Mass Transfer Coefficient of Noble Metals and Gases for Molten Salt Reactor Using Correlations

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ENERGY

Introduction to Mole

- 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
- Mole predicts the formation of fouling, erosion, and corrosion in the molten salt reactor (MSR) fuel cycle and flow loops with **thermophysical/thermochemical properties and phase equilibrium/transitions**
- Mole performs macroscale/mesoscale diffusion or coupling with reactor codes to solve eigenvalue problems/transient analysis with an improved fidelity capability for **MSR safety analysis**
- **Decay and the transmutation of nuclides** can calculate the transition of the parent nucleus to a daughter nucleus of the static/dynamic radioisotopes

Introduction to Mole Code (cont.)

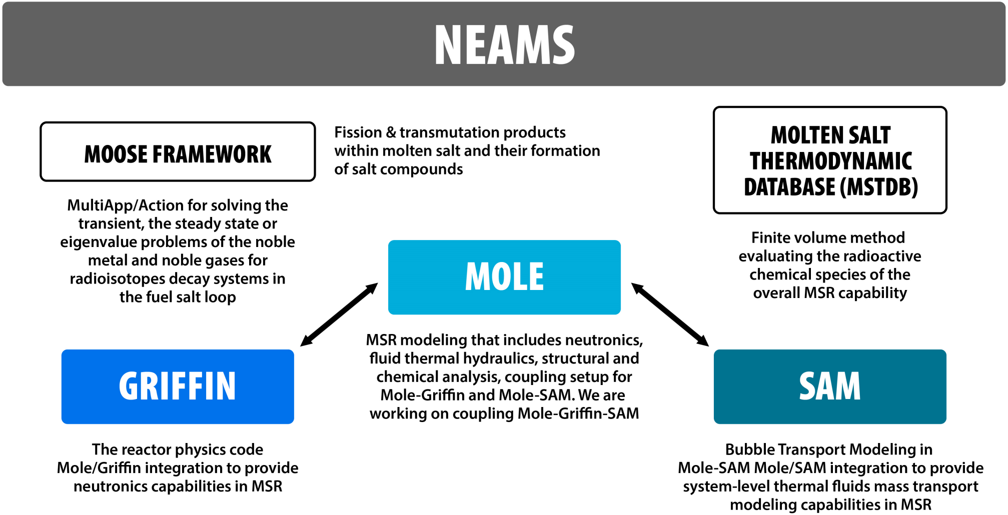
Mole:

- Includes the corrosion and leaching of chromium with heat conduction
- 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

NEAMS, DOE-NE



NEAMS

MOOSE FRAMEWORK

MultiApp/Action for solving the transient, the steady state or eigenvalue problems of the noble metal and noble gases for radioisotopes decay systems in the fuel salt loop

Fission & transmutation products within molten salt and their formation of salt compounds

MOLTEN SALT THERMODYNAMIC DATABASE (MSTDB)

Finite volume method evaluating the radioactive chemical species of the overall MSR capability

MOLE

MSR modeling that includes neutronics, fluid thermal hydraulics, structural and chemical analysis, coupling setup for Mole-Griffin and Mole-SAM. We are working on coupling Mole-Griffin-SAM

GRIFFIN

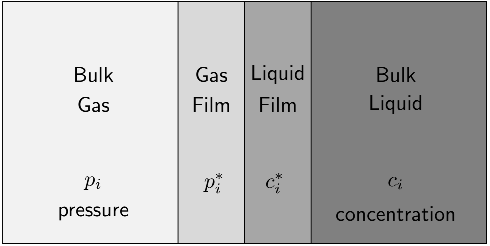
The reactor physics code Mole/Griffin integration to provide neutronics capabilities in MSR

SAM

Bubble Transport Modeling in Mole-SAM Mole/SAM integration to provide system-level thermal fluids mass transport modeling capabilities in MSR

