is required to gas velocity.
- The overall gas mass transfer coefficient can be derived from Herny's gas constant. Thus Herny's gas constant can be considered as a key parameter for gas-liquid interface.
- When $(H/k_g)^{-1} \ll k_l^{-1}$, the overall mass transfer coefficients can be generalized to

$$K_L \approx k_l$$
$$K_G = K_L H$$

Gas Mass Transfer Coefficients

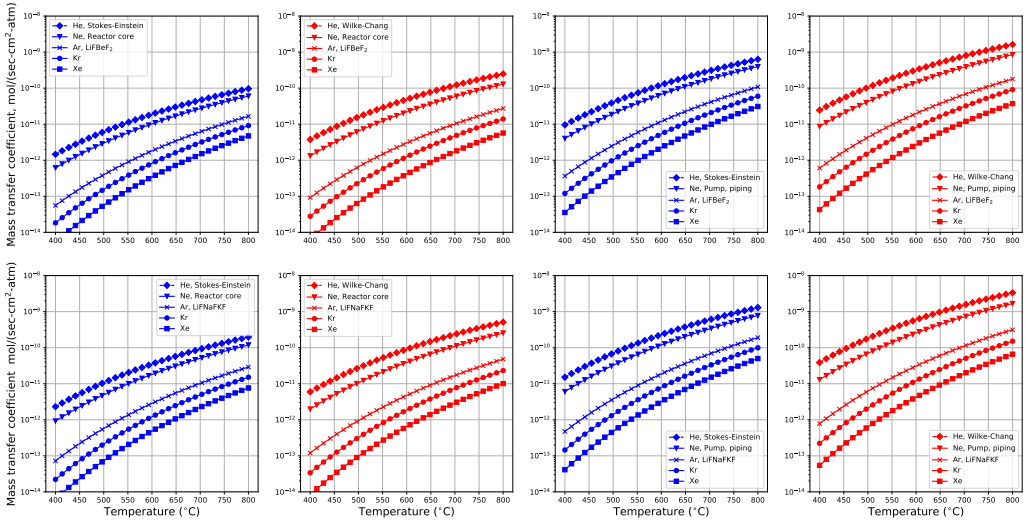


Figure 5: Top: $\mathrm{LiF\text{-}BeF_2}$, Bottom: $\mathrm{LiF\text{-}NaF\text{-}KF}$

Liquid-Gas Mass Transport Using Mole

We have considered the mechanism of mass transfer between phases without convection. The overall mass-transfer coefficients were defined by $c_l = p_g H = c_g HRT$ and $K_G = K_L H$ where $p_g = c_g RT$ and $R = 82.05746[\mathrm{cm}^3 \cdot \mathrm{atm}/(\mathrm{K} \cdot \mathrm{mole})]$

$$\text{Liquid transport:}$$

$$\frac{\partial c_l}{\partial t} = K_L a \left(c_g HRT - c_l\right)$$

$$\text{Gas transport:}$$

$$\frac{\partial c_g}{\partial t} = K_G a \left(c_g RT - c_l/H\right)$$

where c is the concentration of species in liquid, p is partial pressure of species in gas phase, and H is Henry's gas constant. a is gas-liquid interfacial area per unit volume.

Gas-Liquid Interfacial Area

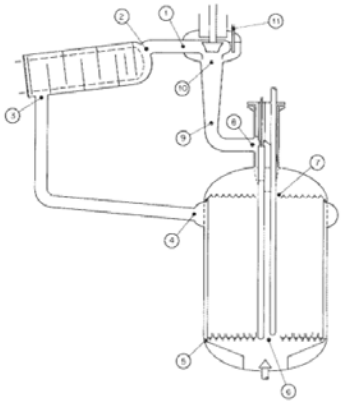


Figure 6: Schematic diagram of MSRE Assembly

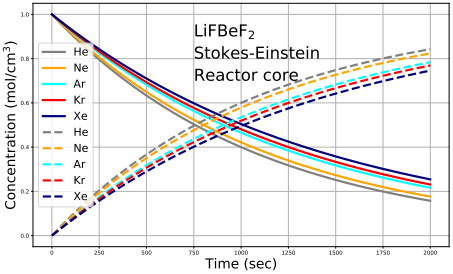
Table 1: MSRE parameters in fuel circulating loop

