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Modeling Thermophysical Properties of Fluoride Molten Salts

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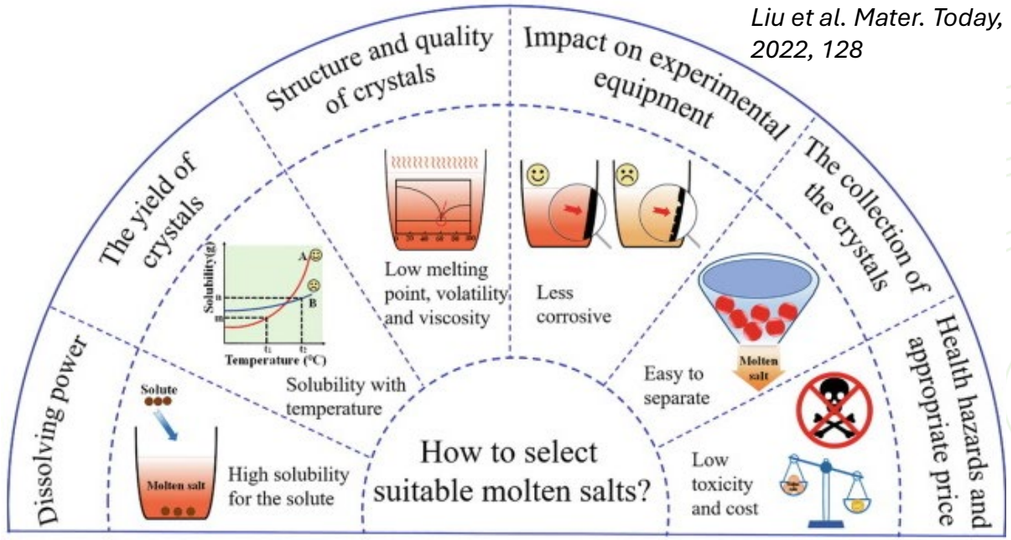
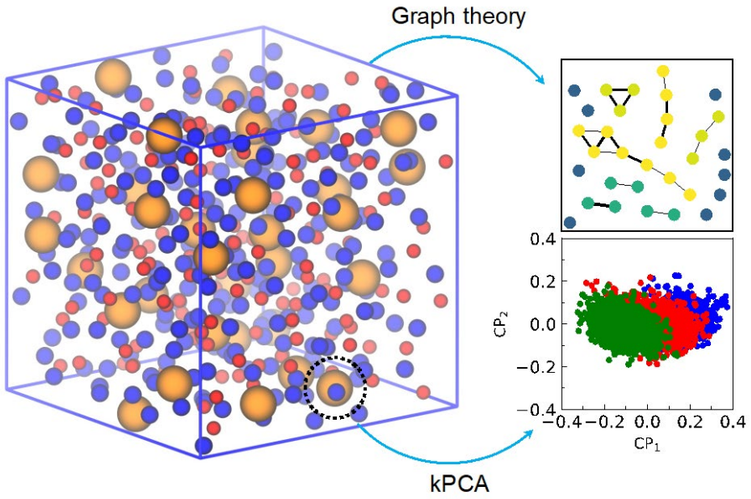
Atomistic simulation aided by AI tools provide an emerging approach for thermo-physical/chemical property prediction

- Computational evaluation of molten salt properties plays an important role to facilitate experimental design of molten salt reactors.
- Experimental measurements:
  - Time, labor, and equipment cost
  - Underlying chemical reactivity and transport
  - Limited data, often unknown errors
- Computational modeling:
  - Fidelity-cost tradeoff
  - AI-acceleration: MLIP, LLM, etc…



Liu et al. Mater. Today, 2022, 128



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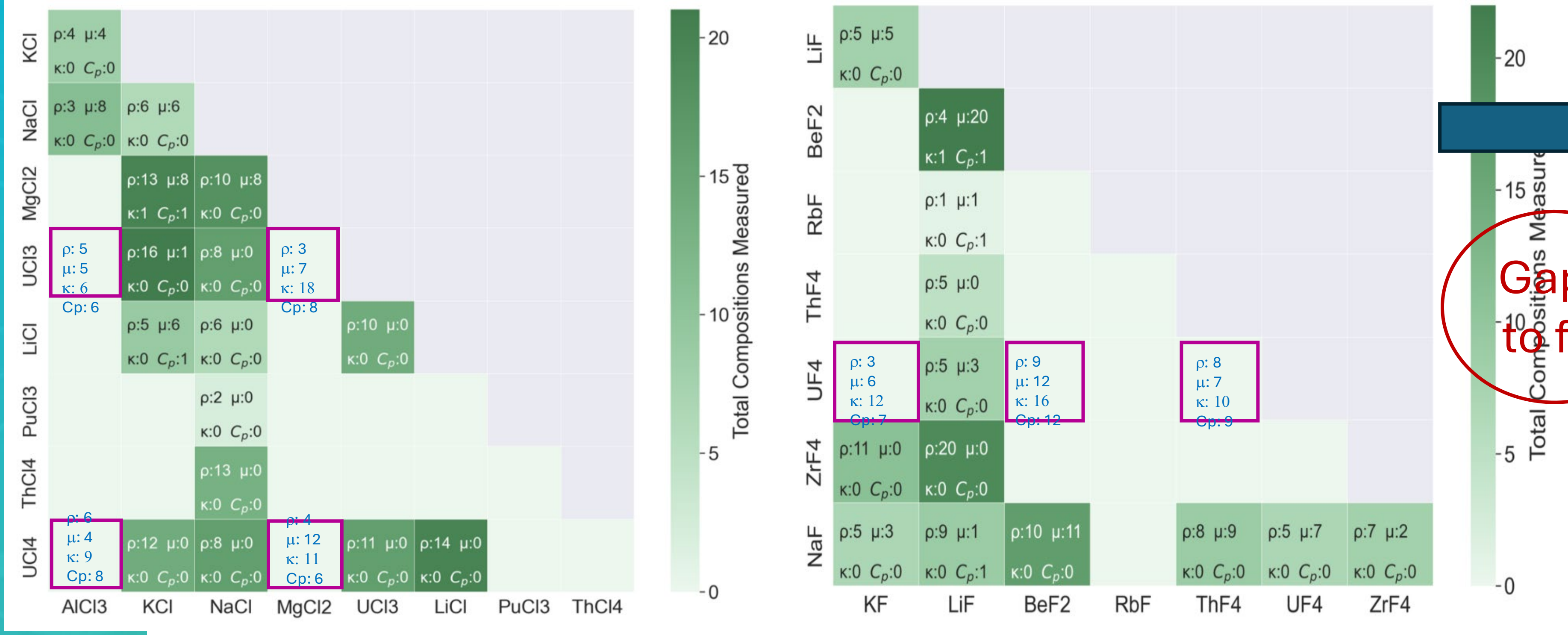
LLM tools facilitate literature search, but significant verification by domain expertise is still required

- LLM tools:
  - FutureHouse Platform (https://www.futurehouse.org)
  - OpenAI ChatGPT
  - Anthropic Claude

