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

Molten Salt Reactor PROGRAM

Ab initio molecular dynamics study of $NaCl–UCl_3–PuCl_3$ mixtures

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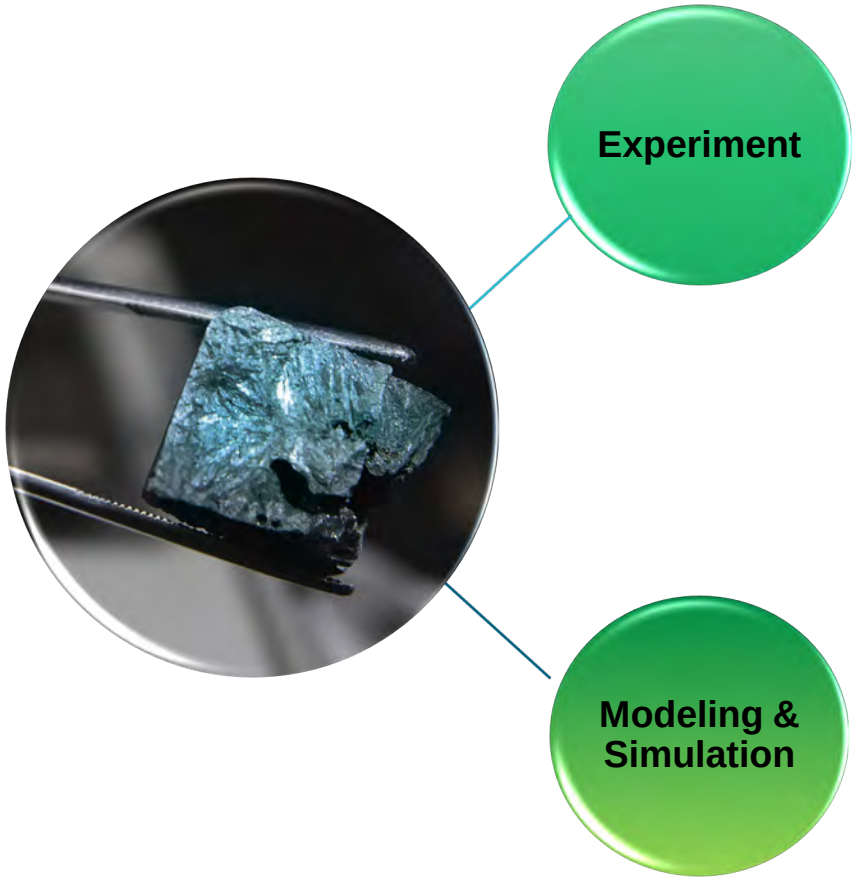
Pacific Northwest National Laboratory

Annual MSR Campaign Review Meeting 2-4 May 2023

Molten salt data base development



Toni Karlsson (INL)

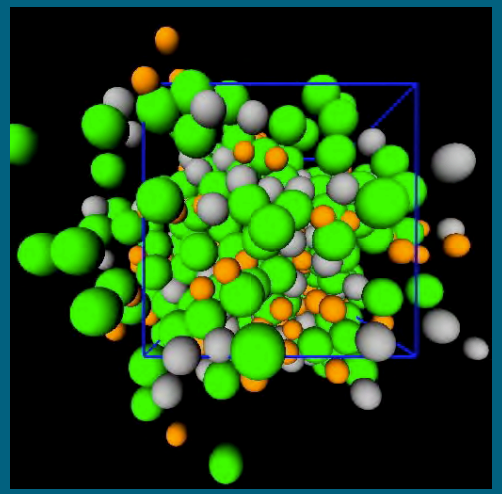
Marisa Monreal (LANL)

Manh Nguyen (PNNL)

Vanda Glezakou (ORNL)

OBJECTIVE

We will generate missing thermophysical, structural, and transport data for NaCl-UCl_3-$PuCl_3$ mixtures at different temperatures and compositions using atomistic modelling and data science.