Mass Transfer across a Gas–Liquid Interface

There are two film theories and the overall mass transfer coefficients



Bulk Gas

p_i

pressure

Gas Film

p_i^*

Liquid Film

c_i^*

Bulk Liquid

c_i

concentration

$$c_i^* = p_i H$$

$$p_i^* = c_i / H$$

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, $\mathbf{v}$ is gas velocity, and 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, ΔS is entropy change, and T is temperature in K. When the temperature of a system changes, Henry's constant changes and is related to the Van 't Hoff equation. The least squares regression can find the arbitrary numbers, α and β.

$$\Delta G = RT \ln H = \alpha 4 \pi r^2 \gamma + \beta$$

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

Henry's gas constant

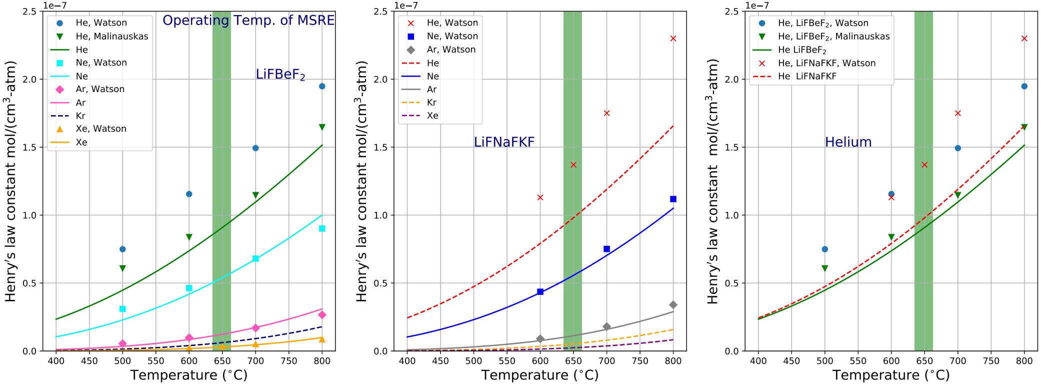


Figure 2: Henry's gas constant dependent on temperature (left $\mathrm{LiF\text{-}BeF_2}$; right $\mathrm{LiF\text{-}NaF\text{-}KF}$)

Liquid Salt Thermophysical and Thermochemical Properties

- $\mathrm{LiF\text{-}BeF_2}$ (64.0-36) at. wt%.
  $\gamma(T) = 235.5 - 0.09T(^\circ\mathrm{C})[\mathrm{erg/cm^2}]$
  $\rho_f(T) = 2.09 - 2.7 \times 10^{-4} T(^\circ\mathrm{C})[\mathrm{g/cm^3}]$
  $\mu_f(T) = 0.0594 \exp\left[4605/T(^\circ\mathrm{K})\right][\mathrm{mPa{-}s}]$

- $\mathrm{LiF\text{-}NaF\text{-}KF}$ (46.5-11.5-42.0) at. wt%.
  $\gamma(T) = 237.0 - 0.0788T(^\circ\mathrm{C})[\mathrm{erg/cm^2}]$
  $\rho_f(T) = 2.47 - 0.68 \times 10^{-3} T(^\circ\mathrm{C})[\mathrm{g/cm^3}]$
  $\mu_f(T) = 0.04 \exp\left[4170/T(^\circ\mathrm{K})\right][\mathrm{mPa{-}s}]$

- 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 the molecular weight solvent, V_B is the molar volume at boiling point, and ϕ is the 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, $\mathrm{Re} > 10{,}000$

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

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

$$\mathrm{Sh} = \frac{h_c L}{D}$$ 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

