Atomistic Simulations for Thermophysical Properties of Molten Salts

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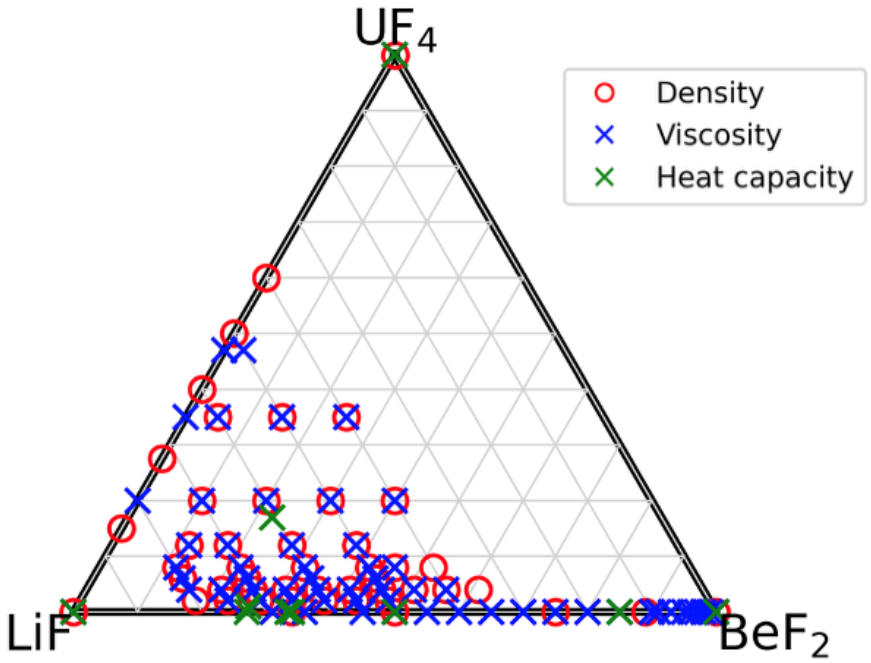
MSR Campaign Annual Review

04/22/2026

U.S. DEPARTMENT of ENERGY | Office of Nuclear Energy

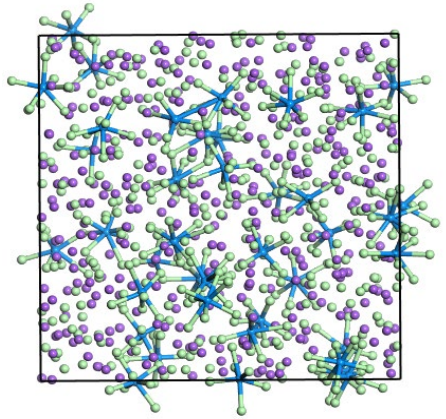
Background and Motivation

- **Thermophysical and thermochemical properties of molten salts are key for design and safe operation of MSRs.**
- **Challenges: High temperature, corrosive, radioactive.**
- **Reported thermophysical properties for some salts have large uncertainties and, in some cases, controversial.**
- ***Atomistic simulations* are critical to addressing these data and knowledge gaps and can provide insights into salt chemistry and inform Molten Salt Database (MSD), particularly MSD-TP.**



Available experimental data for $LiF\text{-}UF_4\text{-}BeF_2$

Atomistic Simulations



Classical Molecular Dynamics

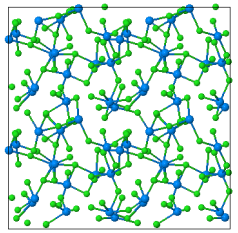
~1,000,000 atoms

~ns

Use empirical potentials

Empirical parameters needed

Intermediate-range structure, transport properties



Machine-learning Molecular Dynamics

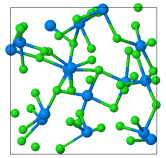
~ 100-1,000,000 atoms

~ns

Use advanced potentials

Potentials training needed

Density, heat capacity, and transport properties



Ab-initio Molecular dynamics (AIMD)

~100 atoms

~ps

Solve Schrödinger equation (DFT)

Computationally demanding

Density, heat capacity, mixing energy, local structure, oxidation states etc.

