# Investigation of Fission Product Influence on the $UCl_3$-LiCl-KCl Eutectic

**Jacob Tellez, Jenifer Shafer, David Wu, and Neil Henson**

*jtellez@mines.edu*

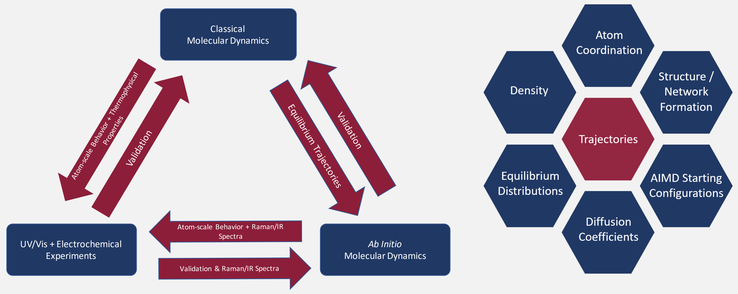
## Objective

To assess the molecular structures and chemistry that govern ***optical indicators*** of molten salt reactors (MSRs) by establishing molten salt spectroscopic monitoring capabilities, developing and validating molecular dynamic (CMD) simulations, and leveraging them for Raman spectroscopic interpretation to develop online monitoring capabilities.

## Overview

**Motivation**

- Monitoring of electronic and vibrational signatures paired with informed models outline nanoscale salt behavior in real time/near-real time.
- Benefits include nondestructive sampling, worker dose reduction, and proliferation risk minimization.




**MD Simulations**

- Classical polarizable potentials designed toward transferability between systems with similar foundational characteristics enable evaluation of equilibrium properties of mixed $MCl_x$ salts. Such potentials complement AIMD simulations, spectral sampling techniques, and electrochemical experiments to classify unique signatures corresponding to atom-scale behavior.

$$V(r_{ij}) = B_{ij}e^{-\alpha_{ij}r_{ij}} - f_{ij}^{6}(r_{ij})\frac{C_{6,ij}}{r_{ij}^{6}} - f_{ij}^{8}(r_{ij})\frac{C_{8,ij}}{r_{ij}^{8}}$$

Predict structural, thermodynamic, and kinetic properties of the eutectic salt → CMD

Predict Raman spectra of the eutectic salt → AIMD

Verify force field implementation for extension to other Ln systems → CMD AIMD

## Force Field Validation

**Validation Parameters and Systems**

- Coordination number pair distributions, activation energies, diffusion coefficients, and crystal properties
- Targeting **$UCl_3$-$LaCl_3$-LiCl-KCl** and relevant subsystems.

*Table 1: Ion self-diffusion coefficients in 5 a/o $UCl_3$-LiCl-KCl at 1023 K.*

| $D_i$ [$10^{-5}$ $cm^2/s$] | $Li^+$ | $K^+$ | $U^{3+}$ | $Cl^-$ |
|---|---|---|---|---|
| This work | 9.37 +/- 1.98 | 7.21 +/- 0.25 | 1.69 +/- 0.00 | 6.10 +/- 0.20 |
| Dai et al. | 9.26 | 7.89 | 1.63 | 7.39 |

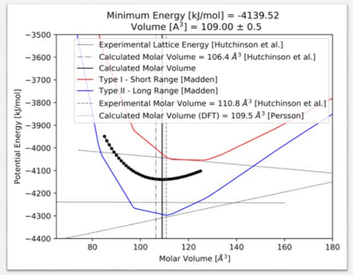



*Figure 1: Molar volume curves for pure $LaCl_3$ (left) and $UCl_3$ (right) crystals, calculated with the PIM, compared with other experimental and simulated data.*

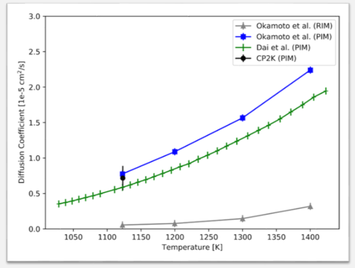



*Figure 2: Comparison of the $U^{3+}$ self-diffusion coefficient to other simulated values in pure $UCl_3$.*

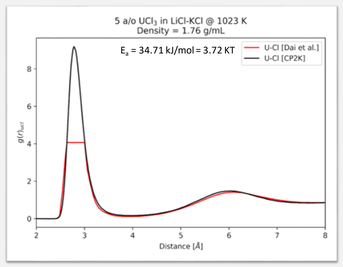



*Figure 3: Comparison of $U^{3+}$-$Cl^-$ PDF in the 5 a/o $UCl_3$ system to another classical potential.*

## Properties of Eutectic Systems

**Network Formation by $U^{3+}$**

- Cation networks, bridged by multiple shared chlorides, form in the molten salts and generate interest surrounding
  - The signatures associated with these networks
  - The impact of fission product species on network stability
- Network (de)stabilization can drastically impact the mobility of ions which influences bulk properties and may raise proliferation concerns.

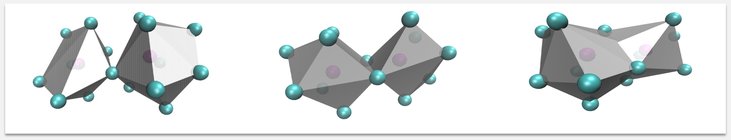


*Figure 4: Corner (left), edge (middle), and face (right) sharing uranium chloride clusters in pure $UCl_3$ at 1023 K. The clusters shown constitute fraction of an extensive network, which becomes smaller with the dilution of $UCl_3$ in LiCl-KCl.*

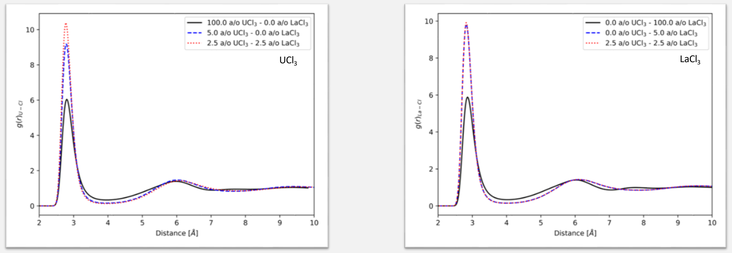



*Figure 5: Comparison of $M^{3+}$-$Cl^-$ pair distribution functions for 2.5, 5, and 100 a/o $MCl_3$ ($M$=$La^{3+}$,$U^{3+}$) at 1023 K.*

**References:**

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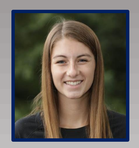
## Project Team

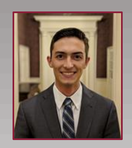

Jenifer Shafer


Nicole Hege


Sam Bryan


Amanda Lines


David Wu


Jacob Tellez


Neil Henson


Aurora Clark


Will Smith

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