Manual verification for atomistic simulations of U-containing molten salts



Number of systems in paper/reports

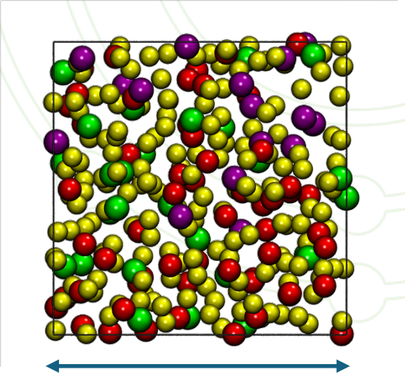
Gaps to fill

Component			ρ	C_p	η
UCl_3	NaCl		21	38	1
UCl_3	KCl		19	26	0
UCl_4	NaCl		12	12	0
UF_4	LiF		22	11	11
UF_4	NaF		10	0	10
UF_4	KF		10	0	10
UF_4	LiF	NaF	1	1	1
UF_4	LiF	BeF_2	8	0	6
UCl_3	KCl	NaCl	2	2	2
UCl_3	KCl	LiCl	5	0	0

Number of data points from existing simulation papers

Ab initio molecular dynamics provide first-principles structure and property information

- AIMD simulations using CP2K under 1100K for 20 ps. Simulation box size is determined by experimental density values from MSD-TP
- UF_4+NaF (0.22-0.78 mol%)
- NaF-KF-UF_4 (57-16-27 mol%)
- UCl_3+NaCl (0.37-0.63 mol%)
- Structural analysis (e.g., pair distribution function) highlights their structural differences and helps thermal conductivity evaluation

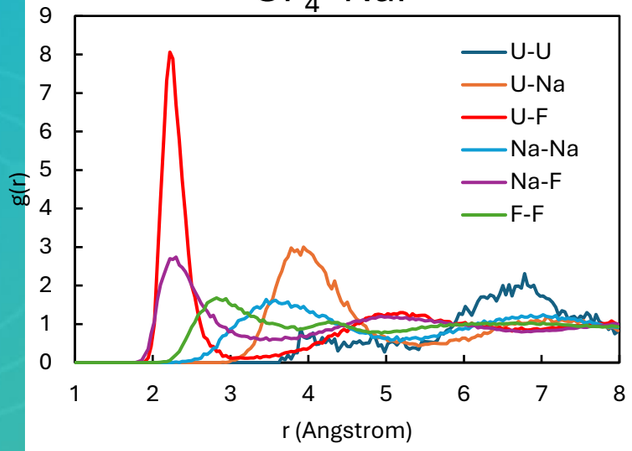


1.7 nm

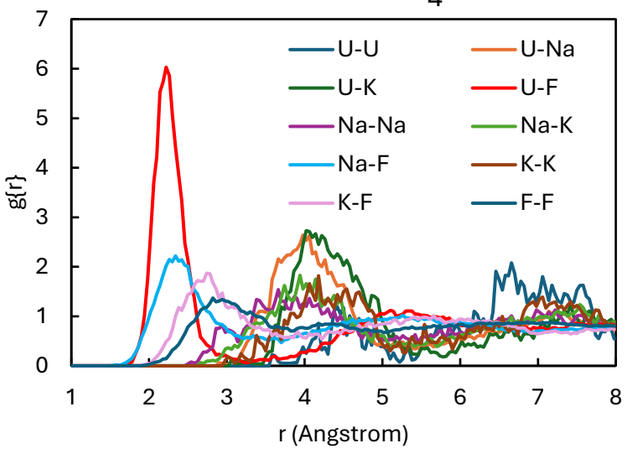
AIMD model of 281 atoms for NaF-KF-UF_4

Estimated thermal conductivity

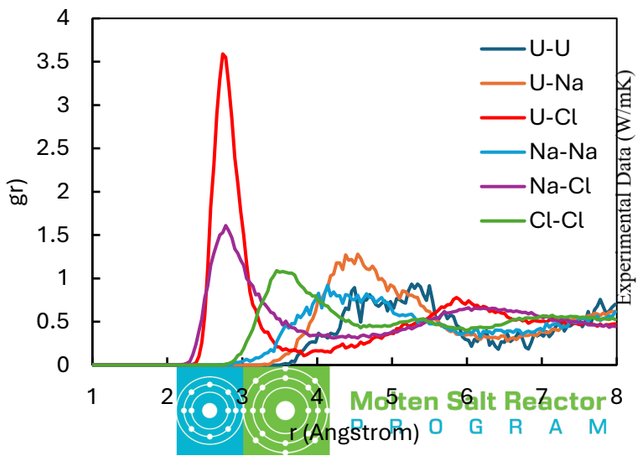
UF_4+NaF

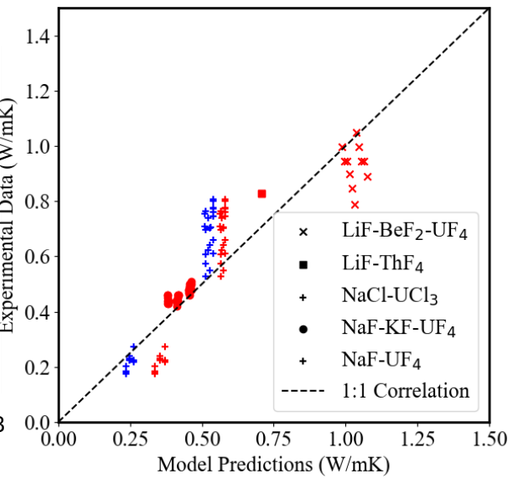


NaF-KF-UF_4



UCl_3+NaCl

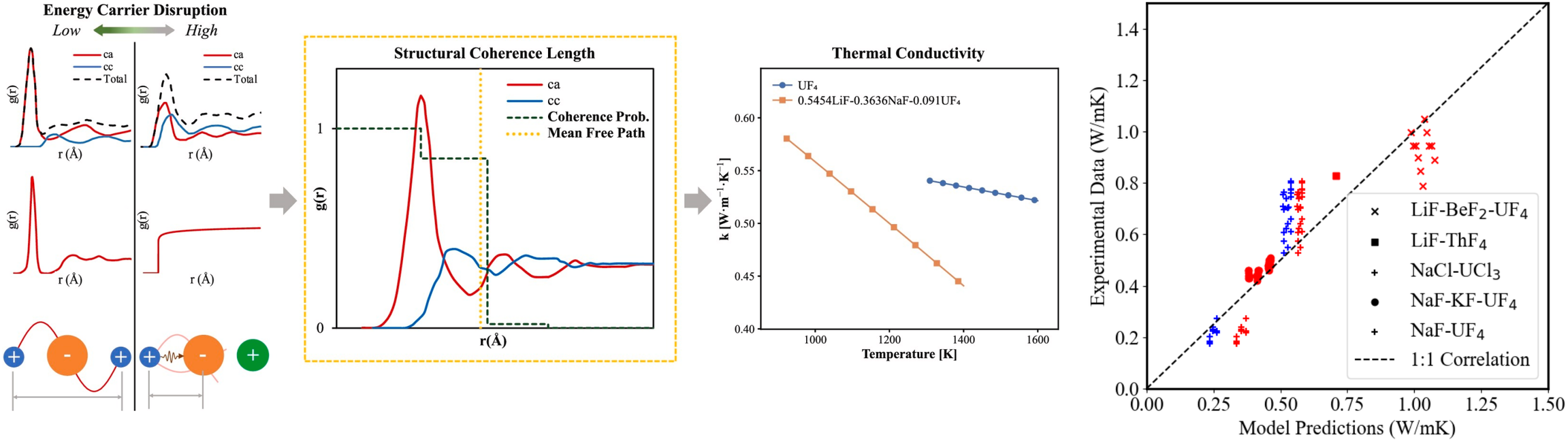




Structural information from AIMD also helps estimate the thermal conductivity

- A recently developed method[1]: Link short-range order structure to mean free path of energy carriers via a simple structural coherence model informed by the partial pair distribution function.