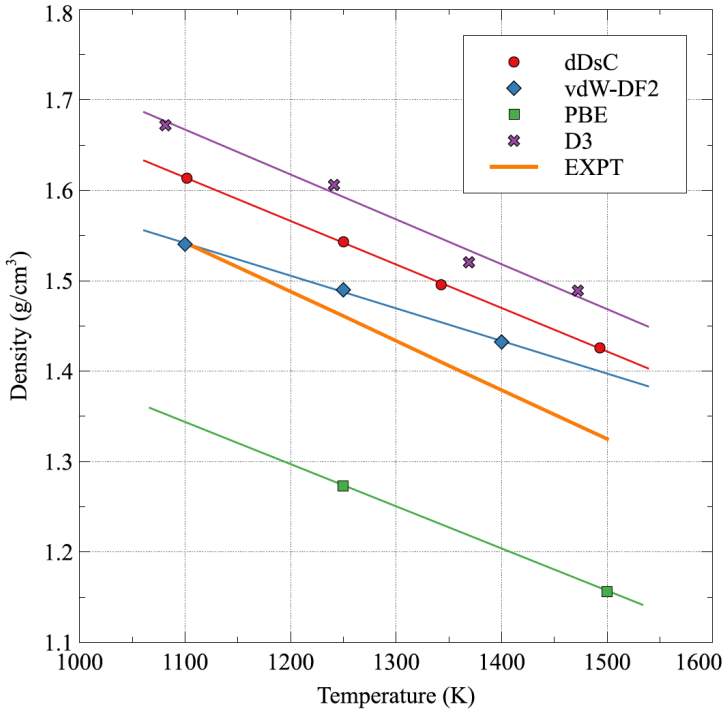
Status of NEAMS-Supported Salt System Simulations

	System	Status	Publication
AIMD	$NaCl\text{-}UCl_3$	✅	**Andersson** and Beeler. *J. Nucl. Mater.* 568 (2022): 153836.
	$KCl\text{-}UCl_3$		**Andersson**, **Wang**, Yang, and Beeler. *J. Nucl. Mater.* 599 (2024): 155226.
	Eutectic $NaCl\text{-}UCl_3$ + corrosion/fission products ($CrCl_3$,$CrCl_2$,$SrCl_2$,CsCl)	✅	**Wang**, Li, Yang, **Besmann**, and **Andersson**. *J. Nucl. Mater.* 615 (2025): 155949.
	$NaCl\text{-}UCl_4$	✅	In preparation
	$LiF\text{-}BeF_2\text{-}UF_4$	✅	In preparation
	$NaF\text{-}ZrF_4\text{-}UF_4$	🔄	On-going
Machine Learning	$MgCl_2\text{-}NaCl\text{-}KCl$	✅	**Jiang**, Guo, **Andersson**, **Schwen**, Benmore, Hoyt, **Spencer**, *J. Mol. Liq.* 403 (2024): 124854.
	$LiF\text{-}BeF_2\text{-}UF_4$	🔄	In preparation
	$LiF\text{-}BeF_2\text{-}ZrF_4$	🔄	On-going

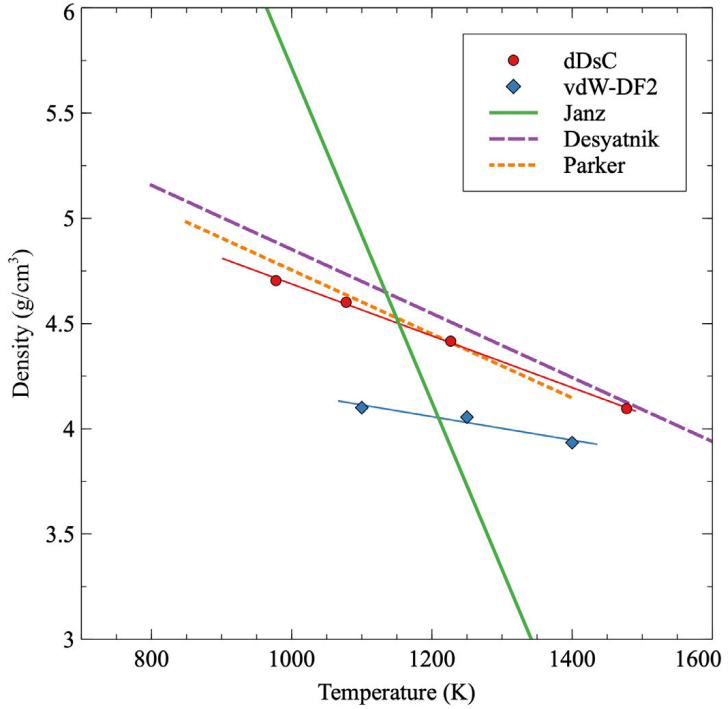
Pure Salts

- Validate computational protocols (dispersion corrections, magnetic moments, Hubbard U, system size, simulation length etc.) against available experimental data.

