Molten Salt Reactor PROGRAM

Machine Learning Prediction and Molecular Dynamics Modeling of Thermophysical Properties of Molten Salts

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Overview

Milestone Number	Milestone title	Due Date	Status
M3AT-25PN0705061	Report on Thermophysical properties of binary molten salt mixtures: machine learning prediction	12/15/2024	Complete
M3AT-25PN0705062	Complete computational investigation of thermophysical (density, specific heat, and viscosity) and structural properties of NaCl-$PuCl_3$-$AmCl_3$ molten salts.	09/30/2025	On schedule

- Milestone M3AT-25PN0705061
- Milestone M3AT-25PN0705062
- Publications & presentations

Milestone #1 - M3AT-25PN070506

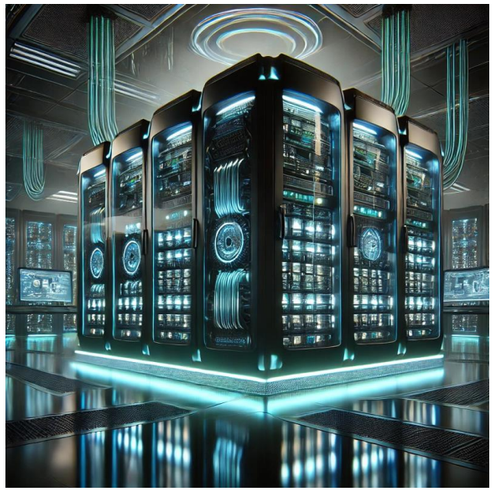
Description: We use machine learning and literature data to

- (i) investigate the correlation between thermophysical properties of binary molten salt systems and physical and chemical properties of elements involved
- (ii) train models for prediction of the thermophysical properties

Milestone: M3, completed (Dec 15, 2024)

Our main motivation is a rapid approach for determining molten salt properties

- Imagine that we can obtain thermophysical properties of a molten salt mixture in seconds, without…



- Data science (AI/ML) enables us to achieve this goal
- Our first attempt is to build machine learning models for prediction of thermophysical properties of *binary halide systems*.

The following machine learning workflow was adopted

Datasets
- Literature data
- Composition
- Temperature

Features
- Atomic properties
- Correlation of features and properties

Regression
- Linear model
- Ensemble method
- Probabilistic method
- Deep learning
- Hyper-parameter tuning

Training/testing
- Error metrics

We used literature data for datasets

- Extracted info:
  1. *Name of mixture*
  2. *Composition*
  3. *Temperature*
  4. *Value of property*
- Mostly from database by Janz *et al*. Data in the MSR campaign also used.
- Density, viscosity, electrical conductivity, and surface tension
- Organized in Excel
- Thousands of data points: 80% training set, 20% test set

Mixture	Composition (mol% of salt #2)	Temperature (K)	Density (g/cc)
AlF3-LiF	90	1130	1.972

RESEARCH ARTICLE | JANUARY 01 1974

Molten Salts: Volume 4, Part 1, Fluorides and Mixtures Electrical Conductance, Density, Viscosity, and Surface Tension Data

G. J. Janz; G. L. Gardner; Ursula Krebs; R. P. T. Tomkins

Check for updates

J. Phys. Chem. Ref. Data 3, 1–115 (1974)
https://doi.org/10.1063/1.3253134

TABLE 23. AlF_3-LiF: Density

Numerical values (gcm^{-3})

Mol percent LiF

T	100	95	90	85	80	75	70	65	60	55
1130	1.800	1.885	1.972	2.037	2.073	2.098	2.088	2.041	1.962	1.861
1140	1.795	1.880	1.966	2.030	2.065	2.089	2.079	2.031	1.952	1.851
1150	1.791	1.875	1.960	2.023	2.058	2.081	2.069	2.021	1.942	1.842
1160	1.786	1.870	1.953	2.016	2.051	2.073	2.060	2.010	1.931	1.832
1170	1.781	1.866	1.947	2.010	2.044	2.064	2.051	2.000	1.921	1.823
1180	1.777	1.861	1.941	2.003	2.037	2.056	2.041	1.990	1.911	1.814
1190	1.772	1.856	1.935	1.996	2.030	2.048	2.032	1.979	1.901	1.805
1200	1.767	1.851	1.929	1.990	2.023	2.039	2.022	1.969	1.891	1.795
1210	1.763	1.846	1.923	1.983	2.016	2.031	2.013	1.959	1.880	1.786
1220	1.758	1.842	1.917	1.976	2.009	2.022	2.004	1.948	1.870	1.776
1230	1.753	1.837	1.910	1.970	2.002	2.014	1.994	1.938	1.860	1.767
1240	1.748	1.832	1.904	1.963	1.994	2.006	1.985	1.928	1.850	1.758
1250	1.744	1.827	1.898	1.956	1.987	1.997	1.976	1.918	1.840	1.748
1260	1.739	1.822	1.892	1.950	1.980	1.989	1.966	1.907	1.829	1.739
1270	1.734	1.817	1.886	1.943	1.973	1.981	1.957	1.897	1.819	1.729
1280	1.730	1.813	1.880	1.936	1.966	1.972	1.948	1.887	1.809	1.720
1290	1.725	1.808	1.874	1.929	1.959	1.964	1.938	1.876	1.799	1.711
1300	1.720	1.803	1.867	1.923	1.952	1.956	1.929	1.866	1.789	1.701
1310	1.716	1.798	1.861	1.916	1.945	1.947	1.920	1.856	1.778	1.692
1320	1.711	1.793	1.855	1.909	1.938	1.939	1.910	1.845	1.768	1.682