Estimated thermal conductivity for UF_4- and UCl_3-containing molten salts using g(r) from our AIMD simulations[2]



[1] Numbers, et al., Journal of Molecular Liquids (2026) 452, 129502
[2] Manuscript in preparation

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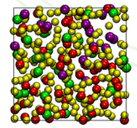
Classical molecular dynamics allow for a larger modeling system and a longer time scale

- Machine learning interatomic potential (MLIP) provides higher accuracy with affordable computational cost.
- Foundational MLIPs (e.g., MACE, UMA, etc.) are less explored for molten salt systems since they are not primarily developed for these systems (with much less training data).
- The transferability of these foundational MLIPs is still under evaluation/re-training

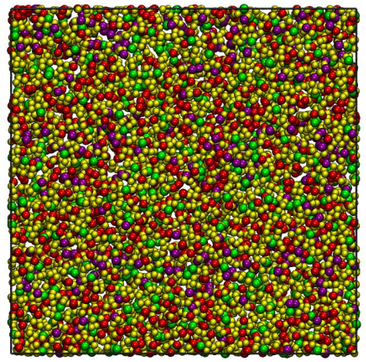
$NaF\text{-}KF\text{-}UF_4$
(57-16-27 mol%)

AIMD model of 281 atoms



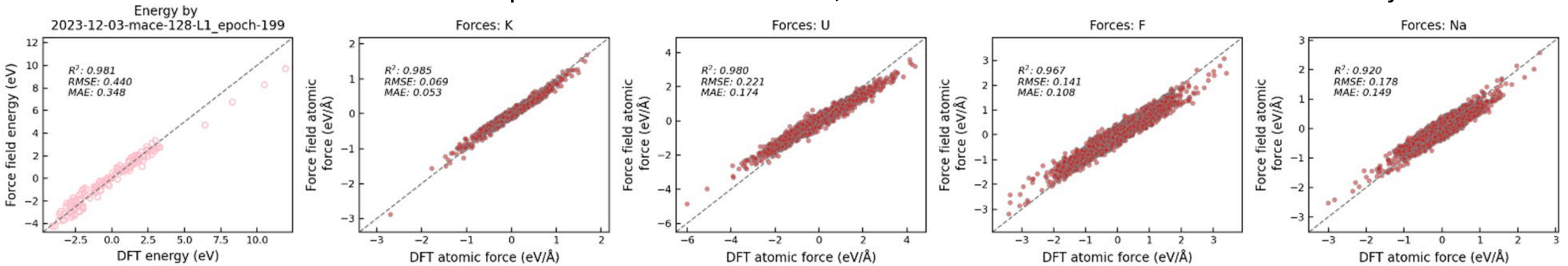
1.7 nm

CMD model 7587 atoms



5.2 nm

Compared to our AIMD results, MACE-2023 MLIP shows a decent accuracy.



Energy comparison

Atomic force comparison by atom species

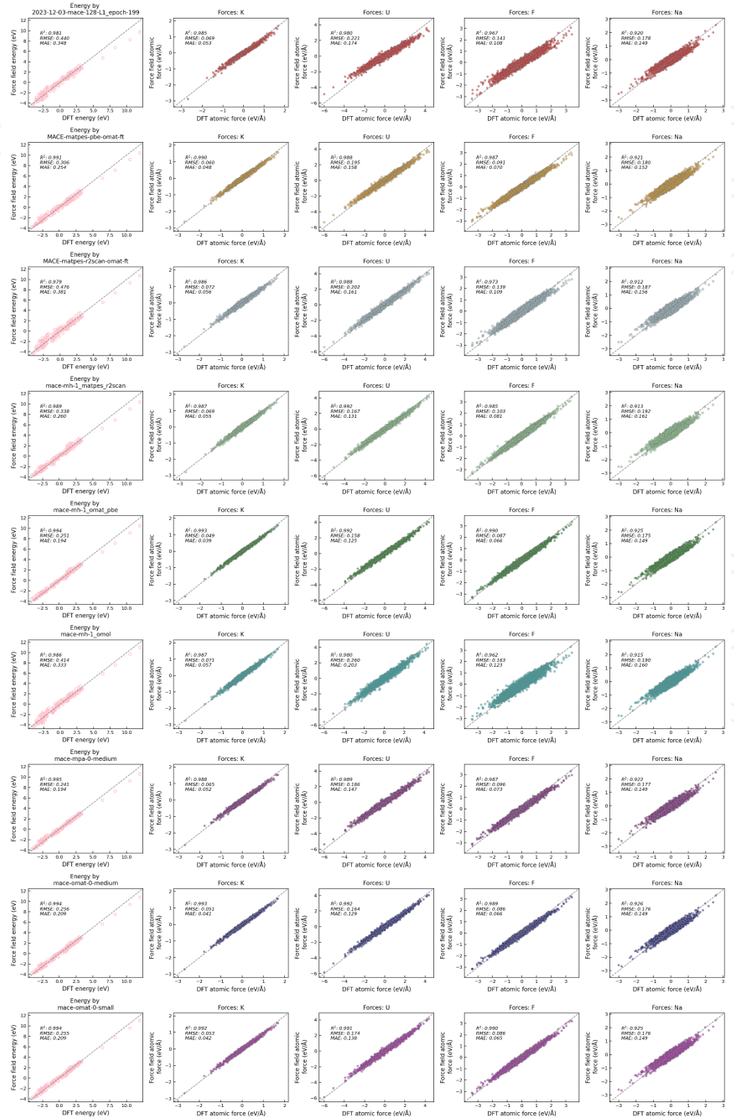
In fact, the MLIPs of MACE family generally show good energy and atomic force accuracy for UF_4+NaF and UF_4+NaF+KF

- Various versions of MACE MLIPs

Model Name	Training Dataset	Level of Theory	Target System	Notes
MACE-MP-0	MPTrj	DFT (PBE+U)	Materials	Initial release of foundation model.
MACE-MPA-0	MPTrj+ sAlex	DFT (PBE+U)	Materials	Improved accuracy for materials, improved high pressure stability.
MACE-OMAT-0	OMAT	DFT (PBE+U) VASP 54	Materials	Most accurate for phonons
MACE-MATPES-0	MATPES	PBE/R2SCAN	Materials	PBE and R2SCAN models for MATPES dataset. Latest recommended ones.
MACE-OFF23	SPICE v1	DFT (wB97M+D3)	Organic Chemistry	Initial release covering neutral organic chemistry.
MACE-MH-1/0	OMAT OMOL OC20 MATPES	DFT (PBE/R2SCAN /wB97M-VV10)	Materials Molecules Surfaces	Latest multihead model for materials chemistry.

