Ab-initio and machine-learning molten salt property modeling

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NEAMS MSR external review

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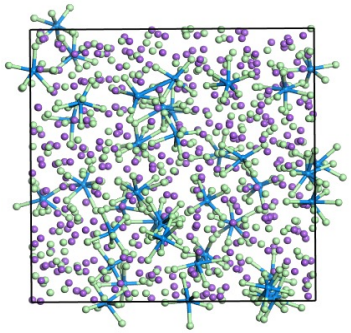
Background and motivation

- **Thermophysical and thermochemical properties of molten salts are key for design and safe operation of MSRs.**
- **Challenges: High temperature, corrosivity, radioactivity.**
- **Significant data and knowledge gaps remain, especially for actinide-containing salts, higher order systems, impurities such as corrosion/fission products.**
- **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, potentially inform MSTDB-TP and MSTDB-TC.**

Atomistic simulations



Classical Molecular Dynamics

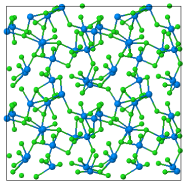
~1,000,000 atoms

~ns

Use empirical potentials

Empirical parameters needed

Intermediate-range structure, transport properties



Machine-learning Molecular Dynamics

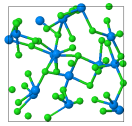
~1,000-100,000+ atoms

~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)

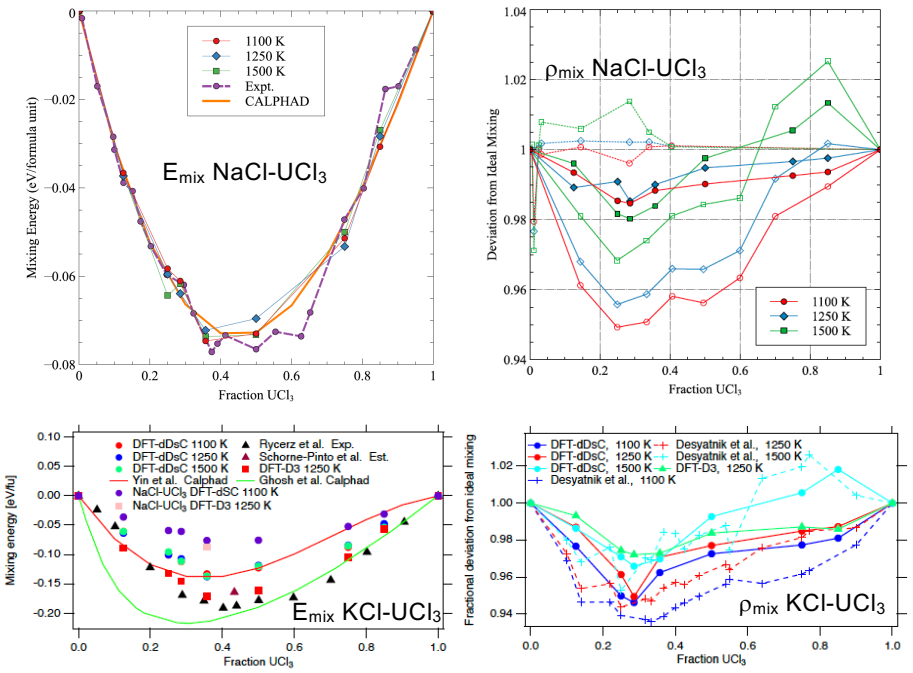
Computationally demanding

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

AIMD simulations of corrosion/fission products

- **Build on previous studies of base salts: NaCl-UCl_3[1], KCl-UCl_3[2], NaCl-$ThCl_4$[3].**
- **Focus on the effects of impurities ($CrCl_3$, $CrCl_2$, $SrCl_2$, CsCl) in NaCl-UCl_3.**
- **AIMD simulations of density, mixing energy, heat capacity, and density deviation.**
- **Structural properties are analyzed by coordination number and radial distribution function (RDF).**

Mixing energy and density of NaCl-UCl_3 and KCl-UCl_3



1. D. A. Andersson and B.W. Beeler, J. Nucl. Mater. 568 (2022) 153836.
2. D. A. Andersson, G. Wang, P. Yang, and B.W. Beeler, J. Nucl. Mat. 599 (2024) 155226.
3. G. Wang, B. Li, P. Yang, and D. A. Andersson, J. Mol. Liq. 385 (2023): 122347.