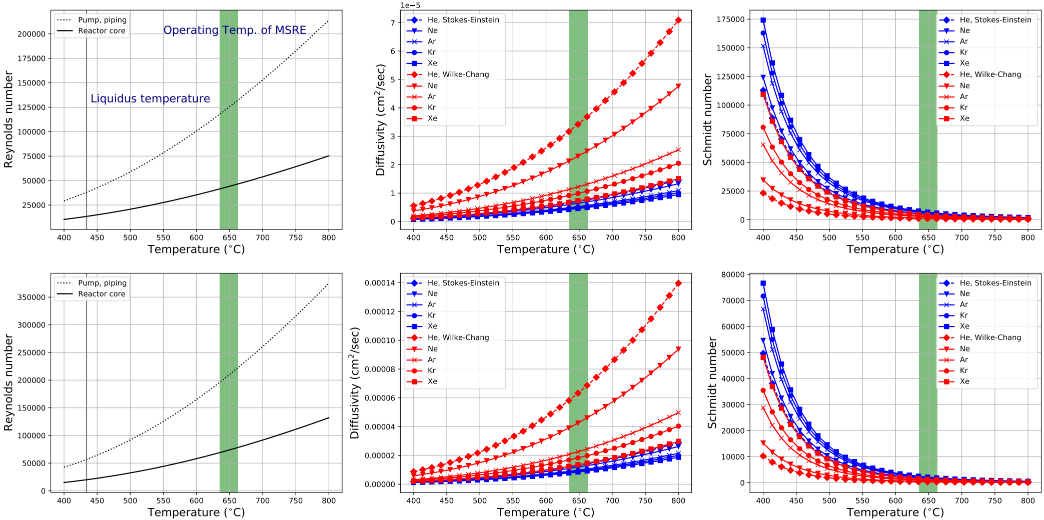


Figure 3: Top: $\mathrm{LiF\text{-}BeF_2}$, Bottom: $\mathrm{LiF\text{-}NaF\text{-}KF}$

Liquid Mass Transfer Coefficients

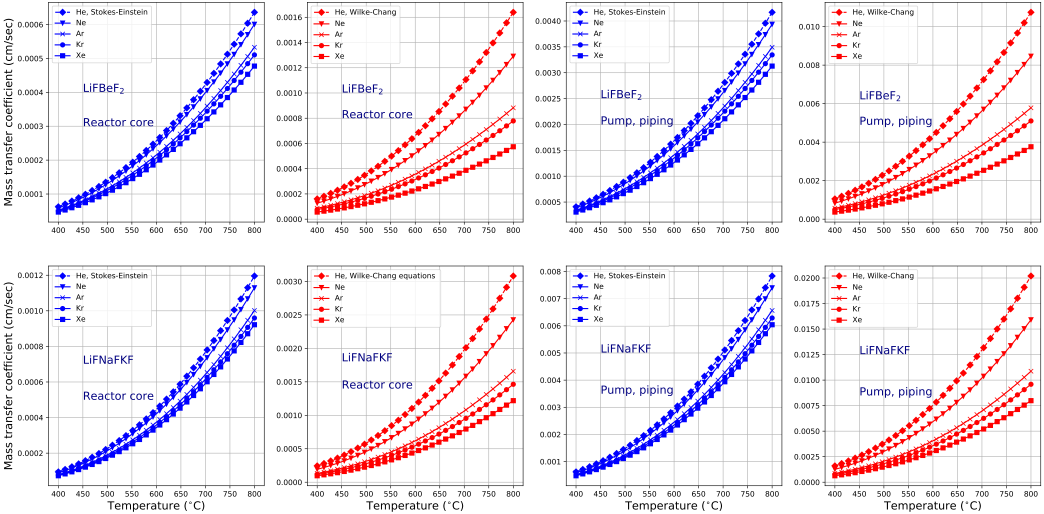


Figure 4: Top: $\mathrm{LiF\text{-}BeF_2}$, Bottom: $\mathrm{LiF\text{-}NaF\text{-}KF}$

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, $[\mathrm{cm}^2/\mathrm{s}]$, 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.
- The definition of gas velocity in the system is complex because Reynolds number is required for 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