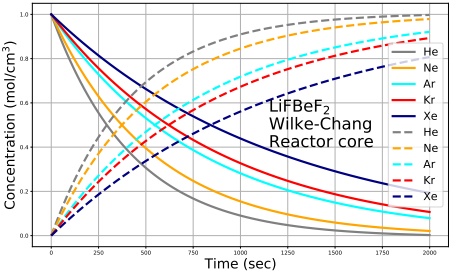
	Volume (ft^3)	Residence Time (sec)	Length (cm)	Fluid velocity † (cm/sec)	(ft/sec)
Pump $^{10\to1}$	1.10	0.41	56.6	149.4	4.90
Fuel Loop Piping $^{1\to2}$	0.76	0.28	39.1	149.4	4.90
Heat Exchanger $^{2\to3}$	6.12	2.29	589.7	279.6	9.17
Fuel Loop Piping, $^{3\to4}$	2.18	0.81	112.2	149.4	4.90
Outer annulus, $^{4\to5}$	9.72	3.63	157.9	47.1	1.55
Lower plenums, $^{5\to6}$	12.24	4.58	18.1	29.9	0.14
Reactor vessel core, $^{6\to7}$	23.52	8.79	149.8	18.5	0.61
Upper plenums, $^{7\to8}$	11.39	4.26	17.4	29.9	0.15
Fuel Loop Piping, $^{8\to9}$	1.37	0.51	70.5	149.4	4.90
Fuel Loop Piping, $^{9\to10}$	0.73	0.27	37.6	149.4	4.90
Total	69.13	25.86	1,249.0		

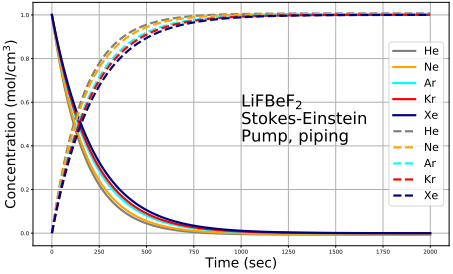
† Volumetric flow rate, 1200 gpm is $Q = A_{\mathrm{eff}}\mathbf{u}$, where $\mathrm{A}_{\mathrm{eff}}$ is the effective cross section and $\mathbf{u}$ is fluid velocity.

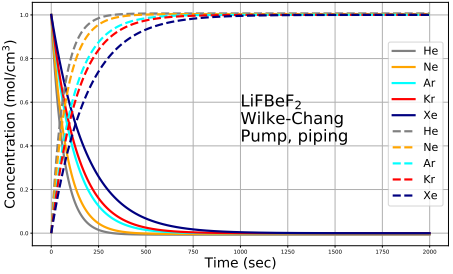
	Surface Area (ft^2)	Interfacial Area (cm^{-1})†
Bubbles of 233U Runs	5581.00	2.5972
Bubbles of 235U Runs	345.00	0.16055

† Interfacial area is $a = \dfrac{A}{V}$, where the volume of a fuel salt in the fuel loop is 70.5 ft^3. The columns of gray color are the data from Kedl's paper.

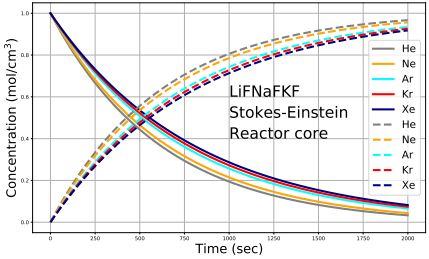

LiFBeF2
Stokes-Einstein
Reactor core
Concentration (mol/cm3)
Time (sec)
He
Ne
Ar
Kr
Xe

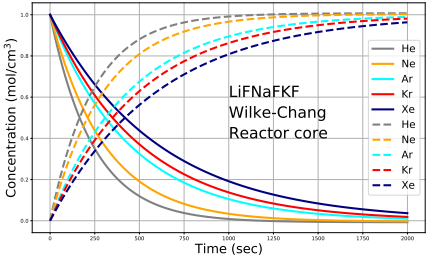

LiFBeF2
Wilke-Chang
Reactor core
Concentration (mol/cm3)
Time (sec)
He
Ne
Ar
Kr
Xe