Molten Salt Reactor PROGRAM

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We built features using atomic properties

Mixture & composition

$x\ AC_n - y\ BD_m$

A, B = metals
C, D = F, Cl, Br, and I
n, m = 1,2,3,…
x,y >0 and x+y=1

- Atomic properties of elements (Number of electron, radius, ionization energy, ect.)
- Average schemes
- Temperature
- Totally, 393 features

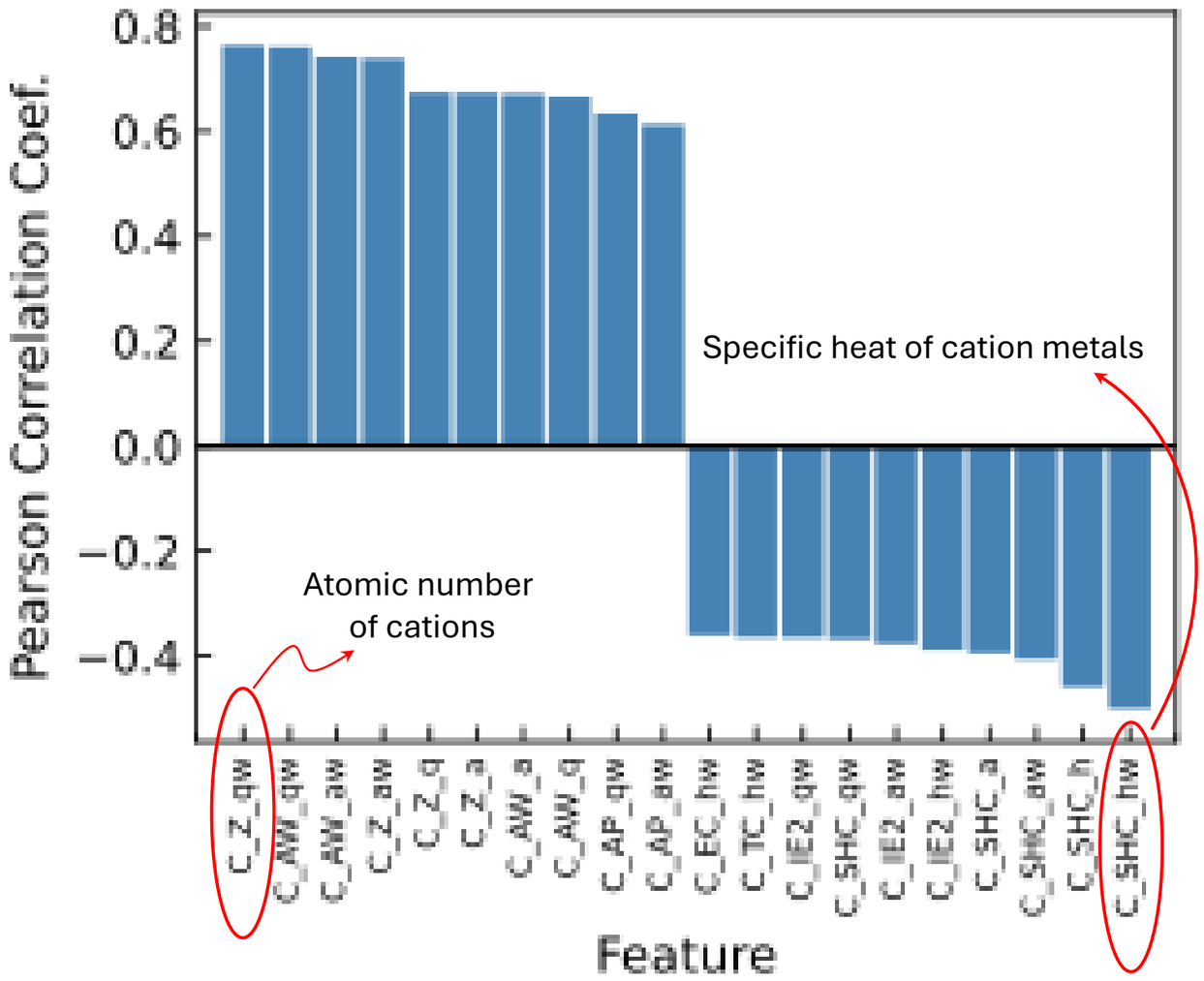
Statistical moments and average methods for elemental feature F in the binary system $(AX_n)_x$-$(BY_m)_y$, A and B are cations, and X and Y are anions in the two salts.

