# Evaluation of Chemical Speciation in Molten Salt Reactors

National Nuclear Security Administration

## Objectives

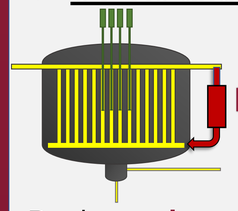
To assess the molecular structures and chemistry that govern ***optical indicators*** of molten salt reactors (MSRs) by establishing molten salt spectroscopic monitoring capabilities, developing and validating classical molecular dynamic (CMD) simulations, and leveraging them for Raman spectroscopic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

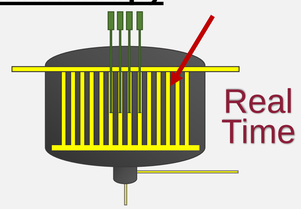
## Experimental

### Online Monitoring

- Uses **electronic** and **vibrational** signatures to characterize and monitor elements in the core
- Key benefits are nondestructive data collection, radiation worker dose reduction, and proliferation risk minimization

### UV-Vis and Raman Spectroscopy



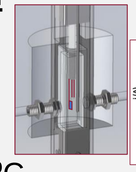



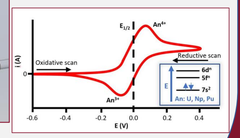

Produces **elemental specific** information for actinides and other fission products such as U, Pu

Probes the **chemical environment**, gives overall geometry and speciation information

### Experimental Design

- Equipped for UV-Vis and electrochemical experiments
- Furnace reaches temperatures of 750ºC



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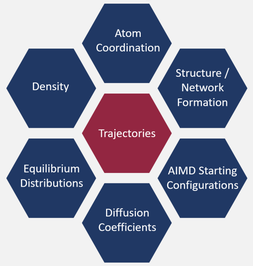
## Classical Molecular Dynamics

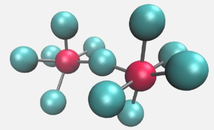
### Modeling Objectives

| Predict structural, thermodynamic, and kinetic properties of the eutectic salt | Predict Raman spectra of the eutectic salt | Verify force field implementation for extension to other Ln systems |
|---|---|---|
| CMD | AIMD | CMD AIMD |

### Classical MD

- Provides trajectories for analysis and AIMD



Snapshot of two $U^{3+}$ cations forming $[UCl_6]^{3-}$ complexes with a single shared chloride. Complexes can share one or two chlorides and form oligomerized chains.

### Verification of CMD Potential

- Against $MCl^{3+}$ coordination number, $MCl^{3+}$ diffusion coefficients, and pair distributions
- **$UCl_3$** system verified
- **$CeCl_3$** system being verified
- **$UCl_3$-LiCl-KCl** system preparing for verification

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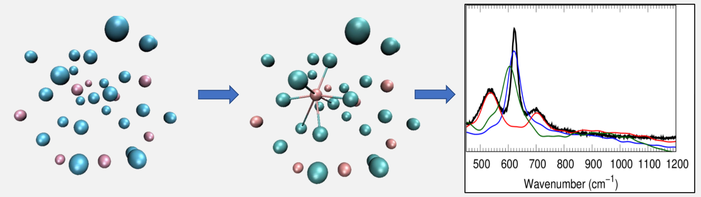
## Ab-initio Molecular Dynamics

### Subensemble Analysis

- Identify unique molecular species and environments using network analysis tools (i.e. graph theory)
- Linear fit of different spectroscopic signatures to yield the total observed in experiment




### *Ab-Initio* MD

- Initial starting coordinates are obtained from the CMD simulation
- Trajectories will be used to predict IR/Raman spectra and provide validation of CMD methods

### Ongoing Verification

- Current pseudopotentials for modeling the uranium atom were optimized for $UO_2$
- Collaboration with Vanda Glezakou at PNNL to improve existing methods

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## Project Team


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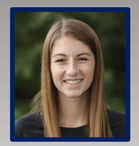
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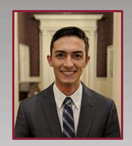
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