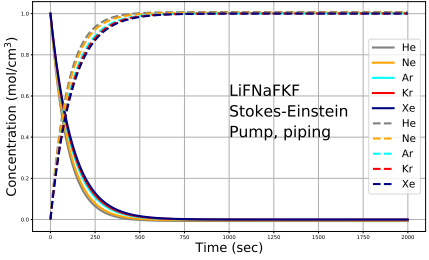

LiFBeF2
Stokes-Einstein
Pump, piping
Concentration (mol/cm3)
Time (sec)
He
Ne
Ar
Kr
Xe

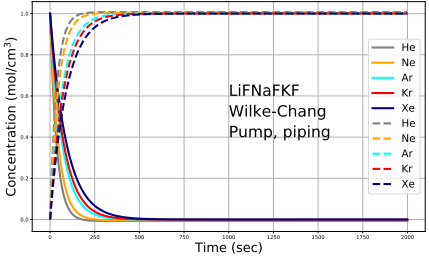

LiFBeF2
Wilke-Chang
Pump, piping
Concentration (mol/cm3)
Time (sec)
He
Ne
Ar
Kr
Xe

Initial constions: liquid 1 mole, gas 0 mole



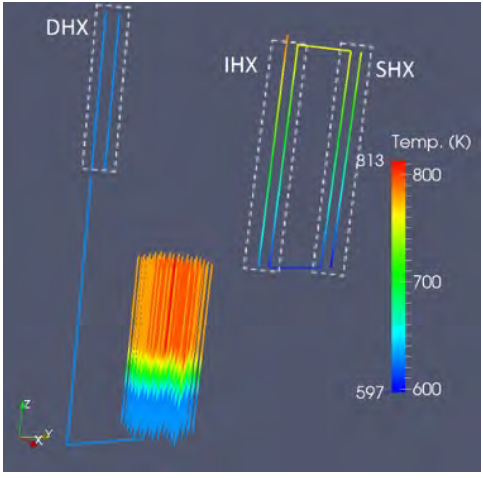






SAM

- SAM is a system-scale thermal-hydraulic modeling tool written using the MOOSE framework
- Solution of mass, momentum, and energy equations on 1D mesh
- Targeted applications include single-phase flow and heat transfer in advanced reactor systems
- Prediction of velocity, temperature in fluids and solids, and pressure during steady-state and transients



SAM gas transport model

- Presence of non-condensable gas phase in some MSR designs plays an important role in MSR behavior
  - Facilitates removal of fission gasses which can impact operational efficiency and safety (e.g., xenon and tritium)
  - Noble metals can affect heat exchanger efficiency and material corrosion and can also interact with entrained gases
- Model added to SAM to capture gas transport and local behavior
- Results in prediction of local gas void, bubble velocity, and bubble diameter/interfacial area

Gas conservation equation added to SAM:

$$\frac{\partial \alpha \rho_g}{\partial t} + \nabla \cdot (\alpha \rho_g u_g) = S \tag{1}$$

Gas velocity calculated using drift-flux model, where gas distribution is dependent on salt viscosity and drift velocity is dependent on surface tension:

$$u_g = C_0 u_l + u_{gj} \tag{2}$$

Bubble diameter depends on surface surface tension and impacts interfacial area calculation:

$$We = \frac{\rho_l u_r^2 d}{\sigma} \tag{3}$$

Model validation

- Kress performed photographic measurements of helium bubbles in flowing salt surrogate fluid in vertical and horizontal orientations
- Bubble injector device allows for known bubble size distribution with bubble size similar to MSRE conditions
- Measurements of volume fraction, bubble diameter, and interfacial area made