Method	Composition-agnostic and composition-based notations	Expression for composition-based features for cation and anions. Likewise, composition-agnostic features with x=y=1
Arithmetric average	$\langle F\rangle_{aw}$ and $\langle F\rangle_{a}$	$\langle F\rangle_{aw}(\text{cation}) = \frac{xF_A + yF_B}{x+y}$ $\langle F\rangle_{aw}(\text{anion}) = \frac{nxF_X + myF_Y}{nx+my}$
Standard deviation	$\langle F\rangle_{sw}$ and $\langle F\rangle_{s}$	$\langle F\rangle_{sw}(\text{cation}) = \frac{\lvert xF_A - yF_B\rvert}{x+y}$ $\langle F\rangle_{sw}(\text{anion}) = \frac{\lvert nxF_X - myF_Y\rvert}{nx+my}$
Harmonic average	$\langle F\rangle_{hw}$ and $\langle F\rangle_{h}$	$\langle F\rangle_{sw}(\text{cation}) = \frac{x+y}{\frac{x}{F_A}+\frac{y}{F_B}}$ $\langle F\rangle_{sw}(\text{anion}) = \frac{nx+my}{\frac{nx}{F_x}+\frac{my}{F_y}}$
Quadratic average	$\langle F\rangle_{qw}$ and $\langle F\rangle_{q}$	$\langle F\rangle_{sw}(\text{cation}) = \sqrt{\frac{xF_A^2 + yF_B^2}{x+y}}$ $\langle F\rangle_{sw}(\text{anion}) = \sqrt{\frac{nxF_X^2 + myF_Y^2}{nx+my}}$

Gharakhanyan et al. Journal of Chemical Physics 2024, 160 (20)

We found unknown correlation between properties of a molten salt mixture and its constituents

Density - Feature correlation



Pearson correlation coefficient

$$\frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2 (y_i - \bar{y})^2}}$$

- Density of molten salts is most anti-correlated with the heat capacity of metals.

We tested different regression methods

- Methods:
  - ✓ Ridge regression
  - ✓ Lasso regression

  } *Linear*

  - ✓ Gradient boosting regression
  - ✓ Random Forest

  } *Ensemble*

  - ✓ Gaussian process regression

  } *Bayesian*

  - ✓ Artificial Neural Network

  } *Deep learning*

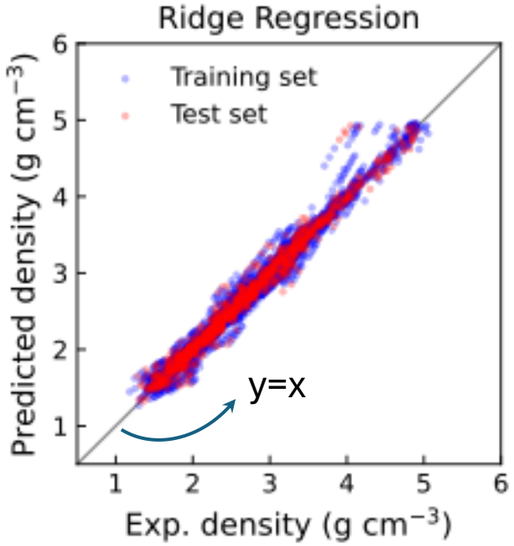
- Hyperparameter tuning:
  - ✓ Parameter grid searches