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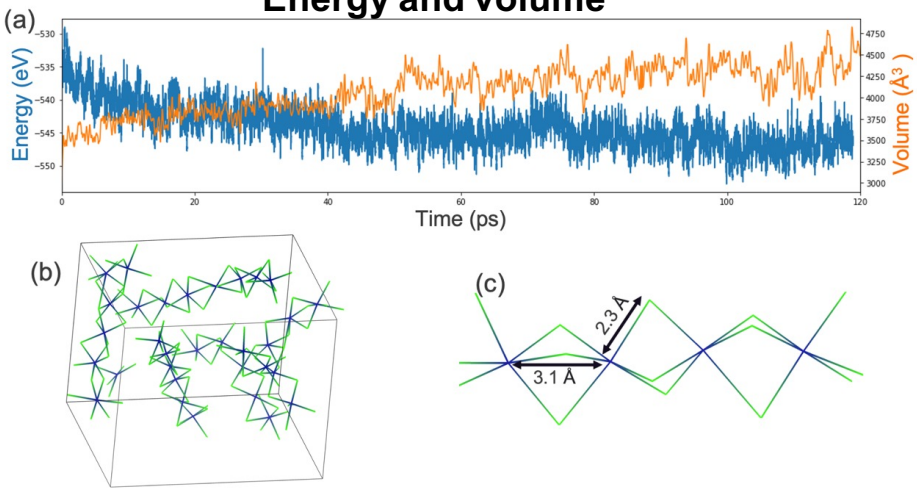
Pure $CrCl_3$ and $CrCl_3$ in eutectic NaCl-UCl_3

- **$CrCl_3$ exhibits valence fluctuations ($2CrCl_3$ → $2CrCl_2 + Cl_2$).**
- **Furthermore, the $CrCl_3$ system requires each Cr^{3+} ion to possess a coordination number of 6 to accommodate the additional Cl atoms.**
- **The stable configuration of 6-fold coordinated Cr can manifest as chain structures or adopt layered structures similar to those observed in bulk solid $CrCl_3$.**
- **Reaching relaxation into these specific configurations demands an extended simulation time and large supercells.**

Energy and volume



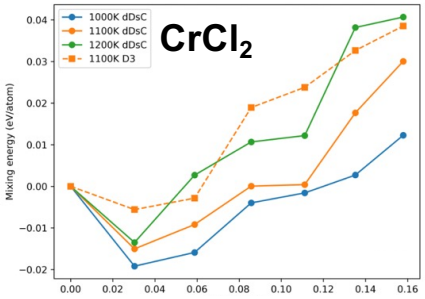
- **In the context of $CrCl_3$ in eutectic NaCl-UCl_3 molten salt, we found that the reaction:**

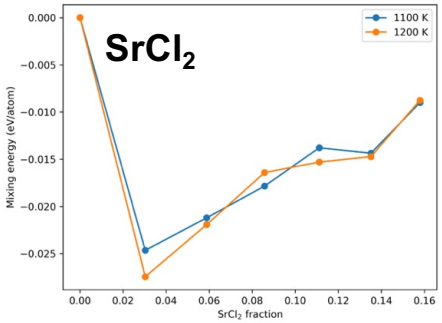
  $UCl_3 + CrCl_3 \rightarrow UCl_4 + CrCl_2$.
- **$CrCl_3$ is not stable in the binary NaCl-UCl_3 mixture.**

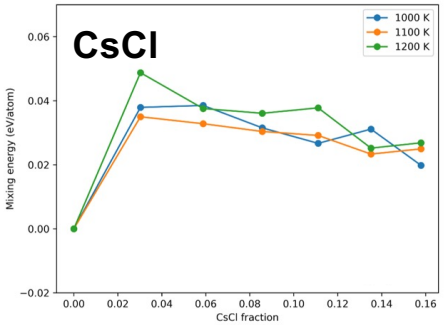
G. Wang, B. Li, P. Yang, T. M. Besmann, and D. A. Andersson, Revision submitted to J. Nucl. Mater.