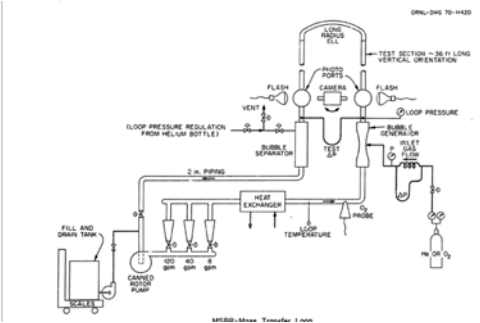


Figure 7: Kress test section

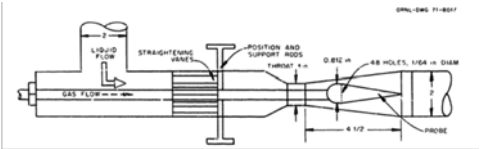


Figure 8: Bubble injector device

Model validation

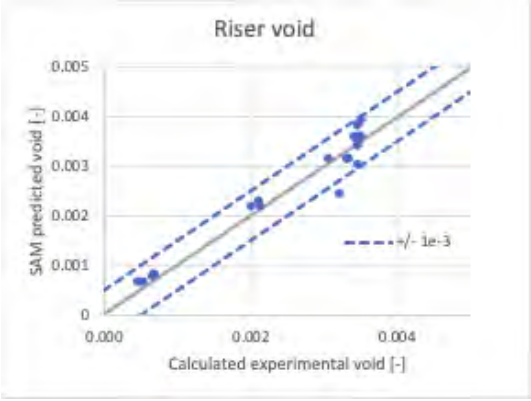


Figure 9: Void prediction in riser

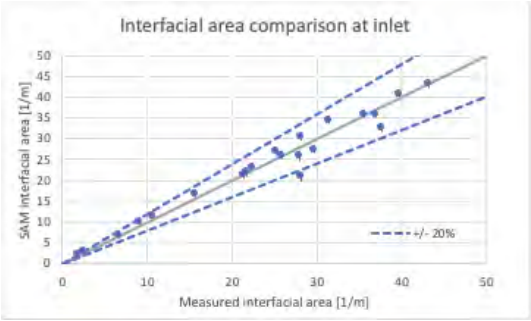
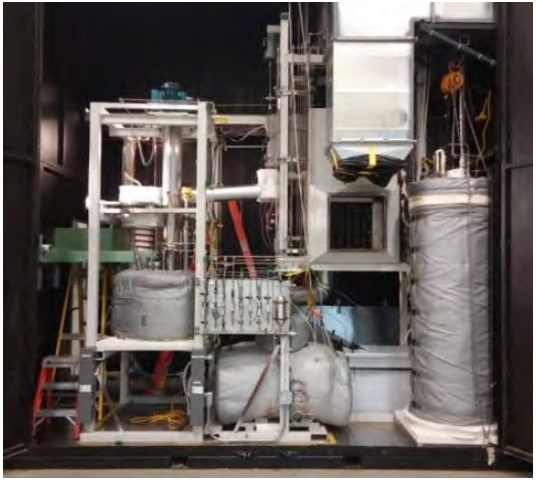


Figure 10: Interfacial area prediction in riser

LSTL Overview

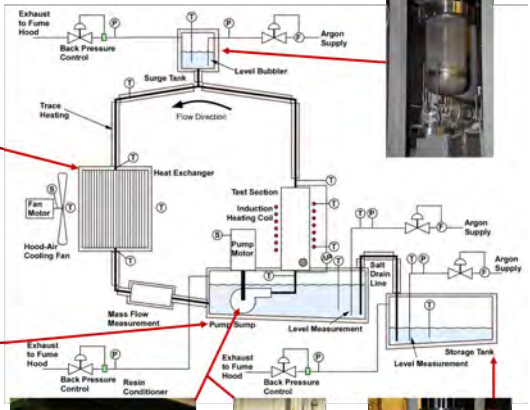
LSTL is located at ORNL and was created as a versatile facility for the development and demonstration of MSR technology

Salt	NaF-Kf-LiF (FLiNaK)
Operating Temp.	700 C
Flow rate	$<=$ 4.5 kg/s
Operating pressure	Near atmospheric
Primary materials	Inconel 600 + SiC
Loop volume	80 liters
Power	200 kW induction
	about 20 kW trace
Primary piping ID	2.67 cm
Initial operation	Summer 2016



LSTL Overview

- Salt pump located in sump tank
- Test section with 200 kW induction heating
- Trace heating of all components and piping
- Surge tank for pressure control
- Air-cooled heat exchanger



SAM LSTL Model

- Previous TRACE model of the facility used in extracting system geometry for SAM
- New heater section modeled as collection of heated flow paths with total power of 200 kW

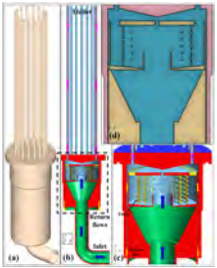


Figure 11: Heater section of facility

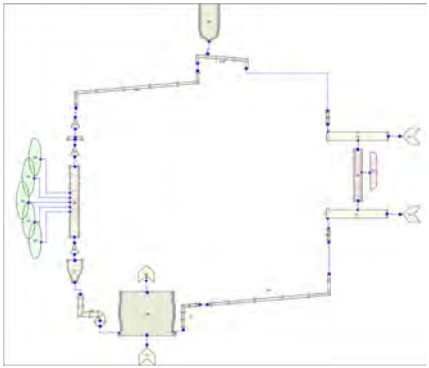


Figure 12: Existing TRACE model of facility[a]

[a] Avigni, et al., "Liquid Salt Test Loop modeling using TRACE", Annals of Nuclear Energy, 2017.