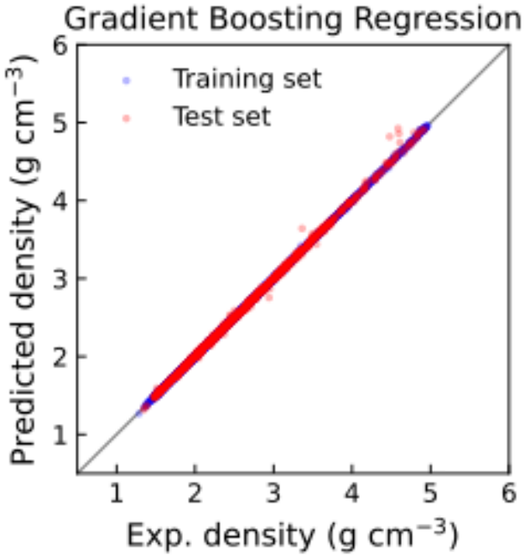
https://scikit-learn.org

TensorFlow

https://www.tensorflow.org

Trained models can accurately predict the density



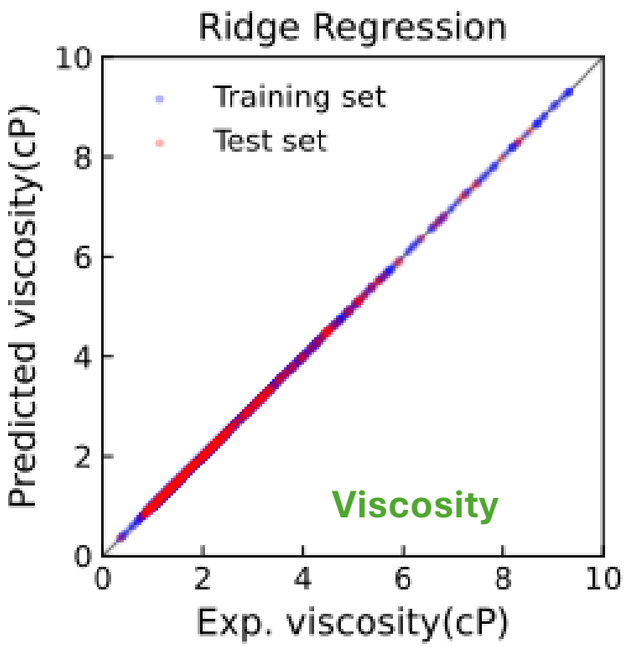


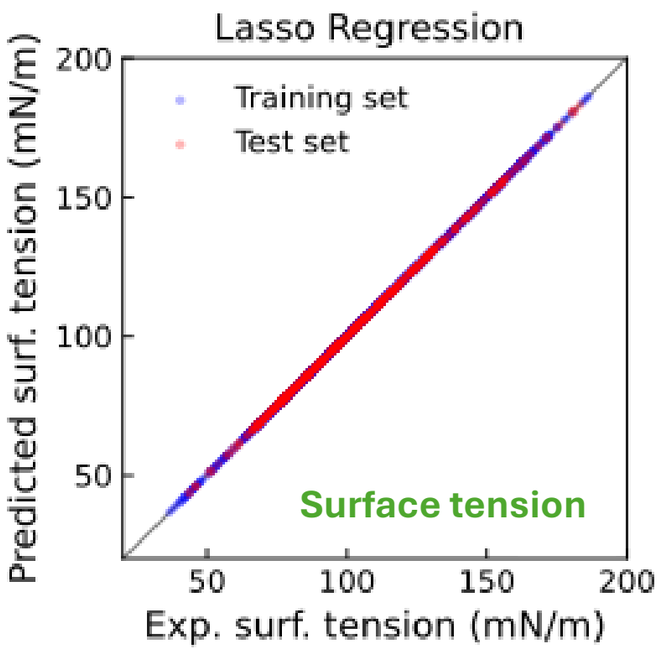
Error metrics

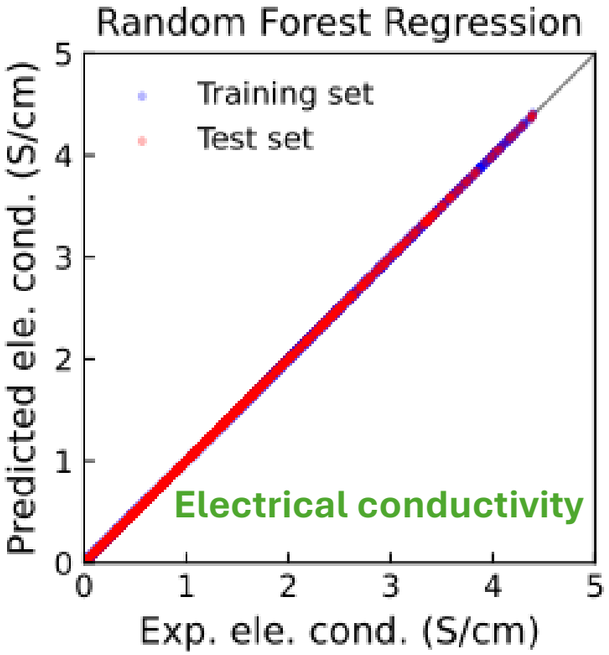
- Root Mean Squared Error (RMSE)
- Mean absolute error (MAE)
- Coefficient of determination (R^2)

Error metrics for regression models of the liquid density						
Regression method	Training set			Test set		
	RMSE, gcm^{-3}	MAE, gcm^{-3}	R^2	RMSE, gcm^{-3}	MAE, gcm^{-3}	R^2
Ridge	0.106	0.071	0.980	0.115	0.074	0.979
Lasso	0.116	0.080	0.977	0.121	0.081	0.978
Gradient Boosting	0.008	0.005	1.000	0.027	0.015	0.999
Gaussian Process	0.050	0.037	0.996	0.216	0.058	0.925
Random Forest	0.016	0.010	1.000	0.041	0.025	0.997
Artificial Neural Network	0.040	0.030	0.997	0.044	0.033	0.997

Likewise, viscosity, surface tension, and electrical conductivity were investigated







- Very promising predictive models were trained.