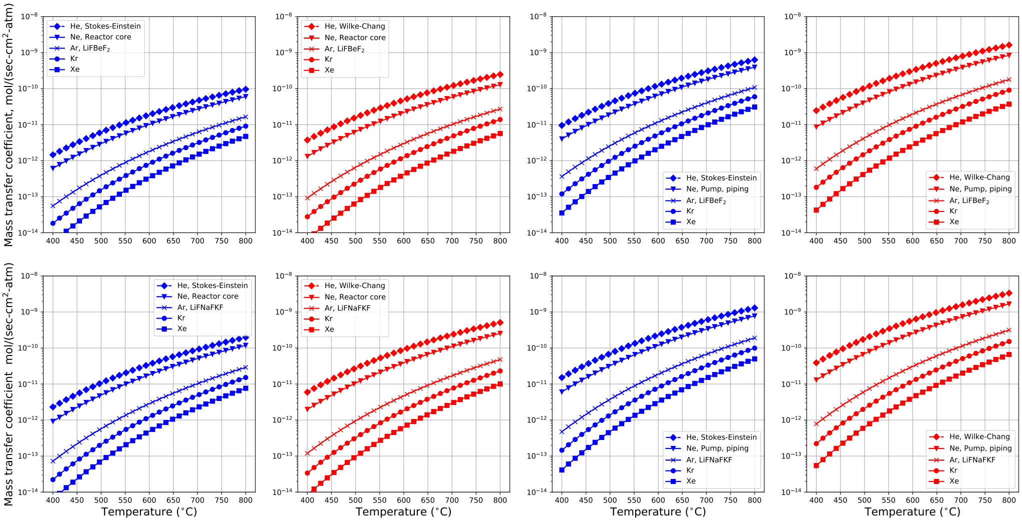


Figure 5: Top: LiF-BeF$_2$, Bottom: LiF-NaF-KF

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

Gas–Liquid Interfacial Area

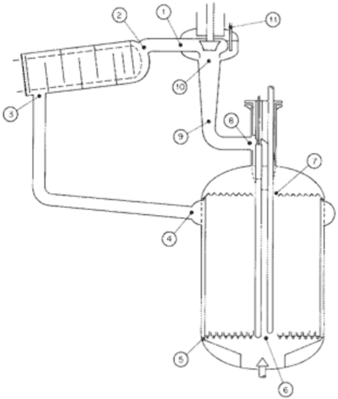


Figure 6: Schematic diagram of MSRE Assembly

Table 1: MSRE parameters in fuel circulating loop

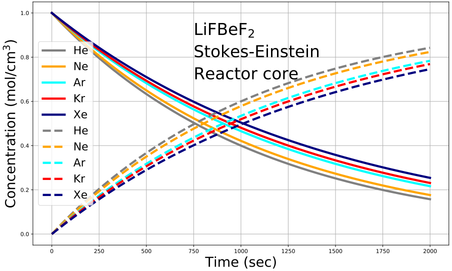
	Volume (ft^3)	Residence Time (sec)	Length (cm)	Fluid velocity † $(\mathrm{cm/sec})$	$(\mathrm{ft/sec})$
Pump $^{10\to1}$	1.10	0.41	56.6	149.4	4.90
Fuel Loop Piping $^{1\to2}$	0.76	0.28	39.1	149.4	4.90
Heat Exchanger $^{2\to3}$	6.12	2.29	589.7	279.6	9.17
Fuel Loop Piping, $^{3\to4}$	2.18	0.81	112.2	149.4	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 A_{eff} is the effective cross section and $\mathbf{u}$ is fluid velocity.

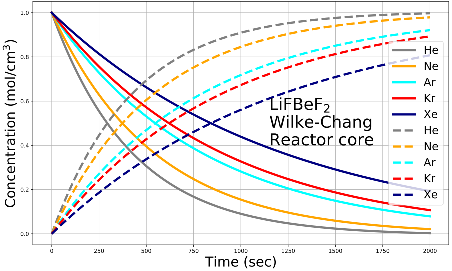
	Surface Area (ft^2)	Interfacial Area (cm^{-1})†
Bubbles of 233U Runs	5581.00	2.5972
Bubbles of 235U Runs	345.00	0.16055

† 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.