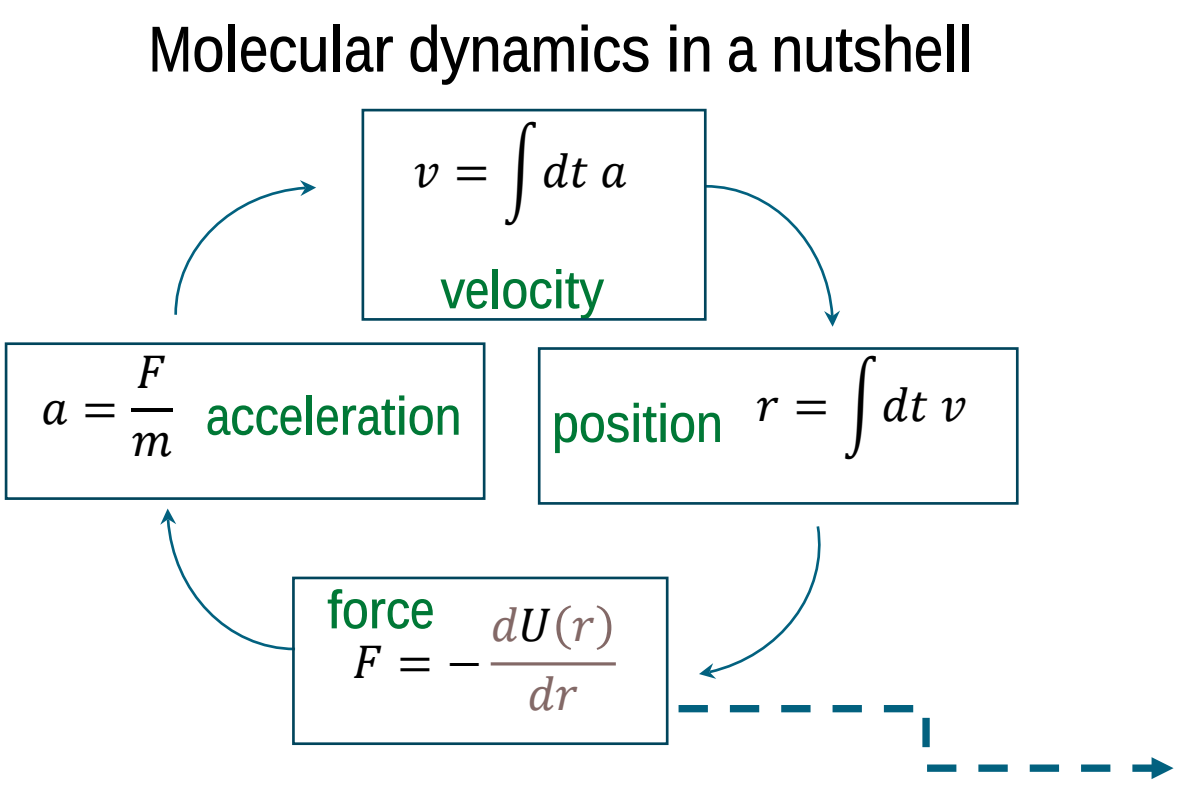
❑Scope

- "For FY23, we propose to investigate the ternary system of NaCl-UCl_3-$PuCl_3$ using AIMD in conjunction with data science approaches. Such a system is highly relevant to molten salt nuclear reactors. This research will be complementary to experimental studies by INL and LANL and theoretical studies by ORNL .The proposed work will consider 2 systems of 1-10 mol% $PuCl_3$ at 3 different temperatures."

❑Milestone

- "Level 3 milestone, Complete Summary report on ternary system of NaCl-UCl_3-$PuCl_3$ using AIMD due September, 30, 2023."

Molecular dynamics is our main approach



- Density; Heat capacity
- Mass transport; Charge transport
- Heat transport; Viscosity
- Enthalpy; Entropy
- Structure