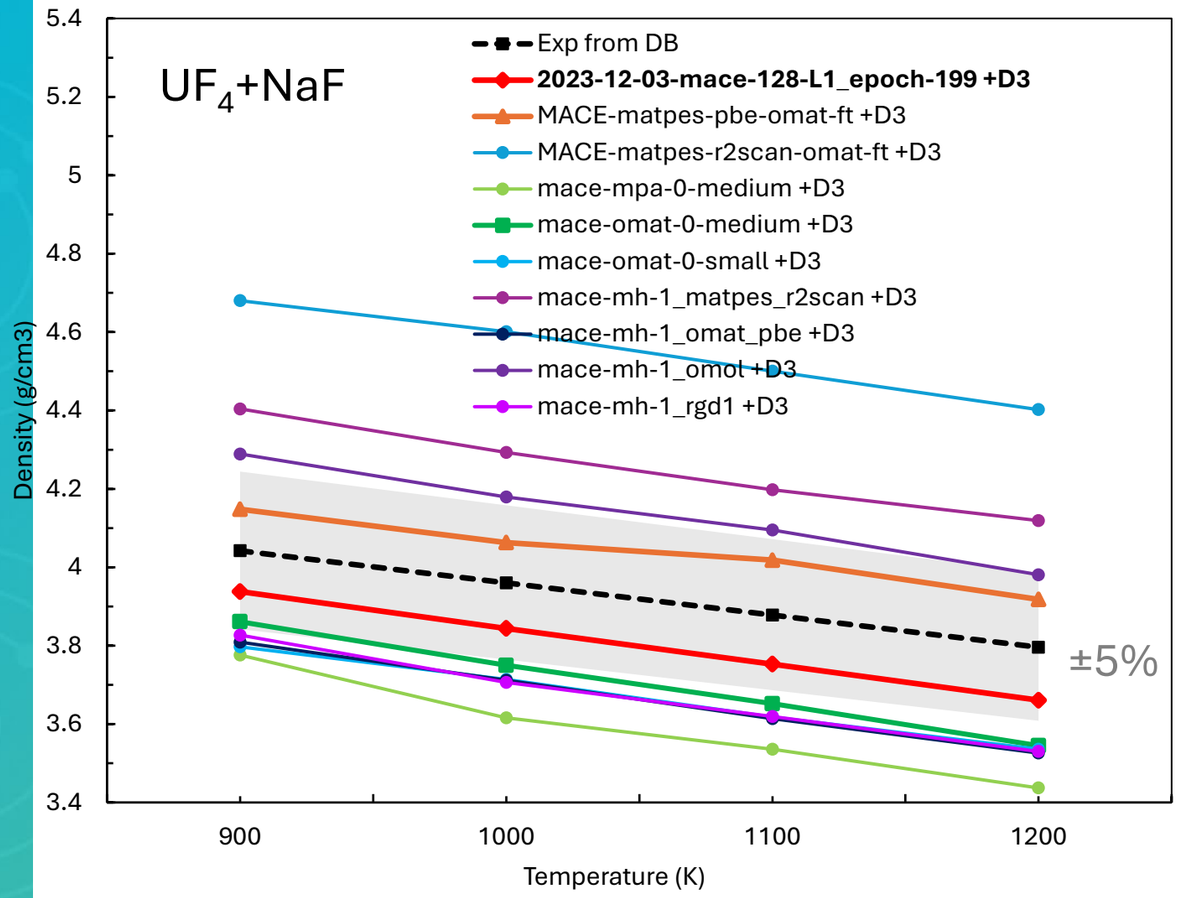
Energy and forces compared to AIMD

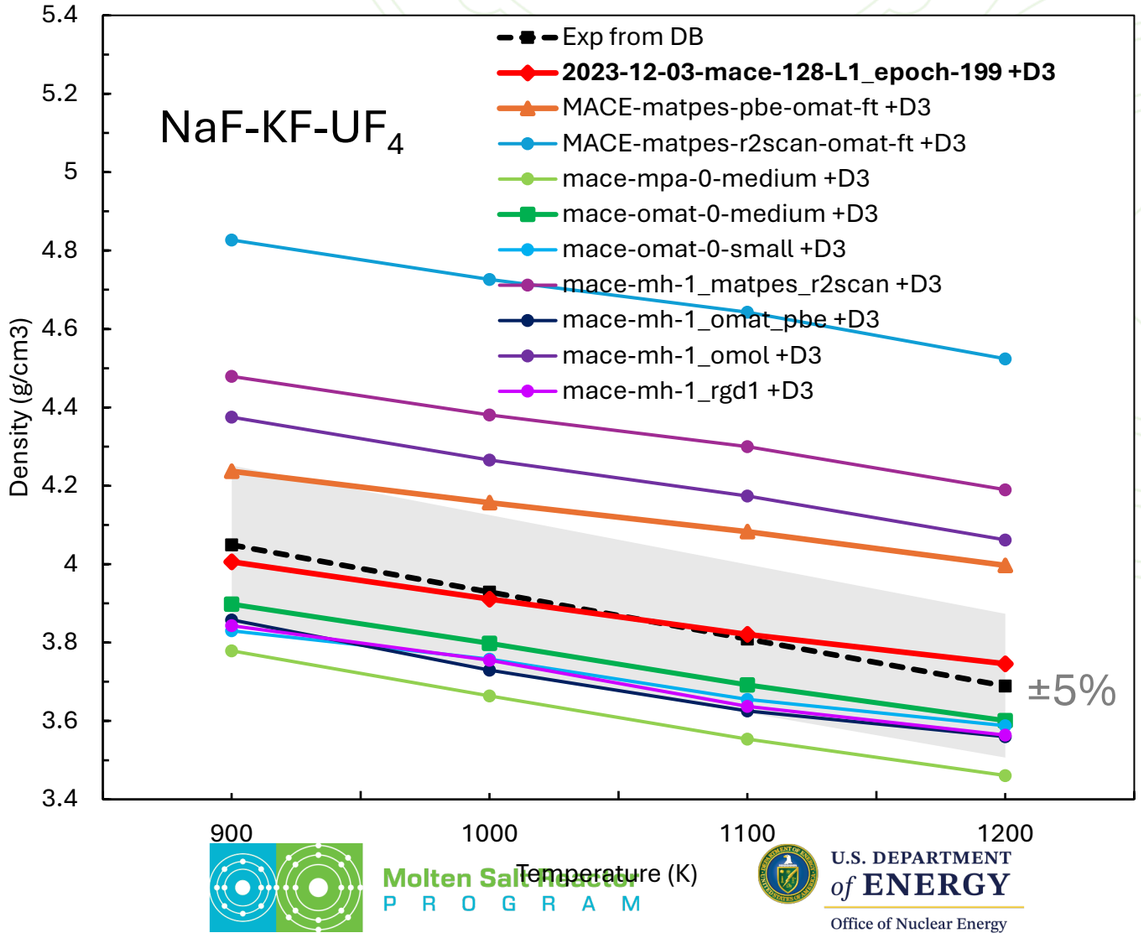
Different MACE MLIPs



The predicted density by the earlier version of MACE MLIP agrees more with experimental density values

- Use UF_4+NaF (0.22-0.78 mol%) and NaF-KF-UF_4 (57-16-27 mol%) as two benchmark systems