Density NaCl



Density UCl_3

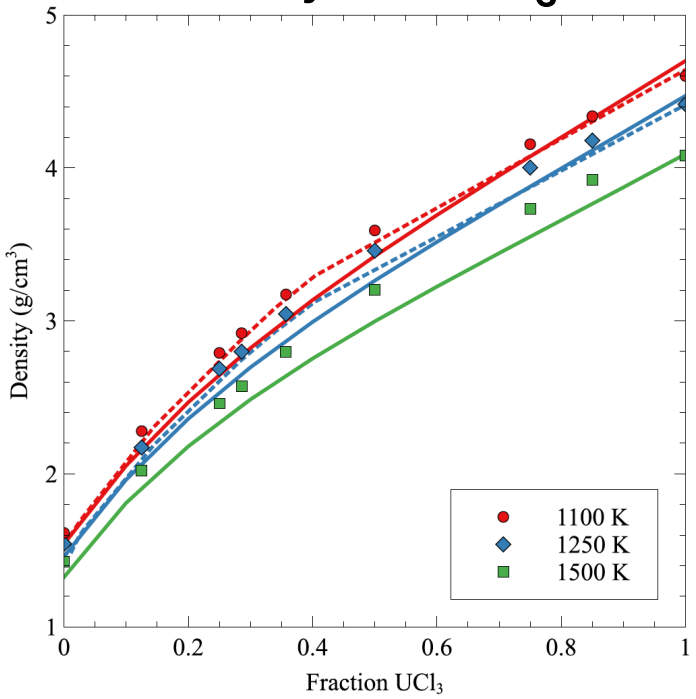


Andersson and Beeler. J. *Nucl. Mater.* 568 (2022): 153836.

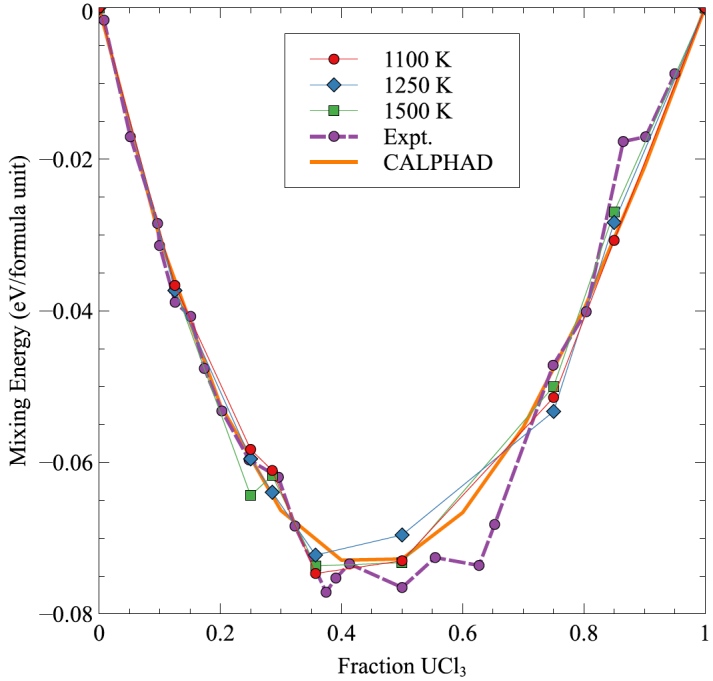
Binary Salts

- Thermophysical properties (density, mixing energy, heat capacity) are predicted within a few percent of experimental data.