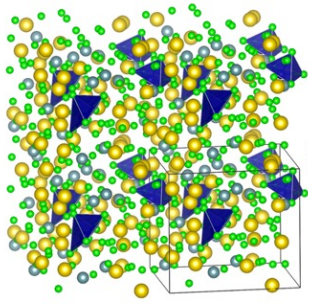
$CrCl_2$, $SrCl_2$ and CsCl in eutectic NaCl-UCl_3

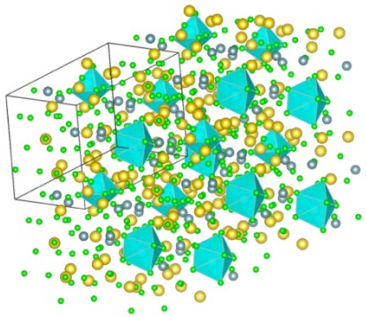
- $CrCl_2$ and $SrCl_2$ exhibit negative mixing energies at molar concentrations below 6%.
- CsCl shows positive mixing energy.

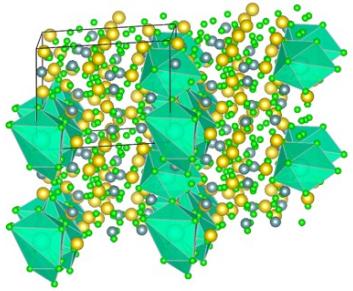








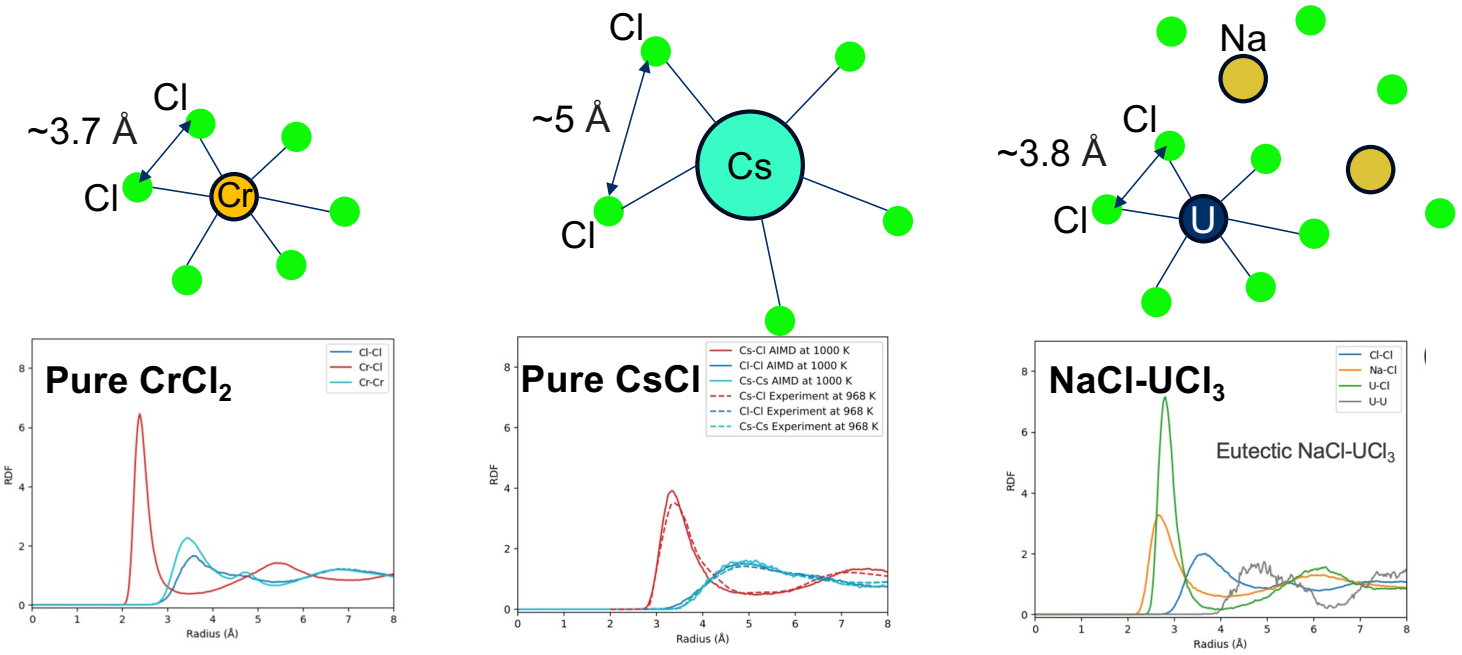




G. Wang, B. Li, P. Yang, T. M. Besmann, and D. A. Andersson, Revision submitted to J. Nucl. Mater.

$CrCl_2$, $SrCl_2$ and CsCl in eutectic $NaCl$-UCl_3

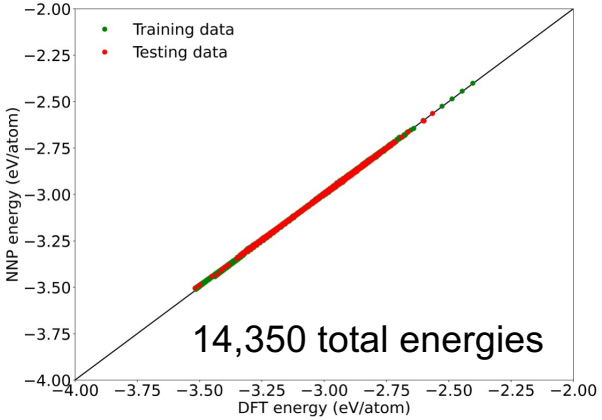
- **The mixing behavior is strongly correlated with the coordination chemistry of the salt.**