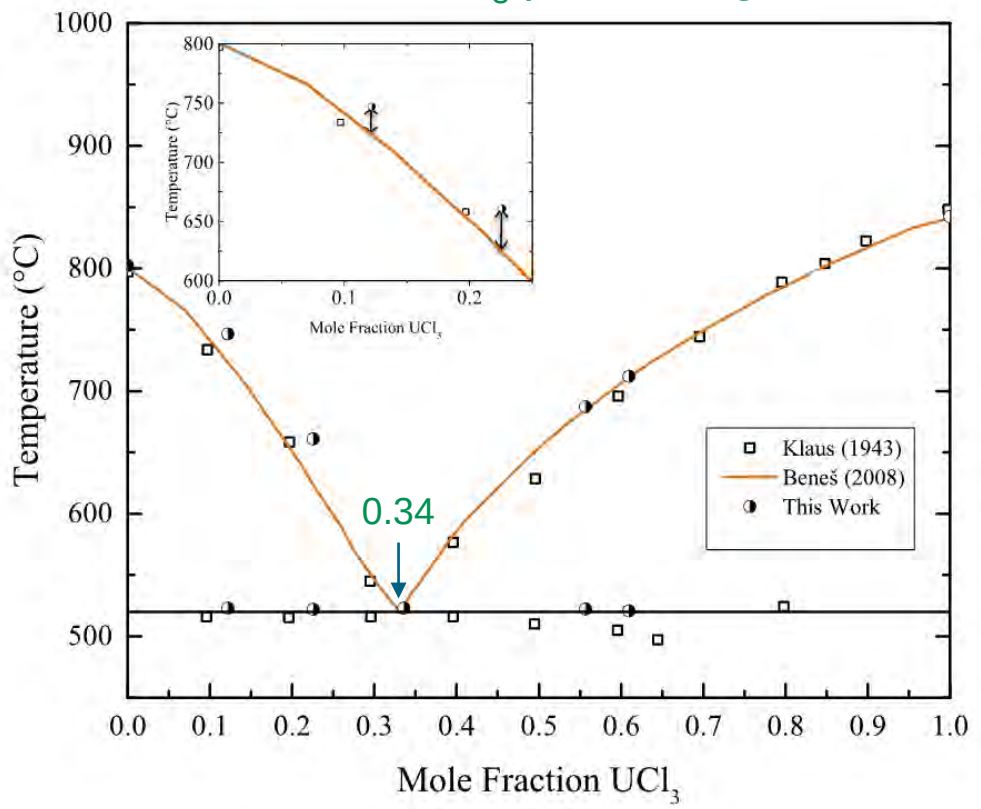
AIMD (in use): potential energy surface U(r) calculated with density functional theory (DFT)

MLD-MD: U(r) calculated with a machine learning interatomic potentials trained on AIMD potentials

The atomic systems

- $NaCl$-UCl_3-$PuCl_3$ is considered as a fuel salt.
- Use eutectic UCl_3-NaCl (34mol%-66mol%) as the “host” molten salt
- **System 1:** 5 mol% $PuCl_3$ added to eutectic UCl_3-NaCl
- **System 2**: 10 mol% $PuCl_3$ added to eutectic UCl_3-NaCl
- Unit cells: ~110 atoms for AIMD, and ~880 atoms for machine learning interatomic potentials MD