Density NaCl-UCl_3

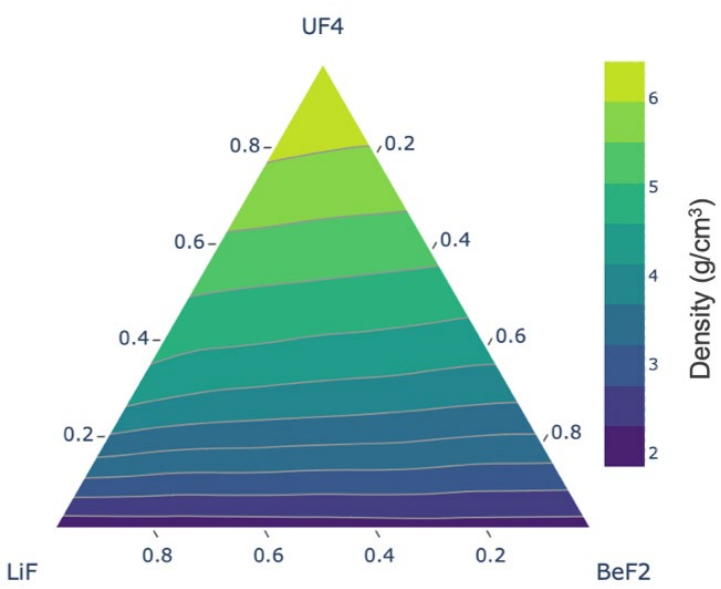


Mixing energy NaCl-UCl_3

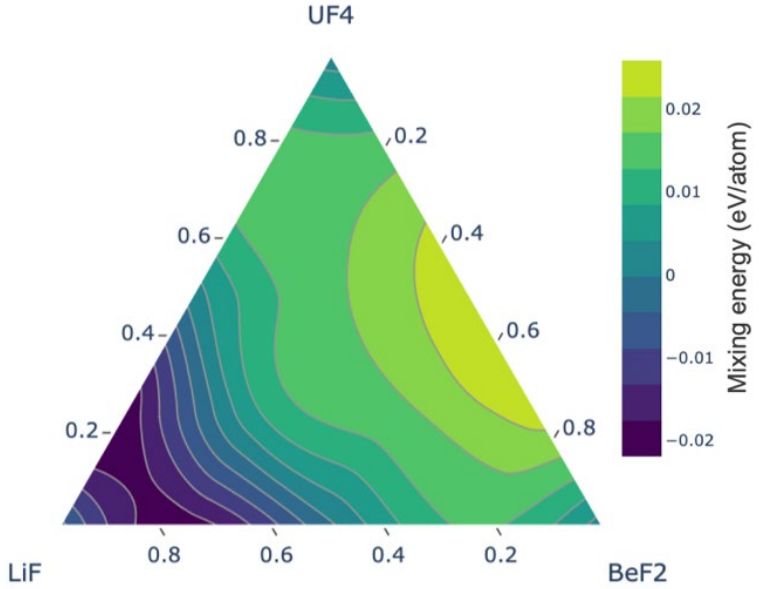


Ternary Salts

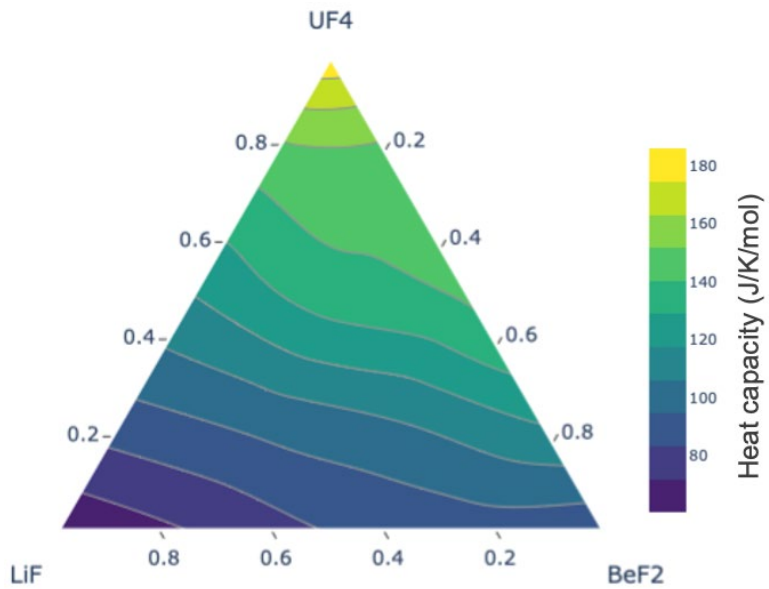
- Thermophysical properties (density, mixing energy, heat capacity) are evaluated for ternary salts across temperature and composition space.



Density



Mixing energy



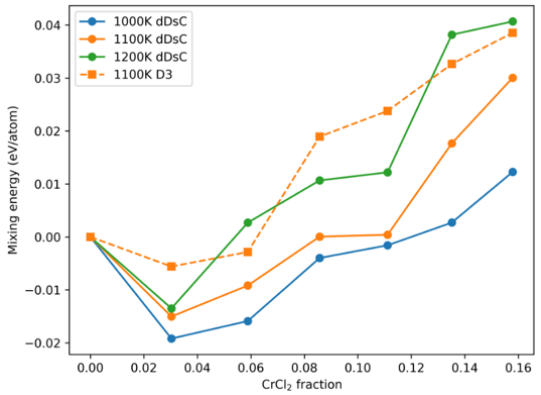
Heat capacity

LiF-BeF_2-UF_4

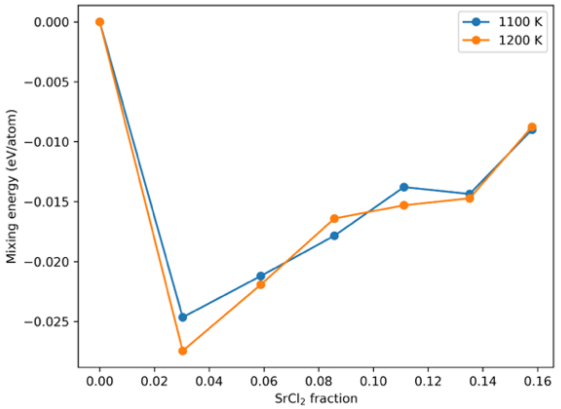
Wang, **Jiang**, **Birri**, and **Andersson**, in preparation.