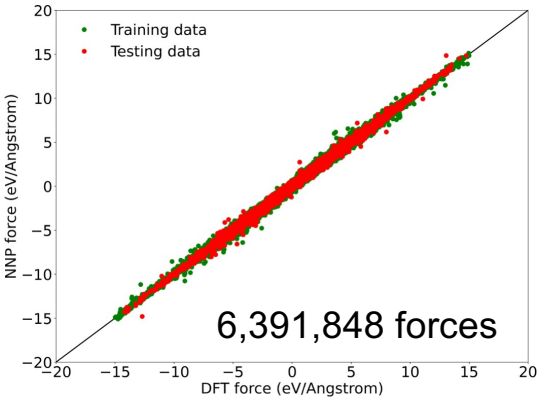


G. Wang, B. Li, P. Yang, T. M. Besmann, and D. A. Andersson, Revision submitted to J. Nucl. Mater.

Machine-learning MD simulations

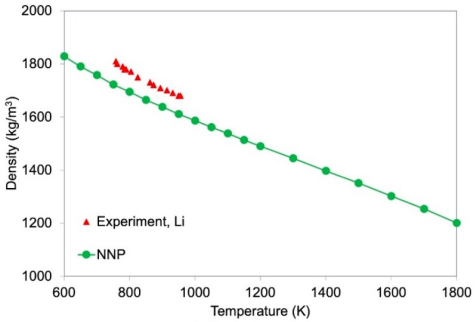
- **A neural network potential (NNP) for $MgCl_2$-NaCl-KCl (MNK) salts was developed using DFT training dataset.**
- **The NNP can satisfactorily reproduce the DFT calculated energies and forces while being many orders of magnitudes faster than DFT.**
- **It can be employed as a low-cost surrogate model for DFT during the calculations of the thermophysical properties of MNK salts.**