Milestone #2 - M3AT-25PN0705062

Description:

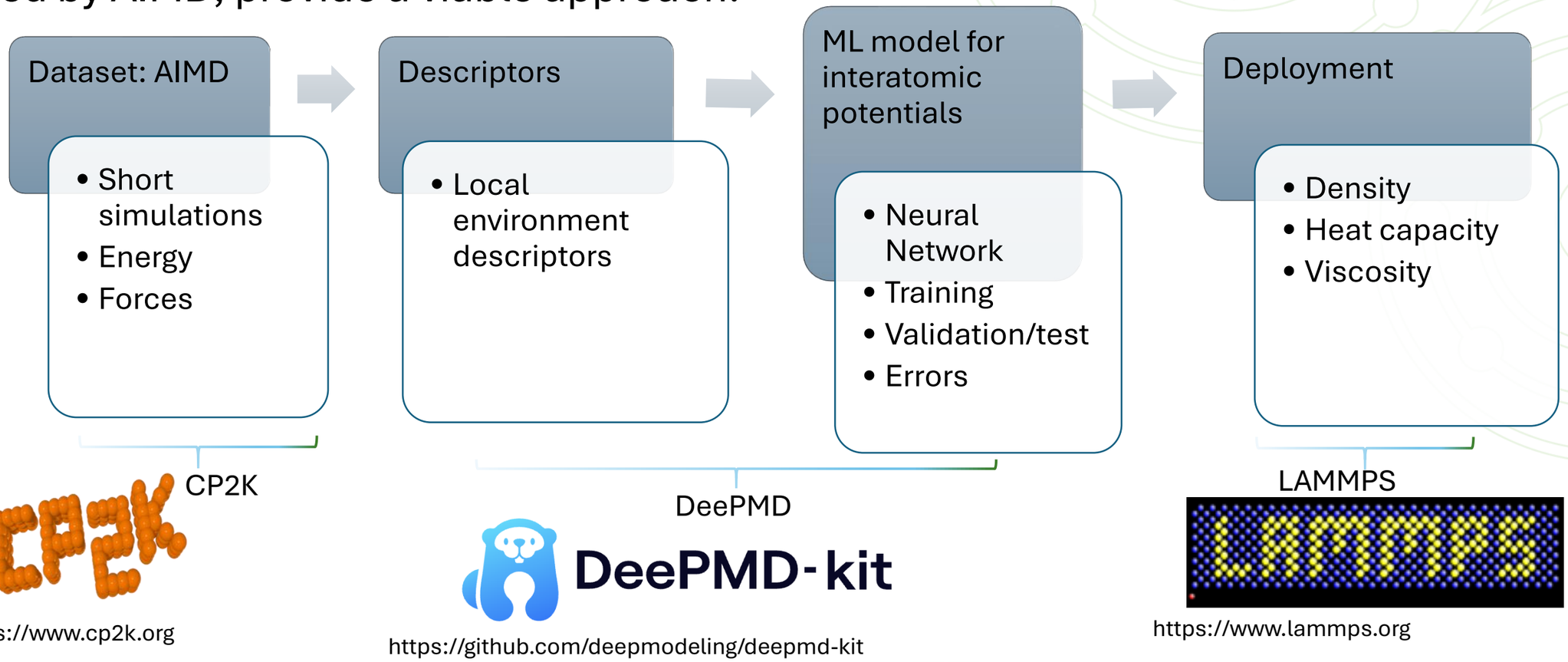
- Determine the thermophysical and structural properties of NaCl-$PuCl_3$-$AmCl_3$ molten salt systems using atomistic modeling.

Milestone:

- M3 (Sep 30, 2025)

We used machine learning interatomic potentials, trained on ab initio molecular dynamics data, to accelerate simulations

- Ab initio molecular dynamics (AIMD): accurate but computationally demanding
- Machine learning interatomic potentials, trained on potential energy surfaces calculated by AIMD, provide a viable approach.