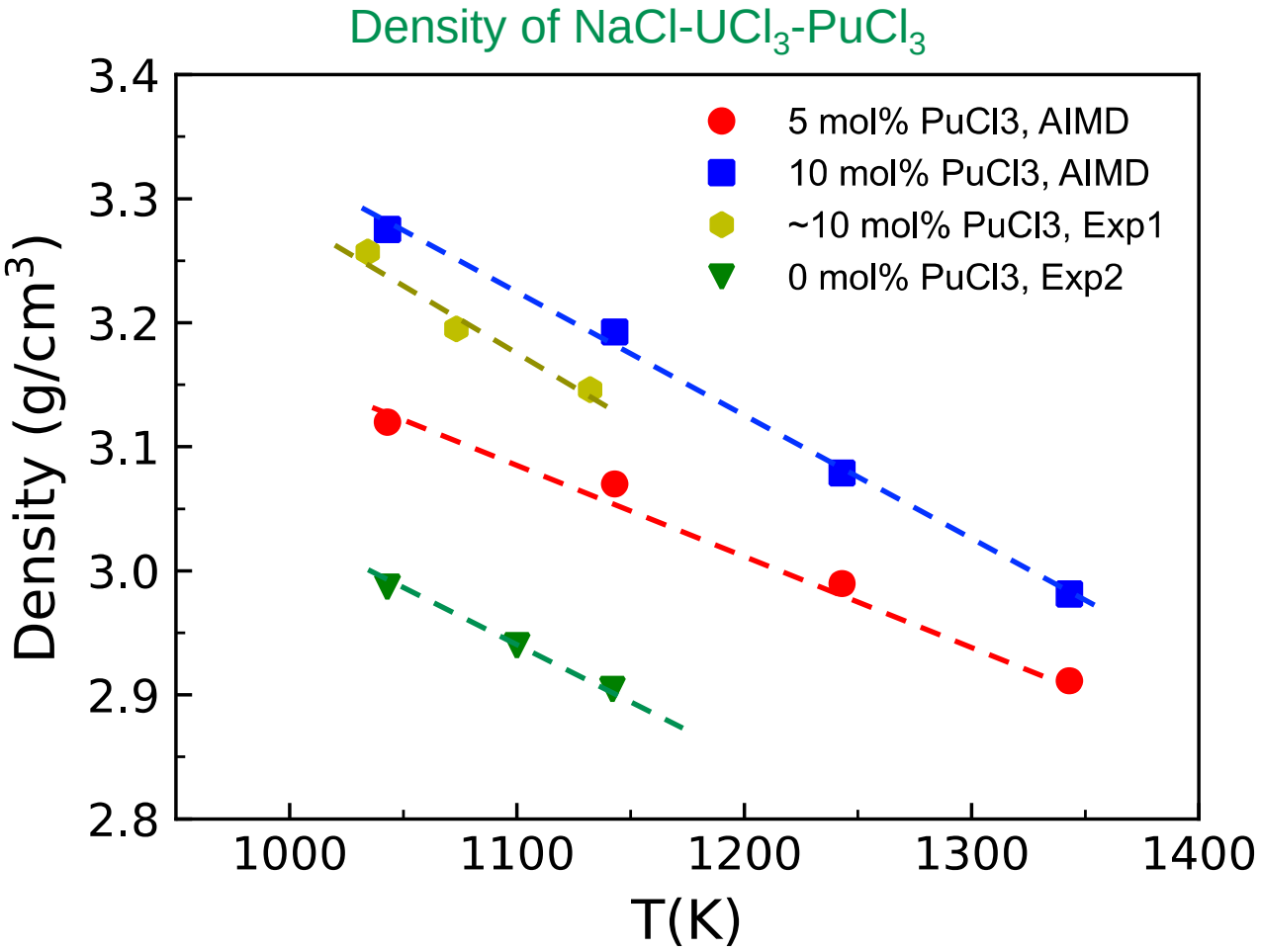
$NaCl$-UCl_3 phase diagram



Sooby et al. Journal of Nuclear Materials 466, 280 (2015)

AIMD predicts MS densities

- NPT ensemble.
- Linear dependence of the density on the temperature.
- Increasing $PuCl_3$ leading to higher density
- No direct comparison with experiment, but the trend looks similar



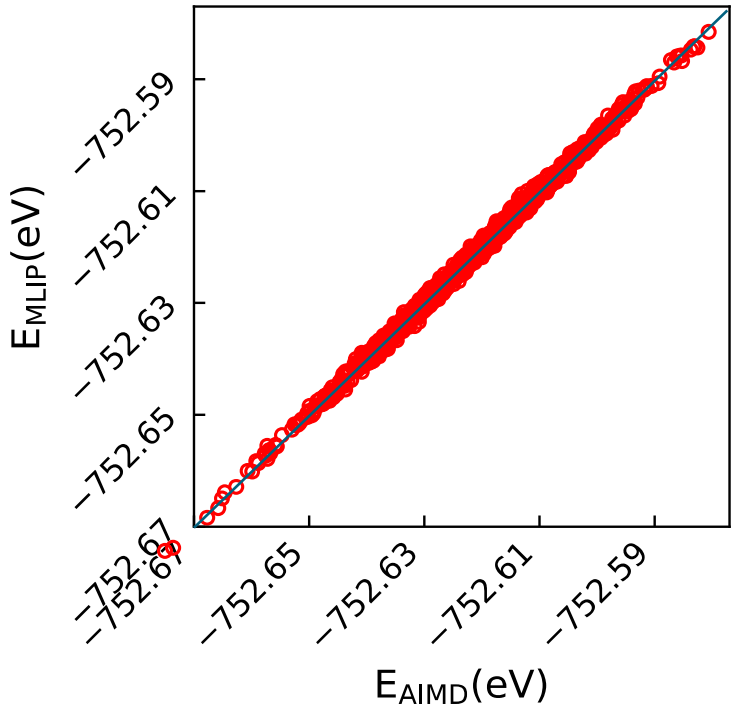
Exp1:Toni Y. Karlsson (INL)

Exp2: Desyatnik et al. Soviet Atomic Energy 39, 649–651 (1975)