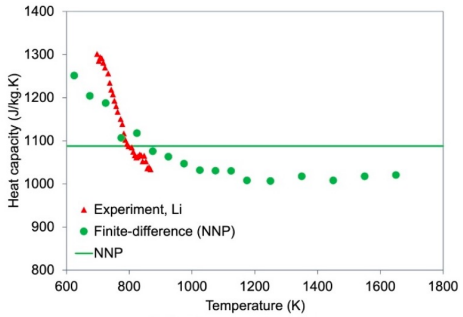


C. *Jiang et al., npj Comput. Mater.10*, no. 1 (2024): 21.

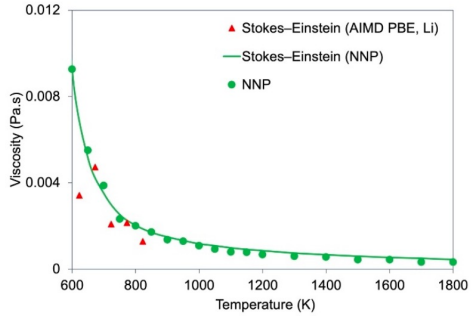
Machine-learning MD simulations



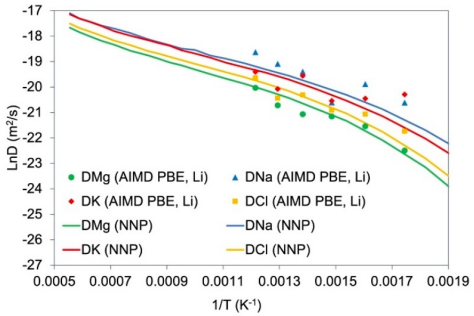
(a) Density



(b) Heat capacity

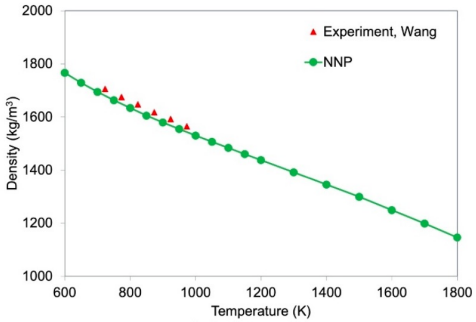


(c) Viscosity

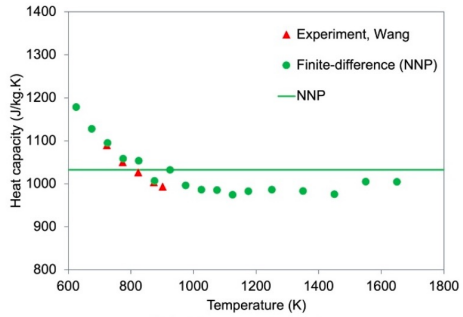


(d) Diffusivity

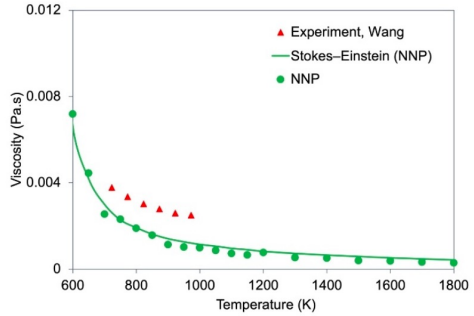
45$MgCl_2$+33NaCl+22KCl



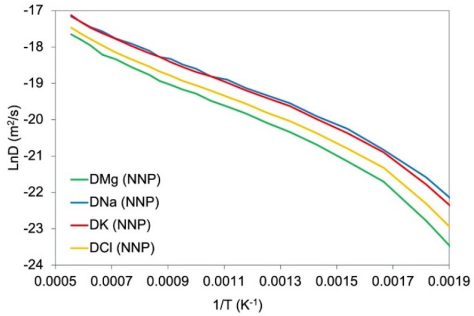
(a) Density



(b) Heat capacity



(c) Viscosity



(d) Diffusivity

38$MgCl_2$+21NaCl+41KCl

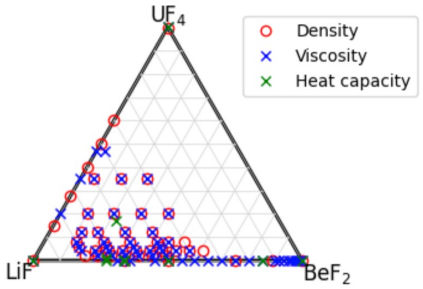
Summary and Ongoing work

FY24:

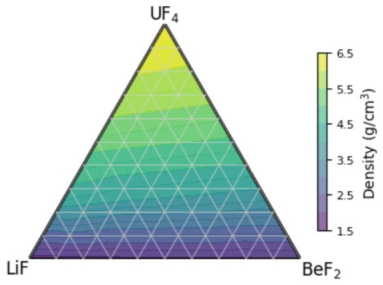
- **AIMD simulations of corrosion/fission products ($CrCl_3$, $CrCl_2$, $SrCl_2$, CsCl) in NaCl-UCl_3.**
- **Machine-learning MD simulations of $MgCl_2$-NaCl-KCl (MNK) salts.**

FY25:

- **Thermophysical properties of LiF-BeF_2-UF_4 salts using AIMD and machine-learning MD.**
- **Inform the development of MSTDB-TP.**



Experiment data[1,2]



AIMD simulated density

1. J. C. Ard et al., *J Nucl. Mater.* 563 (2022): 153631.
2. A. Redkin et al., *J. Mol. Liq.* 341 (2021): 117215.

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