SAM LSTL Model

- Experimental data not yet available for benchmarking of the model
- 100 second transient run to achieve steady state conditions
- Pump head adjusted to achieve 4.5 kg/s flow rate in facility
- Heat exchanger fin multiplier adjusted to remove total input power at steady state conditions
- Total salt mass of system is 164.6 kg compared to 150 kg in TRACE model

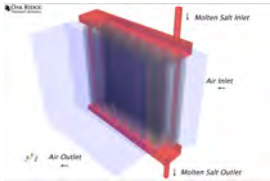
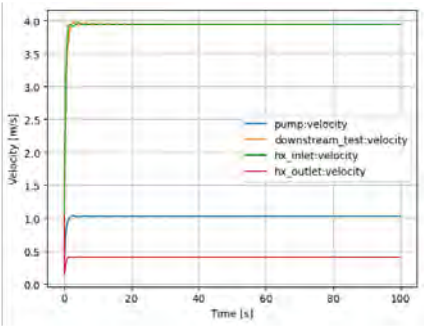
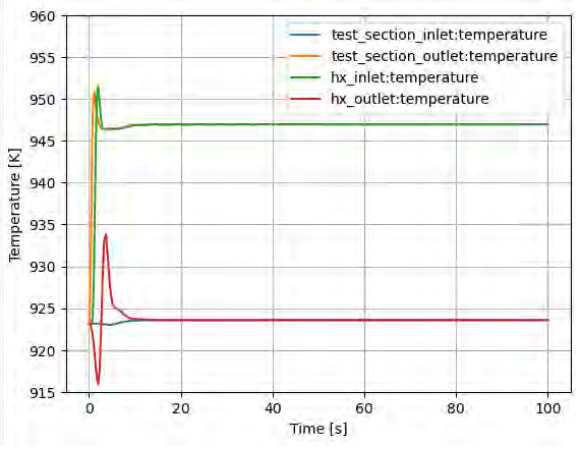
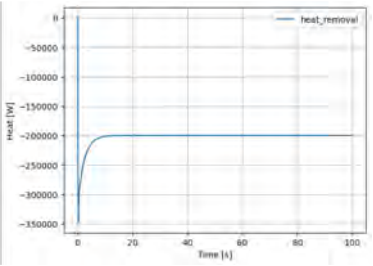


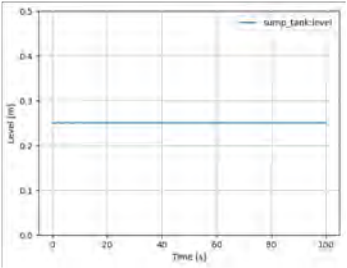
Figure 13: LSTL heat exchanger



SAM LSTL Model







SUMMARY I

The liquid-gas transport of the off-gas system can determine high-quality parameters. The liquid-gas transport accounts for salt compounds and noble gas behavior. This includes the physical and chemical processes that affect the distribution and range of movement between the various parts of the MSR system.

- We have developed for the coupling liquid-gas mass transport using Mole and SAM.
- The off-gas system of Mole is based on the fundamental correlations of liquid or gas.
- The experimental data of Henry's gas constant are not sufficient to verify the theory.
- Gas velocity and gas diffusivity are complex to define theory, Thus the experimental results can provide to establish a theory's validity.
- For liquid diffusion, Wilke-Chang's distribution of noble gases is wider than that of Stokes-Einstein.
- For liquid-gas mass transport, all liquid-gas transitions tend toward a state of chemical equilibrium because Henry's gas constant is very low.

SUMMARY II

- From the concentration versus time graph, the concentration rate is dependent on the Reynolds number or diffusivity.
- Gas transport model implemented into SAM for determination of interfacial area and bubble velocity needed for mass transport calculation
- Preliminary validation of gas transport model completed, but more data is needed
- SAM model setup for LSTL facility, which will be benchmarked as experimental data becomes available