$CrCl_3$, $CrCl_2$, $SrCl_2$, CsCl in Eutectic NaCl-UCl_3

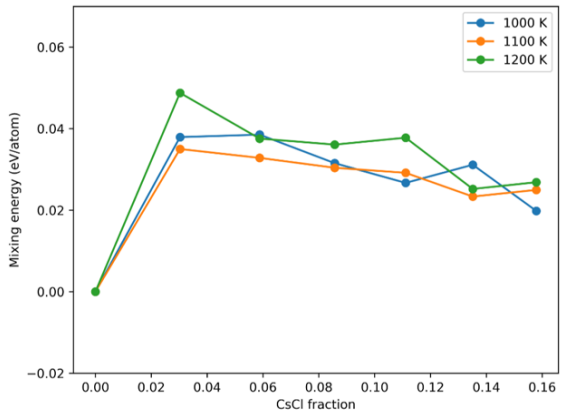
- $CrCl_3$, $CrCl_2$, $SrCl_2$, CsCl exhibit different behaviors in NaCl-UCl_3, driven by redox and coordination chemistry.
- Cr^{3+} instability in NaCl-UCl_3: $CrCl_3 + UCl_3 \rightarrow CrCl_2 + UCl_4$
- Negative mixing energies for $CrCl_2$ and $SrCl_2$, positive mixing energy for CsCl
- CsCl shows a strong tendency to form clusters.



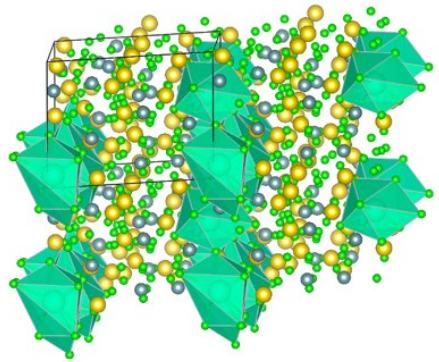
$CrCl_2$



$SrCl_2$



CsCl

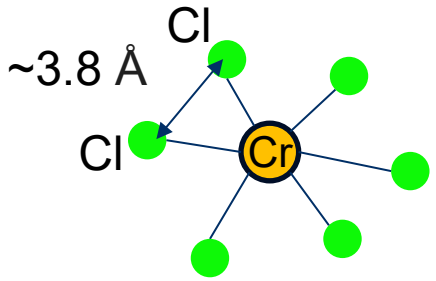


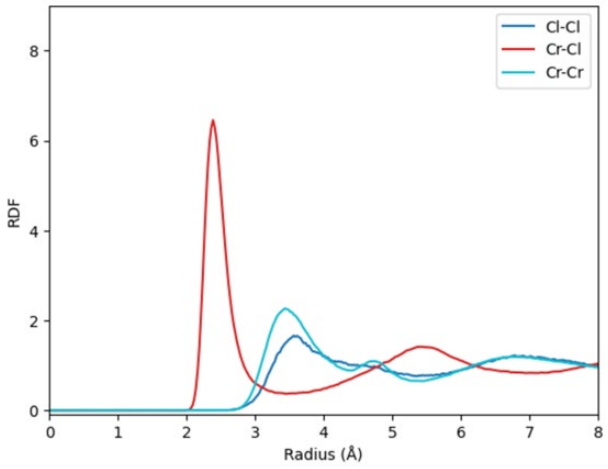
Snapshot of CsCl in NaCl-UCl_3

Wang, Li, Yang, **Besmann**, and **Andersson**. *J. Nucl. Mater.* 615 (2025): 155949.