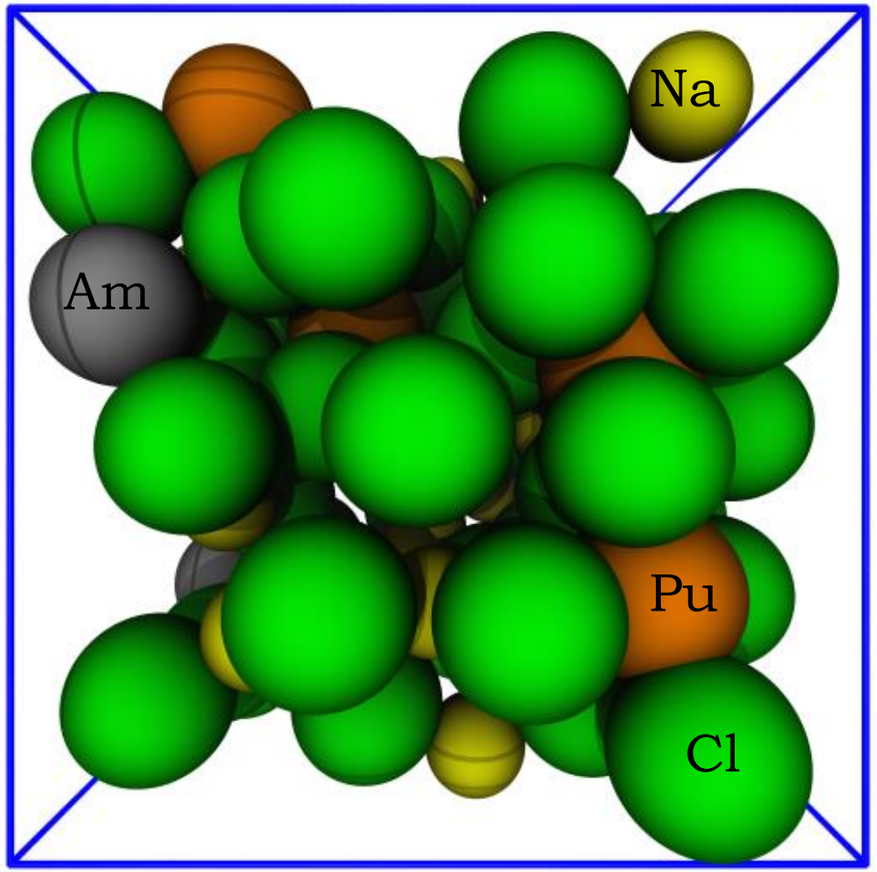
Datasets were generated using AIMD

Computational recipe:

- revPBE-vdW density functional
- DZVP basis sets
- GTH pseudopotentials
- NPT ensemble, dt=2.5 fs
- Classical molecular dynamics – equilibrated systems as starting points
- T= 900, 1100, and 1100 K
- CP2K code

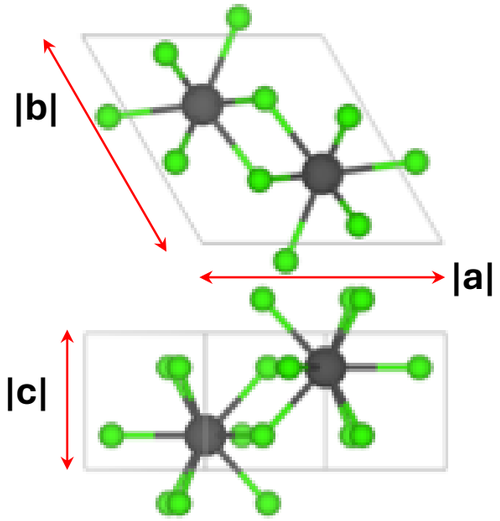
$NaCl$-$PuCl_3$-$AmCl_3$ system:

- Composition: $NaCl:PuCl_3:AmCl_3=0.55:0.31:0.14$
- Totally, 84 atoms / box

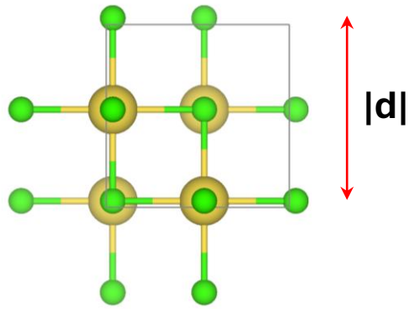


Computational method accurately predicts lattice constants of solids

$AmCl_3$ & $PuCl_3$



NaCl

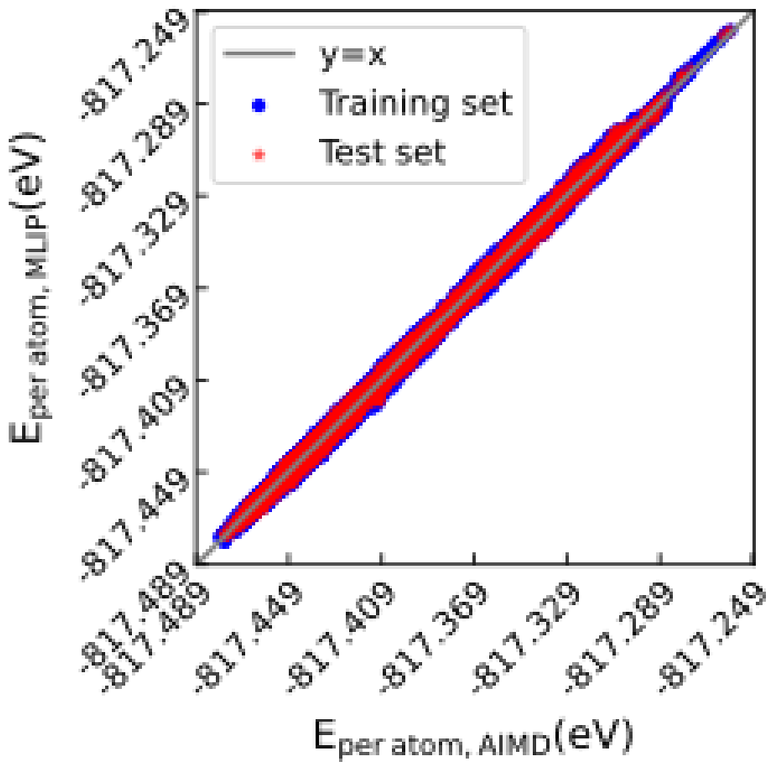


Asprey et al., Inorg. Chem. 1965, 4, 7, 985–986
Burns et al., J. Inorg. Nucl. Chem. 1975, 37, 743-749

- $AmCl_3$, $PuCl_3$: hexagonal P6_3/m space group
- NaCl: cubic Fm-3m space group
- Theory in agreement with experiment

	$AmCl_3$			$PuCl_3$			NaCl		
	Theory	Exp.	Error(%)	Theory	Exp.	Error(%)	Theory	Exp.	Error(%)
|a|=|b| (Å)	7.337	7.390	0.7	7.330	7.394	0.8			
|c| (Å)	4.234	4.234	~0.0	4.296	4.243	1.3			
|d| (Å)							5.665	5.640	0.4

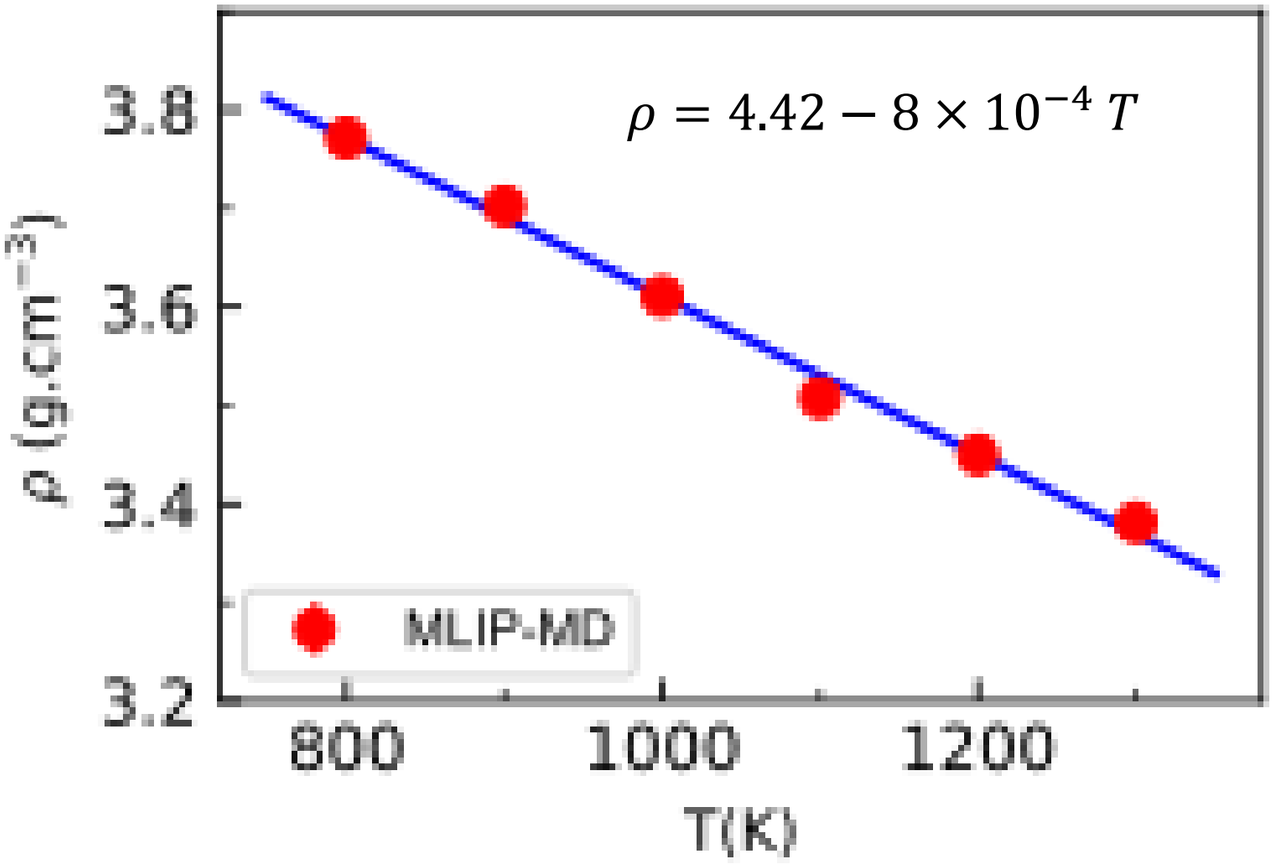
Machine learning interatomic potential (MLIP) is accurate