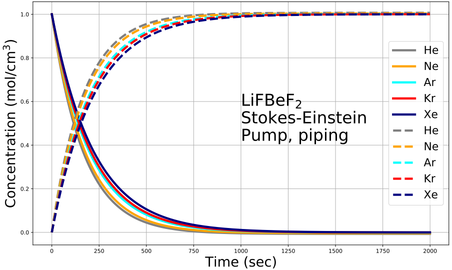
$LiBeF_2$



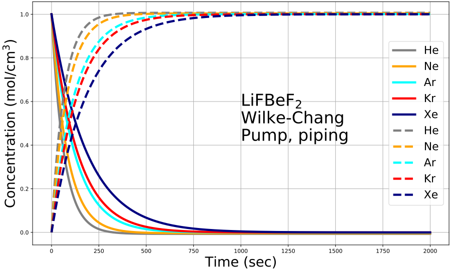
LiFBeF$_2$
Stokes-Einstein
Reactor core
Concentration (mol/cm^3)
Time (sec)
0.0
0.2
0.4
0.6
0.8
1.0
0
250
500
750
1000
1250
1500
1750
2000
He
Ne
Ar
Kr
Xe
He
Ne
Ar
Kr
Xe



LiFBeF$_2$
Wilke-Chang
Reactor core
Concentration (mol/cm^3)
Time (sec)
0.0
0.2
0.4
0.6
0.8
1.0
0
250
500
750
1000
1250
1500
1750
2000
He
Ne
Ar
Kr
Xe
He
Ne
Ar
Kr
Xe



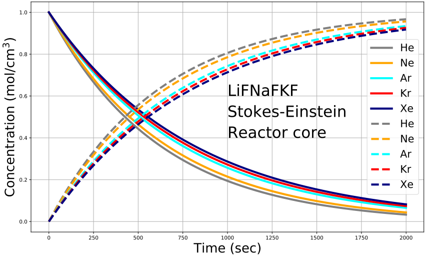
LiFBeF$_2$
Stokes-Einstein
Pump, piping
Concentration (mol/cm^3)
Time (sec)
0.0
0.2
0.4
0.6
0.8
1.0
0
250
500
750
1000
1250
1500
1750
2000
He
Ne
Ar
Kr
Xe
He
Ne
Ar
Kr
Xe

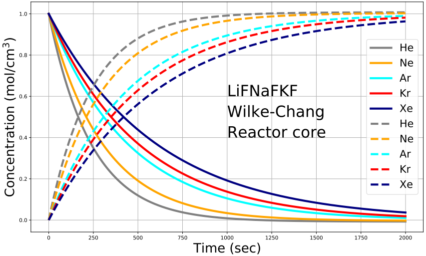


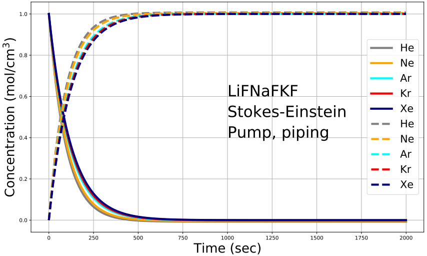
LiFBeF$_2$
Wilke-Chang
Pump, piping
Concentration (mol/cm^3)
Time (sec)
0.0
0.2
0.4
0.6
0.8
1.0
0
250
500
750
1000
1250
1500
1750
2000
He
Ne
Ar
Kr
Xe
He
Ne
Ar
Kr
Xe

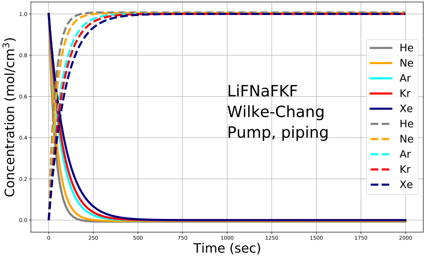
LiFNaFKF

Initial constions: liquid 1 mole, gas 0 mole









SUMMARY

The liquid–gas transport of the off-gas system can determine high-quality parameters. It 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.
- 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 theories are difficult to define; thus, the experimental results can help to validate them.
- For liquid diffusion, Wilke–Changs 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 Henrys gas constant is very low.
- From the concentration versus time graph, the concentration rate is dependent on the Reynolds number or diffusivity.