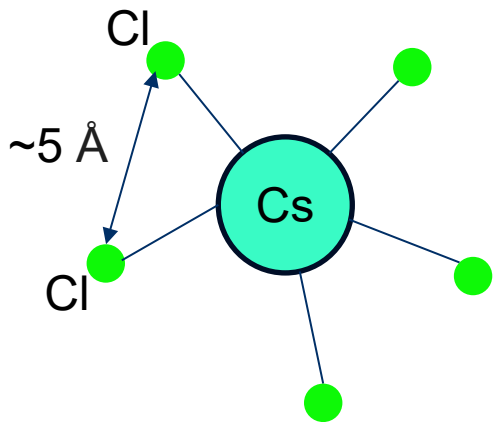
Coordination-Driven Mixing Behaviors

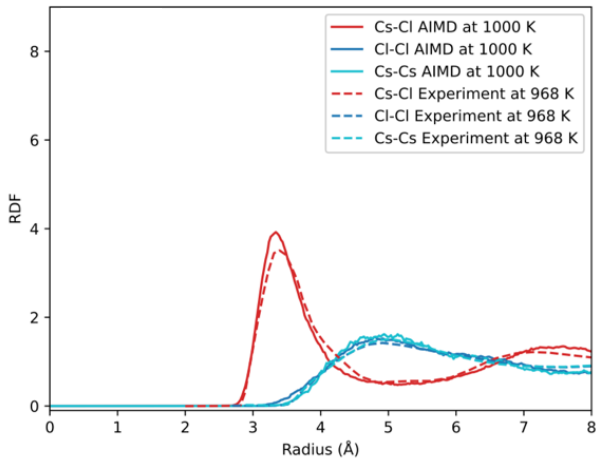
Pure $CrCl_2$



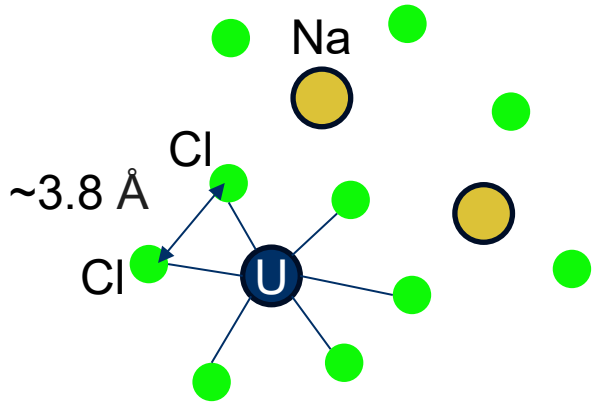


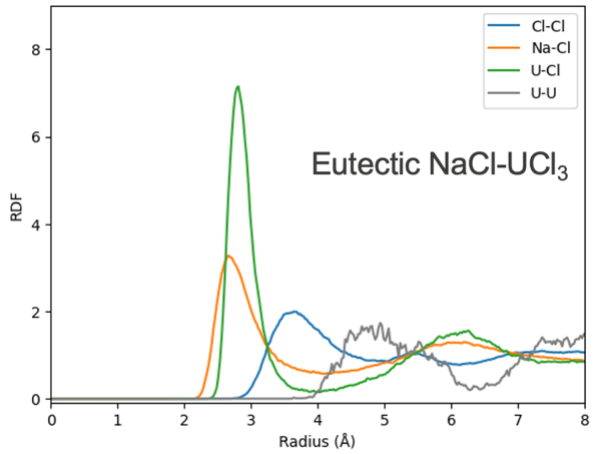
Pure CsCl





$NaCl\text{-}UCl_3$

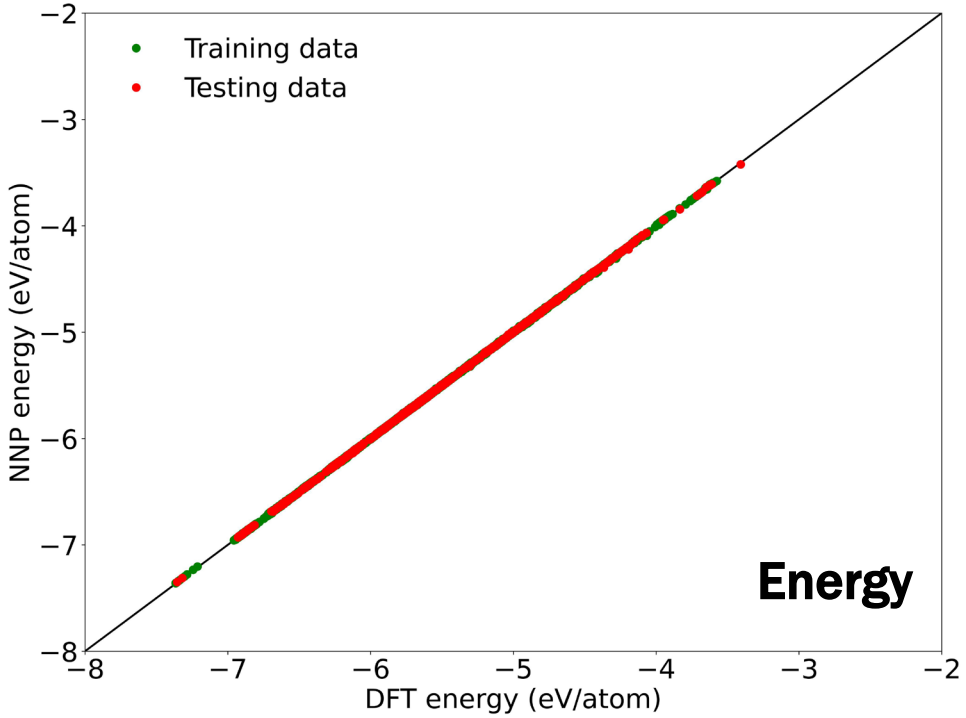


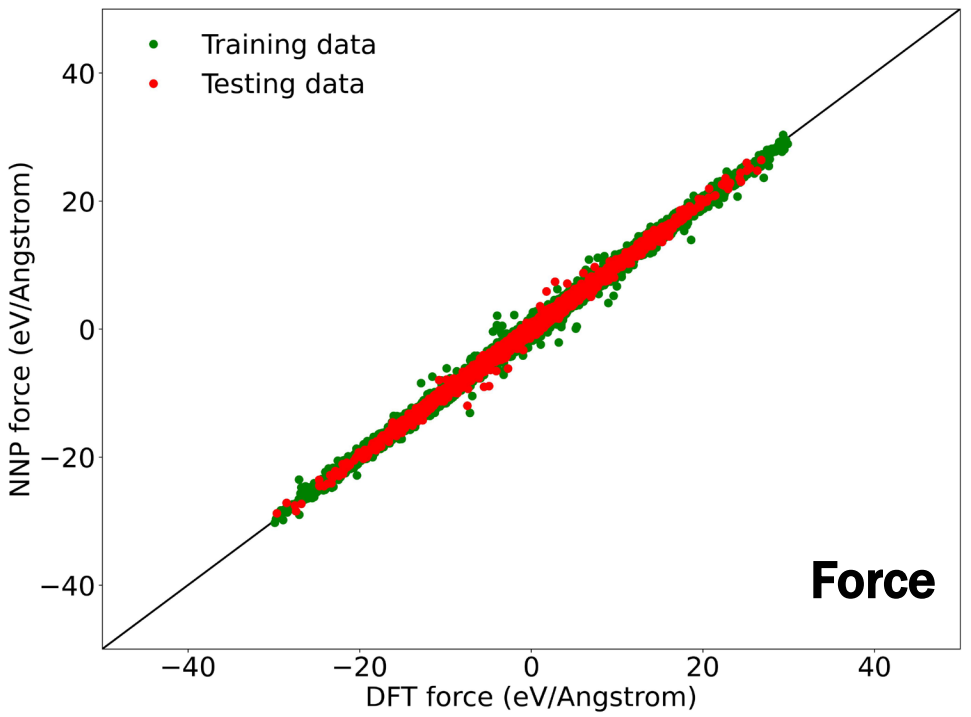


Wang, Li, Yang, **Besmann**, and **Andersson**. *J. Nucl. Mater.* 615 (2025): 155949.

Developing machine learning potentials for $LiF\text{-}BeF_2\text{-}UF_4$

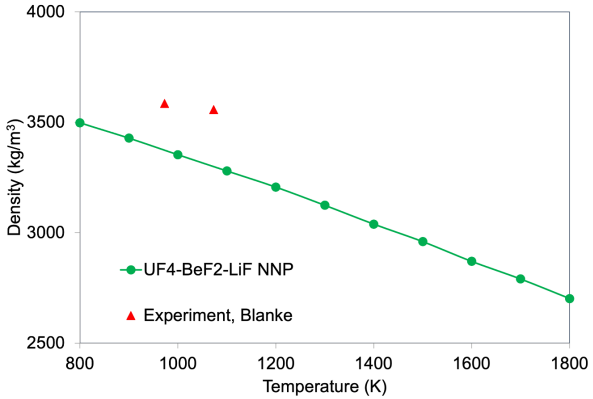
- A neural network-based machine learning potential (NNP) is developed.
- NNP can accurately reproduce DFT potential energy surfaces.
- NNP are orders of magnitude faster than DFT, enabling large-scale MD simulations.