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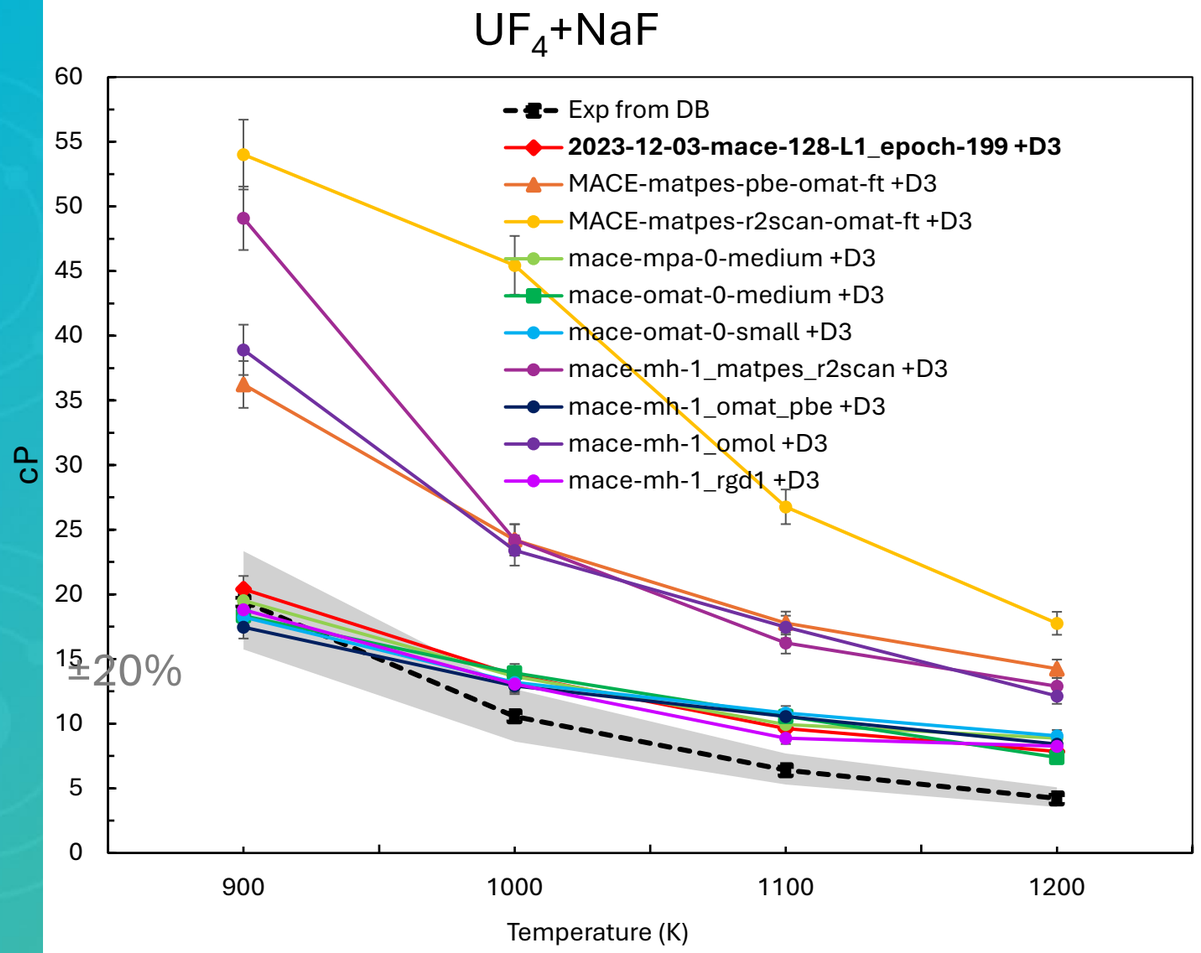
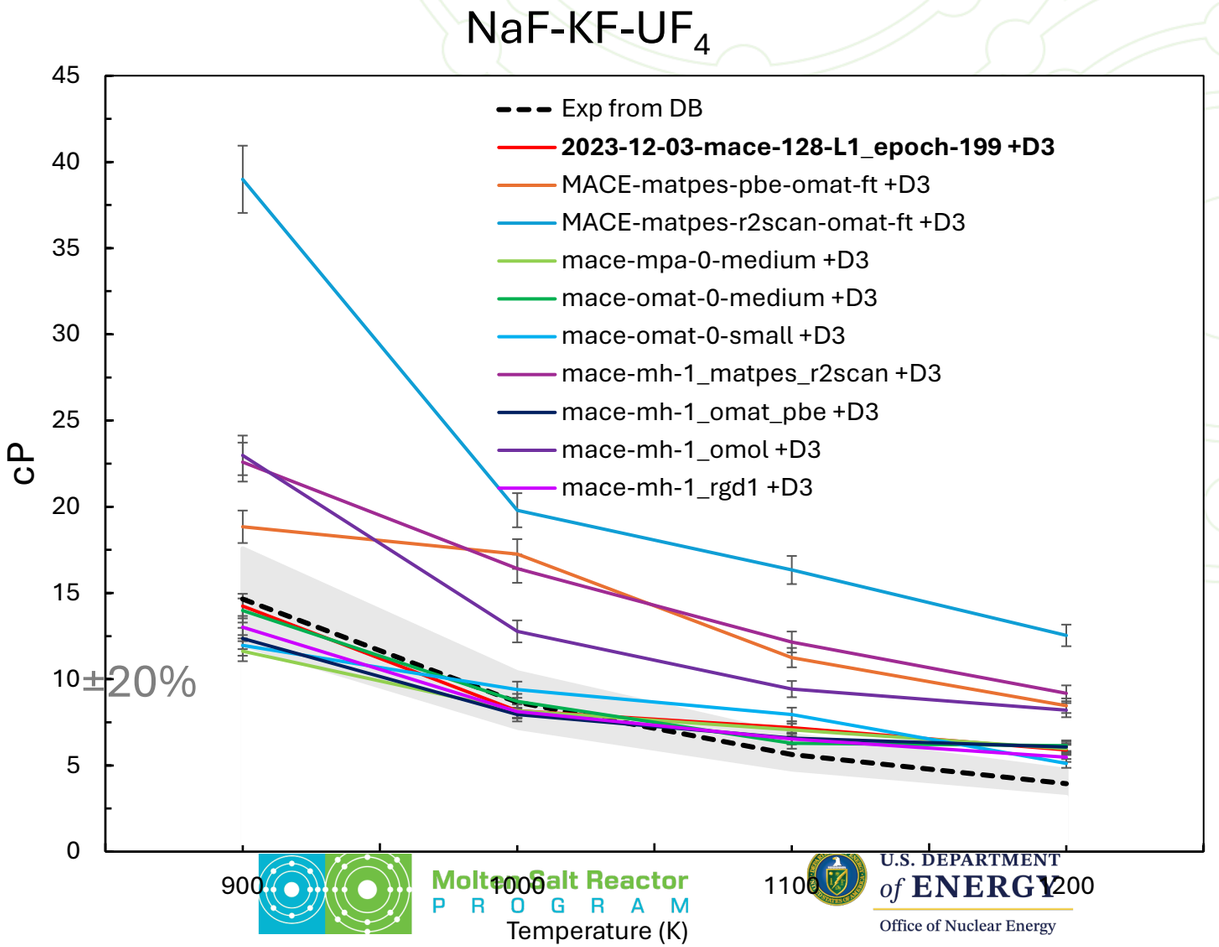
Most MACE-MLIPs also show predicted viscosity in good agreement with experimental results

- The 2023 version of MACE MLIP reproduces both density and viscosity for the two benchmark systems