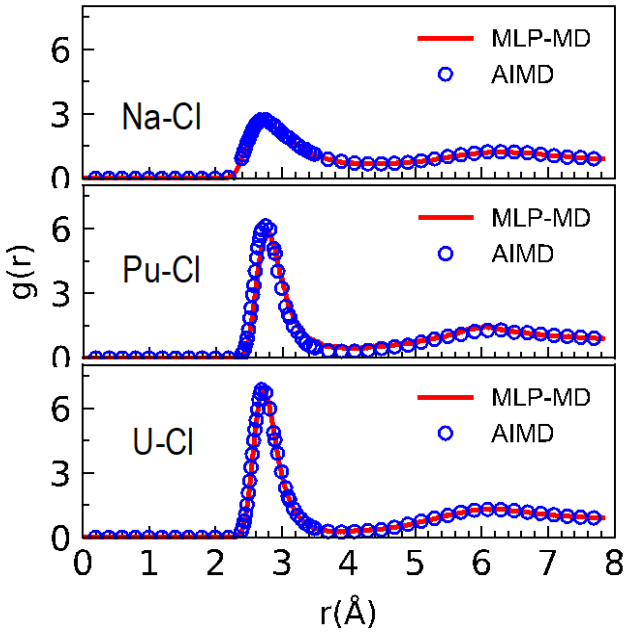
Machine learning interatomic potentials help accelerate transport studies

- Transport properties (e.g., diffusion) require intensive sampling
- AIMD is time-consuming
- Machine learning interatomic potentials, ***trained on AIMD potential energy surfaces***, offer a more feasible approach

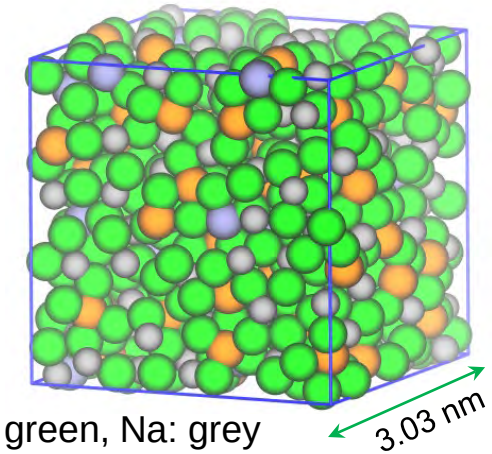
Potential energy



Radial distribution functions



Machine learning interatomic potential molecular dynamics predicts diffusion coefficients



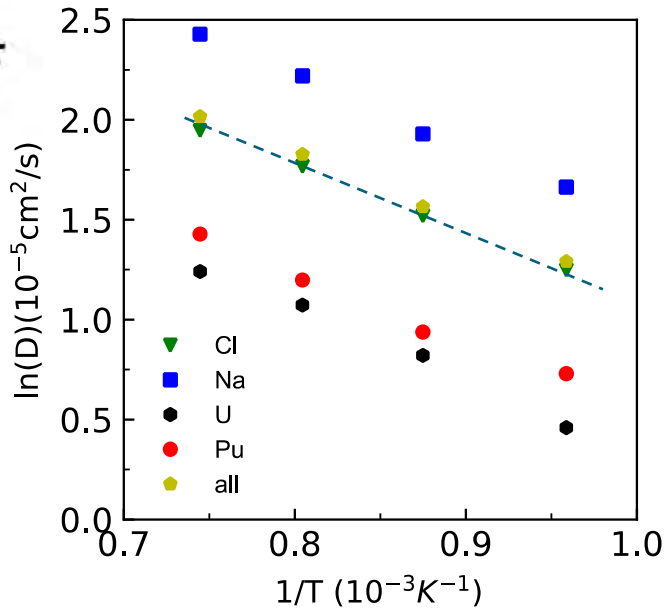
Cl: green, Na: grey
U: orange, Pu: purple

Diffusion coefficients

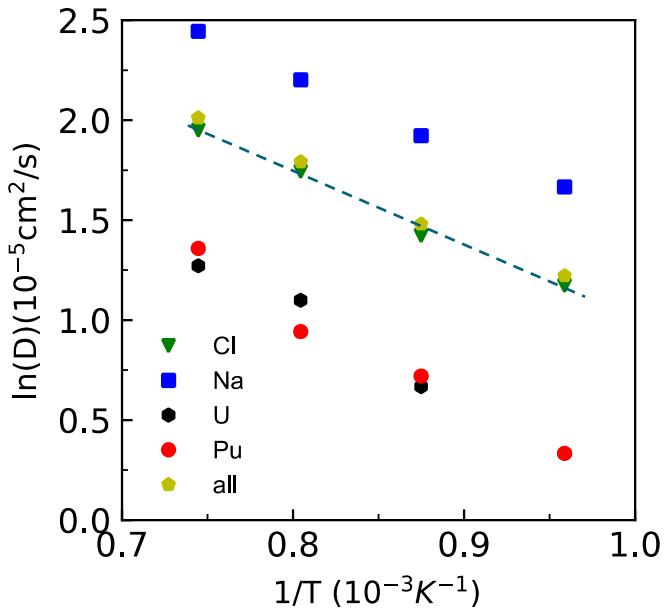
$$D = \frac{1}{3}\int_0^\infty \langle v(0)v(\tau)\rangle \, d\tau$$