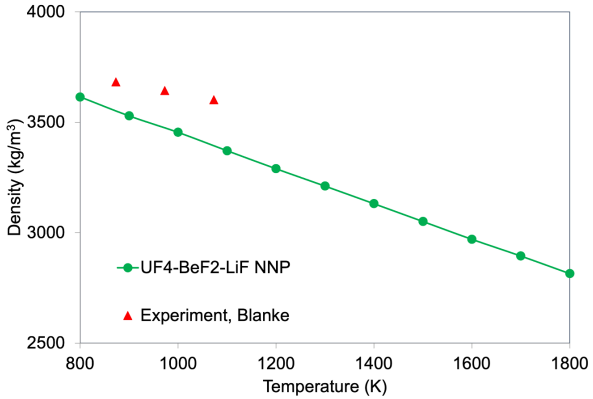


Salt Properties from MD Simulations Using NNPs

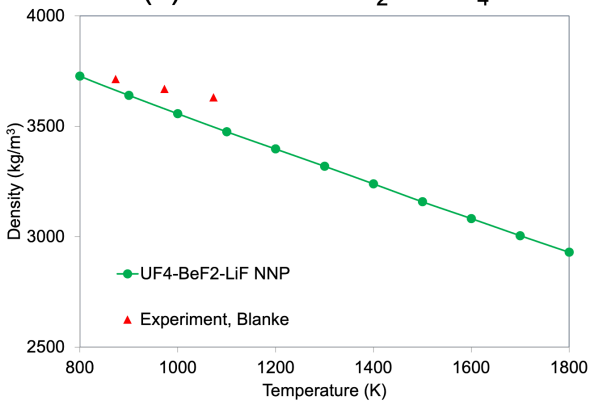
Density



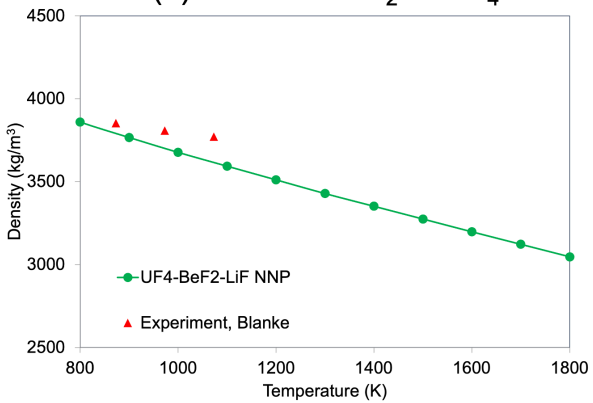
(a) 40LiF-40BeF_2-20UF_4



(b) 50LiF-30BeF_2-20UF_4

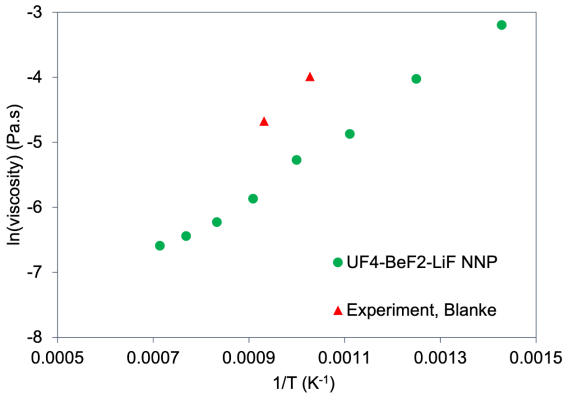


(c) 60LiF-20BeF_2-20UF_4

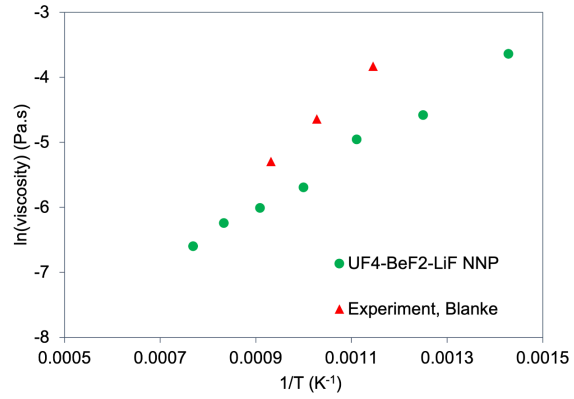


(d) 70LiF-10BeF_2-20UF_4

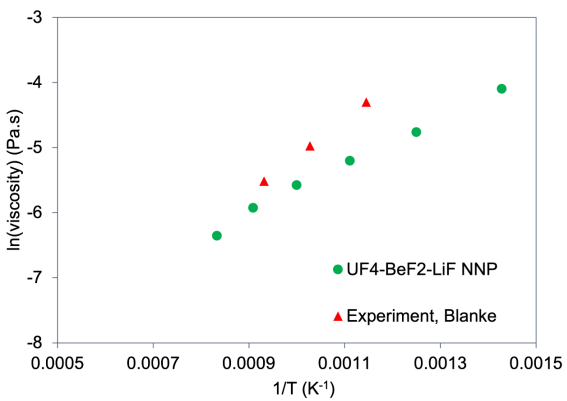
Viscosity



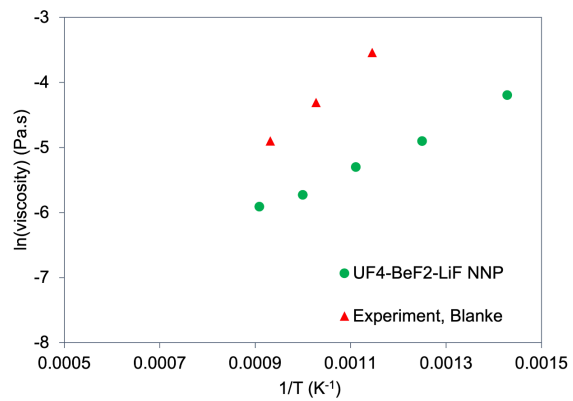
(a) 40LiF-40BeF_2-20UF_4



(b) 50LiF-30BeF_2-20UF_4



(c) 60LiF-20BeF_2-20UF_4



(d) 70LiF-10BeF_2-20UF_4

Summary

- AIMD simulations are performed for thermophysical properties of actinide-containing molten salts (NaCl-UCl_3, KCl-UCl_3, NaCl-UCl_4, NaCl-UCl_3+corrosion/fission products, LiF-BeF_2-UF_4, NaF-ZrF_4-UF_4 etc.).
- Machine learning potentials are developed and used to simulate transport properties of $MgCl_2$-NaCl-KCl and LiF-BeF_2-UF_4 molten salts.
- Atomistic simulations are critical to address data and knowledge gaps.
- Efforts underway to integrate the simulation results into MSD (MST-TP in particular).

Acknowledgements

- **Collaborators:**
  - Danny Perez, LANL
  - Ping Yang, LANL
  - Marisa Monreal, LANL

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- **Funding:**
  - DOE NE NEAMS

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- **Facilities:**

LANL Institutional Computing Program

INL High Performance Computing Center

Thank you for your attention!