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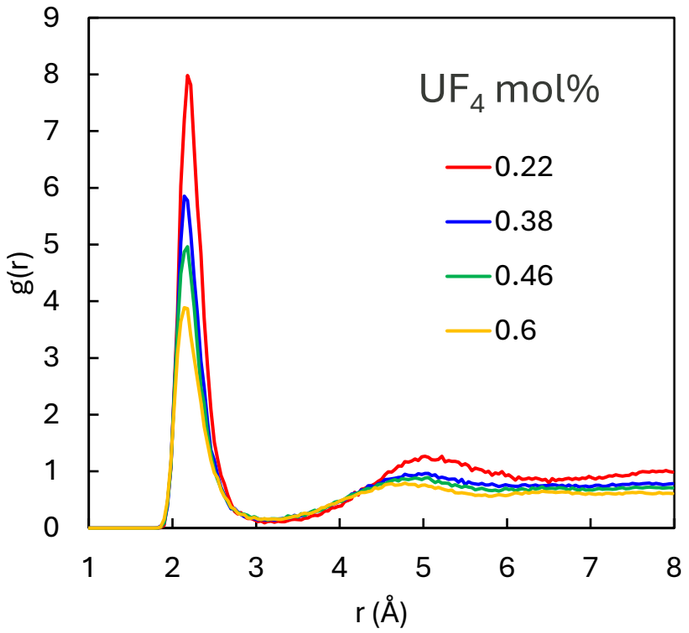
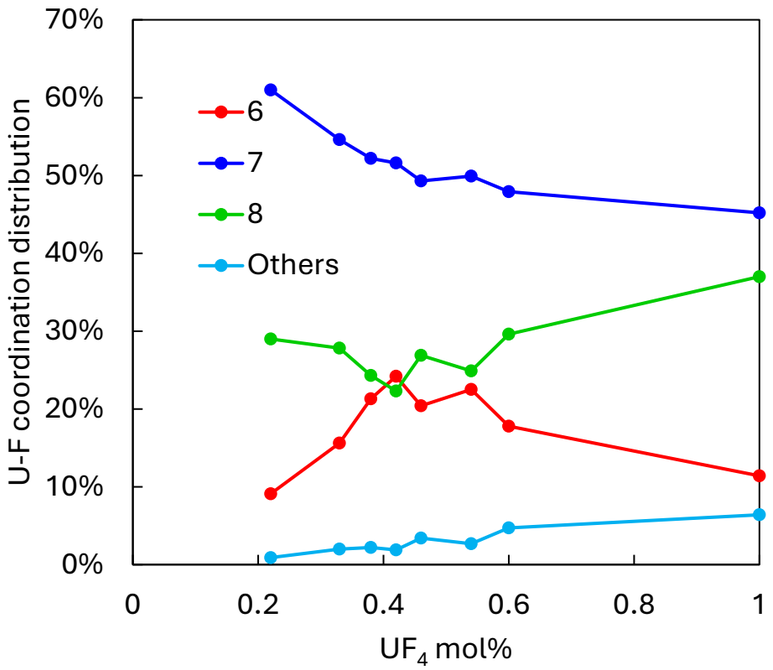
Dynamic $[UF]_n$-complex with different compositions and temperatures

- U-F coordination number distribution in different compositions:

U-F pair distribution function across compositions

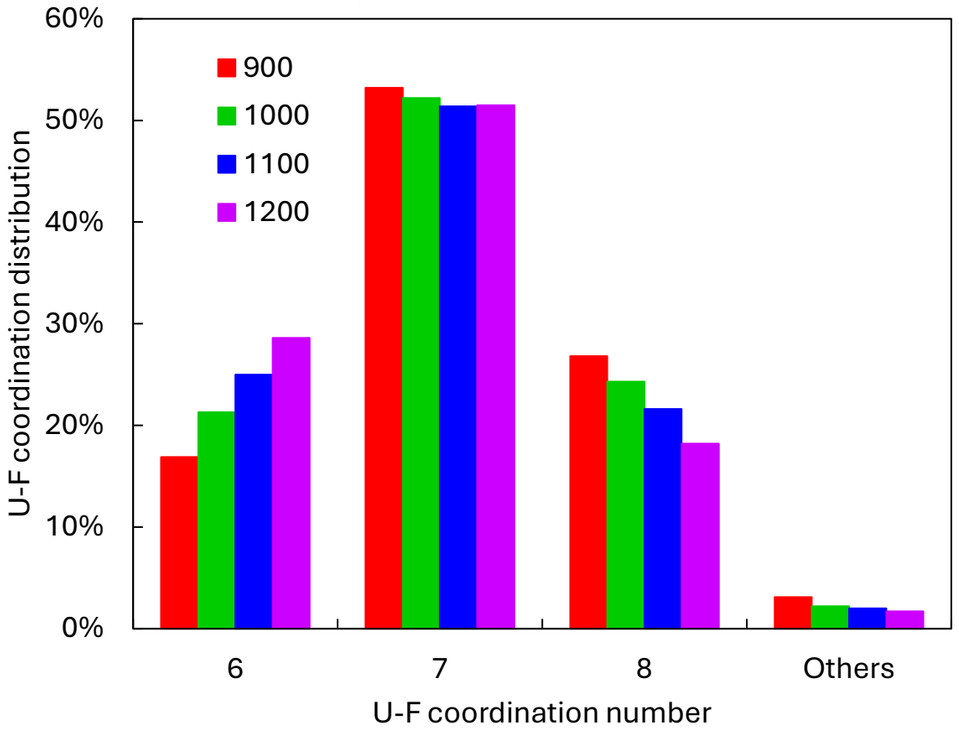
UF_4+NaF at 1200K





U-F coordination number distribution in different temperatures

UF_4+NaF (0.38-0.62)



Similar results in other compositions or temperatures.

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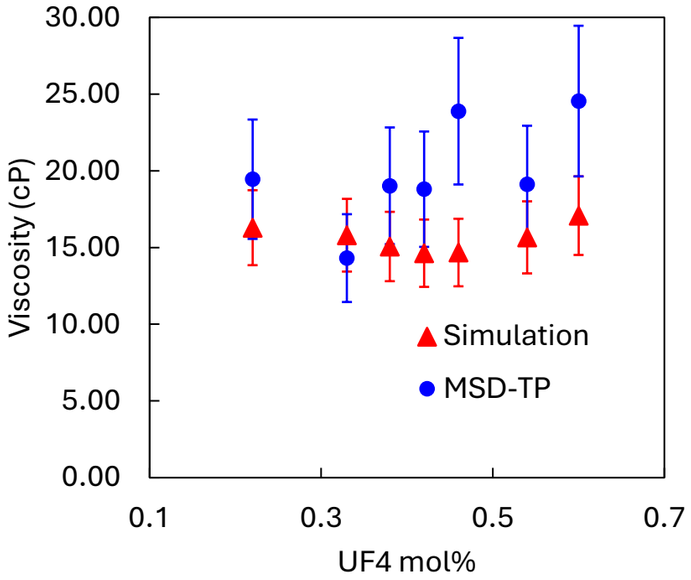
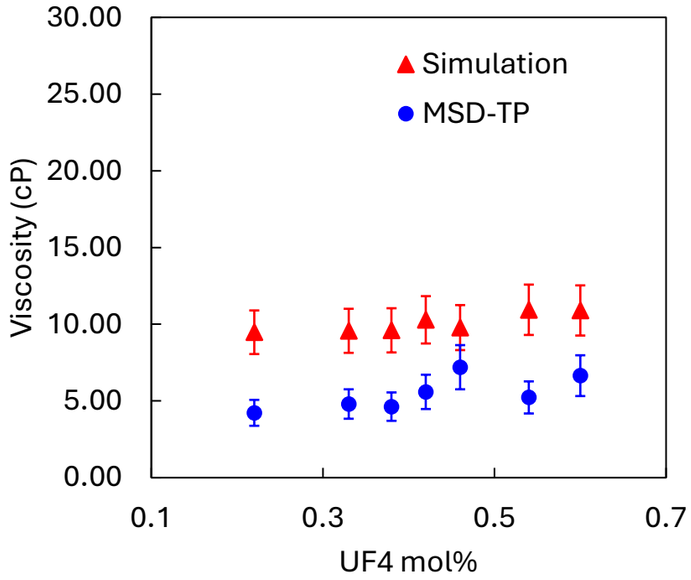
Computed viscosity captures quantitative trends across compositions, with lower accuracy at higher temperatures

- Simulated viscosity compared to experiments at different compositions:

UF_4+NaF at 900K

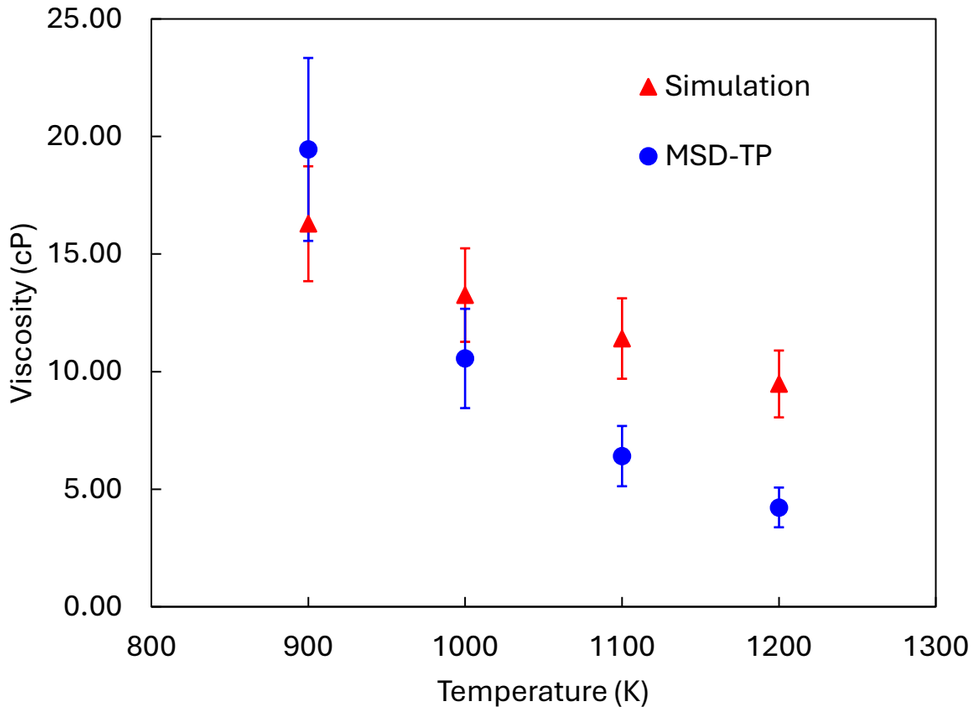


UF_4+NaF at 1200K



Simulated viscosity at different temperatures:

UF_4+NaF (0.22-0.78)



Viscosity is computed based on Stokes–Einstein correlation using diffusion coefficients and estimated hydrodynamic radius.[1]

[1] Nguyen et al., Journal of Molecular Liquids, 326, 115262

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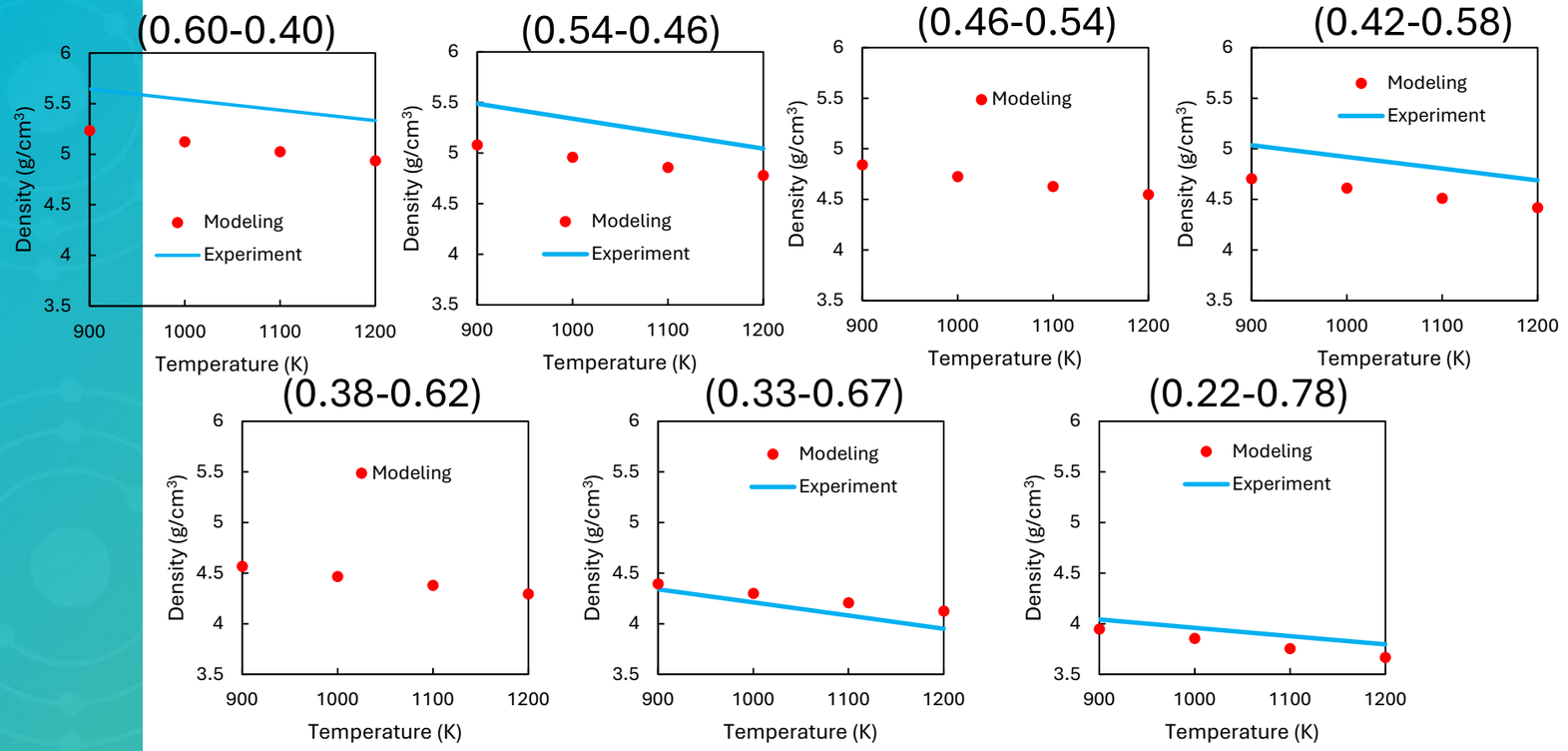
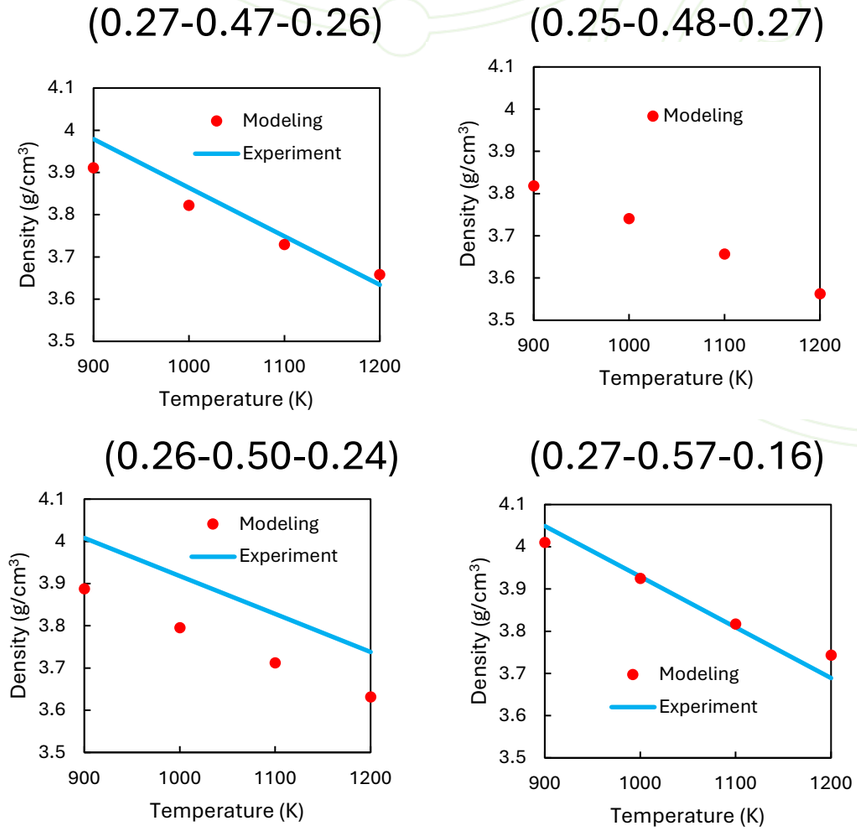
The MACE MLIP (2023) shows robustness across various compositions and temperatures for UF_4-containing systems