$$D_{all} = \sum \alpha_i D_i$$

5 mol% $PuCl_3$



10 mol% $PuCl_3$

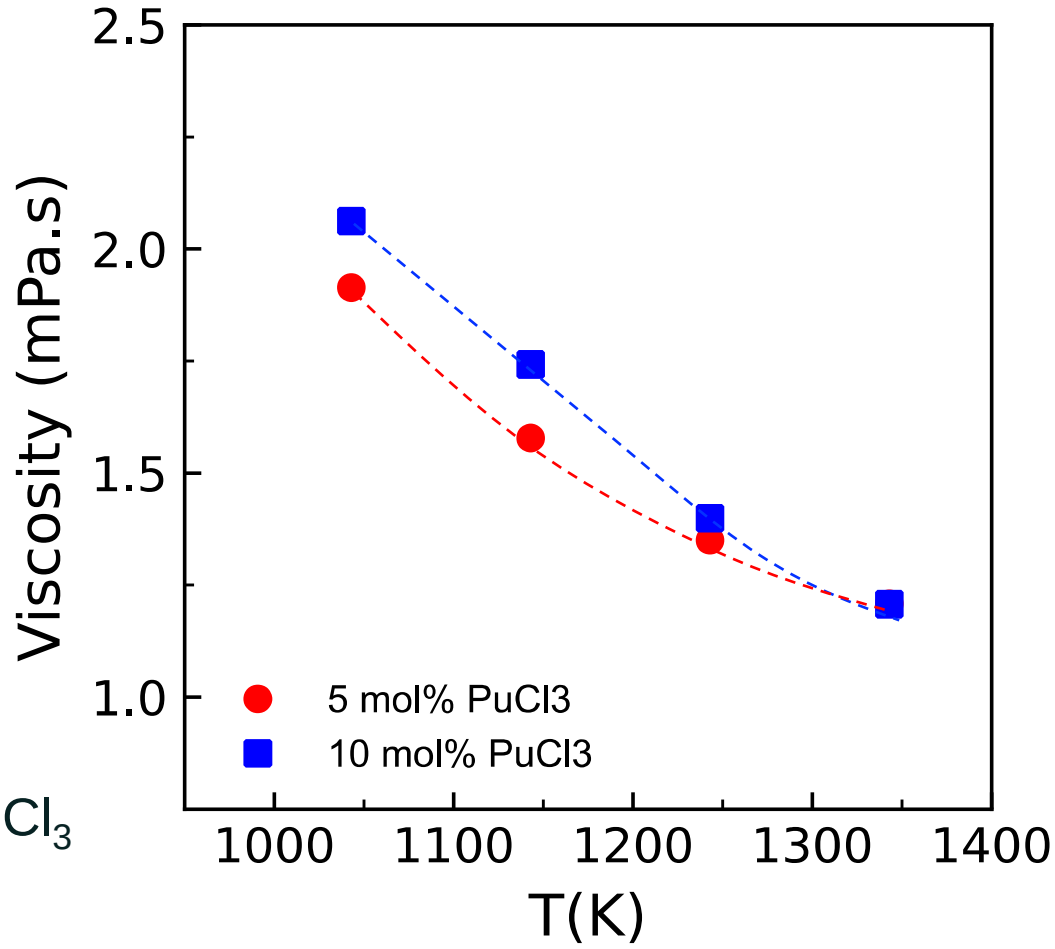


- Arrhenius-type diffusion
- Diffusion coefficient trend: $Na^+ > Cl^- > Pu^{3+} \geq U^{3+}$
- Diffusion coefficient of the whole system is similar to that of Cl.