- CMD simulations using the MACE-2023 MLIP under 900-1200K for 2 ns (NPT), to determine density of molten salts
- Provide reliable estimates (with errors) when experimental data are not available

UF_4+NaF (mol%)

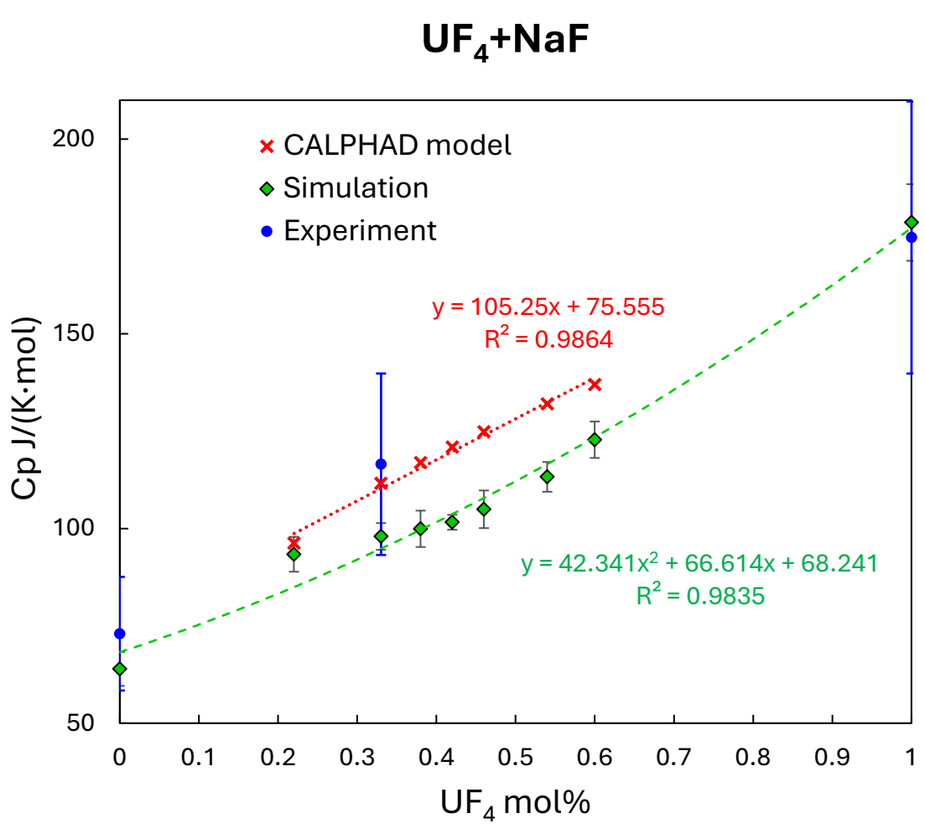


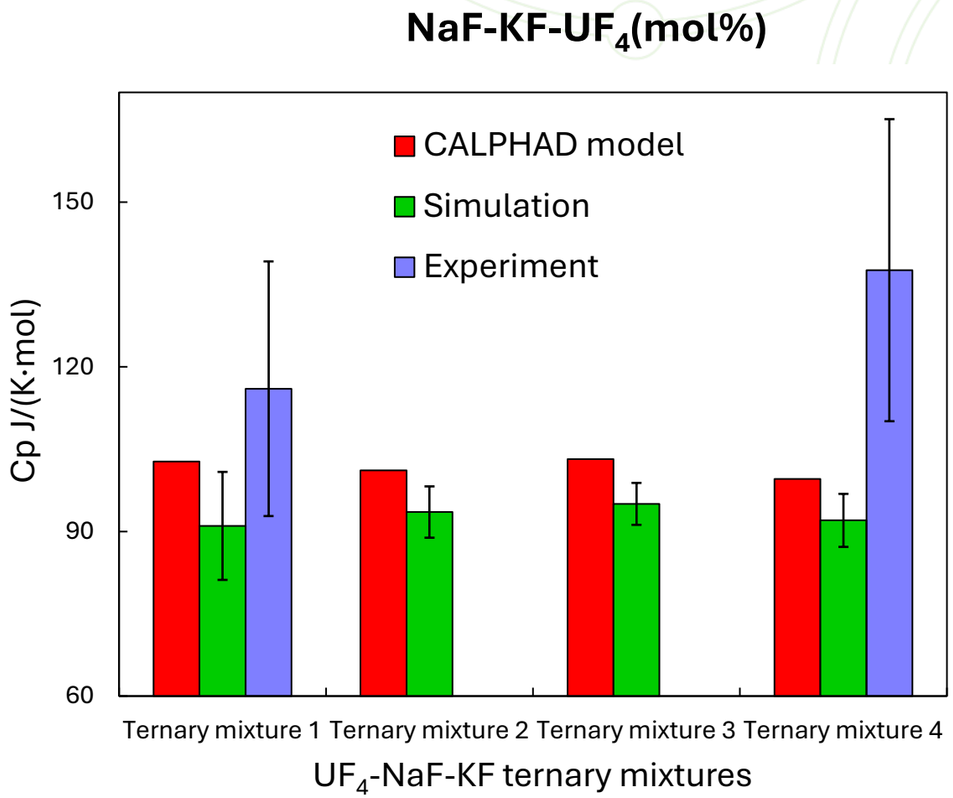
NaF-KF-UF_4(mol%)



The MLIP also provides a quick estimation of heat capacity for these UF_4-containing molten salts

- Heat capacity (Cp) is extracted from the CMD simulations using the MACE-2023 MLIP
- When both simulated Cp and CALPHAD results deviate from experimental data, a new validation experiment is suggested





Summary and Outlook

- LLM can accelerate data-mining and reveal the data gaps, but it can also extract incorrect results from existing data or generate fake results. A post-validation is still needed
- AIMD simulation is limited by its modeling size and time scale due to high computational cost. Its relative high accuracy can generate high-fidelity structural information of molten salt, which can be passed to other analysis or prediction methodologies
- Foundational MLIP provides a base for developing MLIPs with a higher accuracy for specific systems (e.g., UF_4-containing molten salts), but this is still underdeveloped
- **Milestone/Deliverable: Thermal properties of 3-5 different uranium-fluoride/chloride salt compositions. Joint report with experimental team (on track for Sep 30 2026)**
- **Ongoing work**
- Fine-tuning MACE foundational MLIP using our DFT/AIMD data to improve the MLIP accuracy for UF_4-containing binary and ternary molten salt systems.
- Report and manuscript in preparation

Acknowledgement

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