Viscosity is determined from diffusion coefficient

- Stokes-Einstein relation[a] $\eta = \frac{k_B T}{2\pi D d}$
- Combination rule for hydrodynamic radius

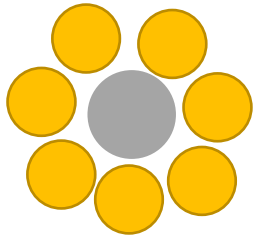
$$d = (d_{ClCl} + d_{NaNa} + d_{UU} + d_{PuPu} + 2d_{ClNa} + 2d_{ClU} + 2d_{ClPu})/10$$

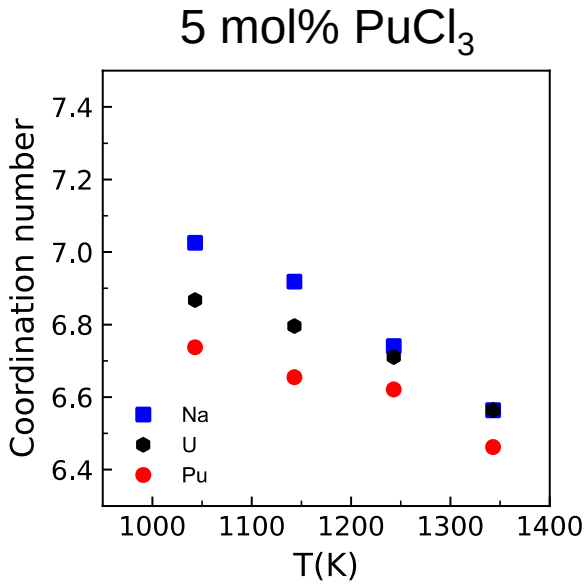


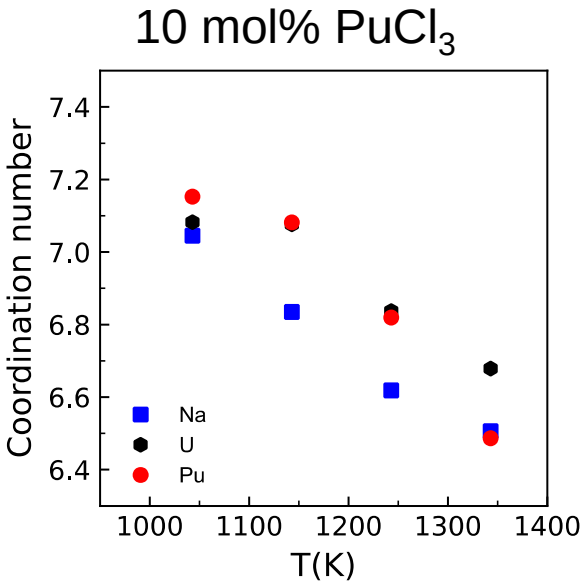
- Trend: the mixture is more viscous with higher content of $PuCl_3$
- We will also calculate the viscosity with pressure tensors

[a]D. Corradini, F.X. Coudert, R. Vuilleumier J. Chem. Phys., 144 104507 (2016)

Higher $PuCl_3$ concentration leads to higher coordination numbers







- Coordination number is decreased with temperature
- All cations have quite similar coordination numbers

Summary and Look ahead

- The density of the ternary system $PuCl_3$-NaCl-UCl_3 was determined for 2 compositions at 4 temperatures
- Transport (diffusion coefficient and viscosity) properties are composition- (and temperature) dependent
- Coordination number is increased with temperature
- We will investigate specific heat and thermal conductivity

Outputs

❑ **Publications**:

- Toni Karlsson, Scott C. Middlemas; Manh-Thuong Nguyen; Michael E. Woods; Kevin R. Tolman; Vassiliki-Alexandra Glezakou; Steven D. -Herrmann; Juliano Schorne-Pinto; Ryan D. Johnson; Shawn E. Reddish; Stephen A. Warmann; Patricia D. Paviet, "Synthesis and Thermophysical Property Determination of NaCl-$PuCl_3$ Salts" Journal of Molecular Liquids (under revision)
- Manh-Thuong Nguyen et al. "NaCl-$PuCl_3$ Molten Salts: Transport and Structural Properties from Machine Learning Interatomic Potentials and Data Analytics" (to be submitted)

❑ **Presentations:**

- Conference (invited talk): Manh-Thuong Nguyen "Computational Approaches to Study Heavy Element Materials" at "6th International Symposium on Frontiers in Materials Science (FMS 2022)", Phu Quoc, Vietnam, November 21-23, 2022.

Thank you

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