- DeePMD method
- $4x10^4$ frames for training set
- $1x10^4$ frames for test set
- Loss function for energy and force
- 6.5 Å cutoff
- {250,250,250} fitting network
- Small energy and force errors

	Training set	Test set
Energy MAE/atom (meV)	1.59	1.60
Energy RMSE/atom (meV)	1.99	2.01
Force MAE (meV/Å)	60	65
Force RMSE (meV/Å)	81	84

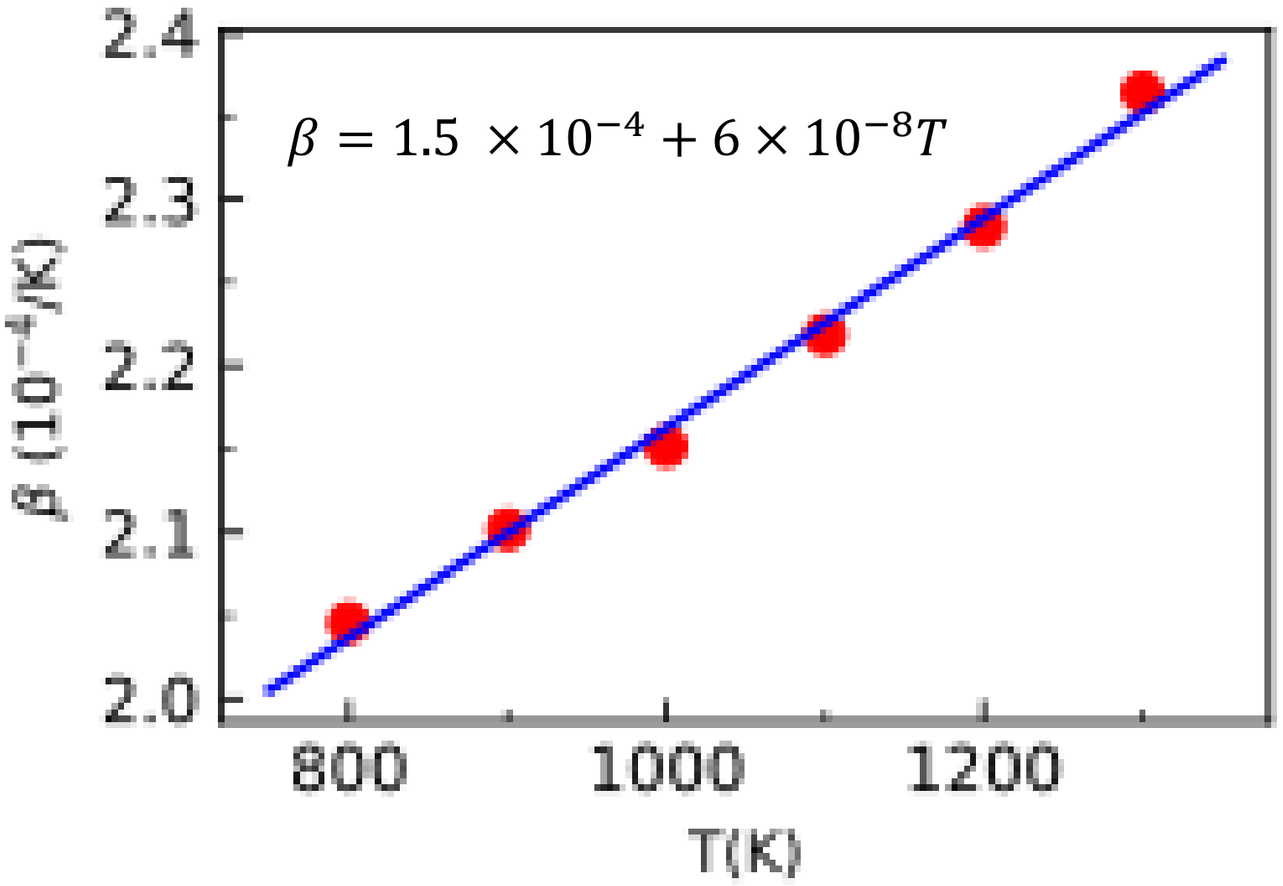
Liquid density was calculated over a range of temperatures



$$\rho = \frac{\sum_i N_i M_i}{V N_A} = \frac{m}{V}$$

- MLIP molecular dynamics
- NPT ensemble, dt=1fs
- T= 800-1300 K
- LAMMPS code
- $\rho = \rho_0 - A \times T$

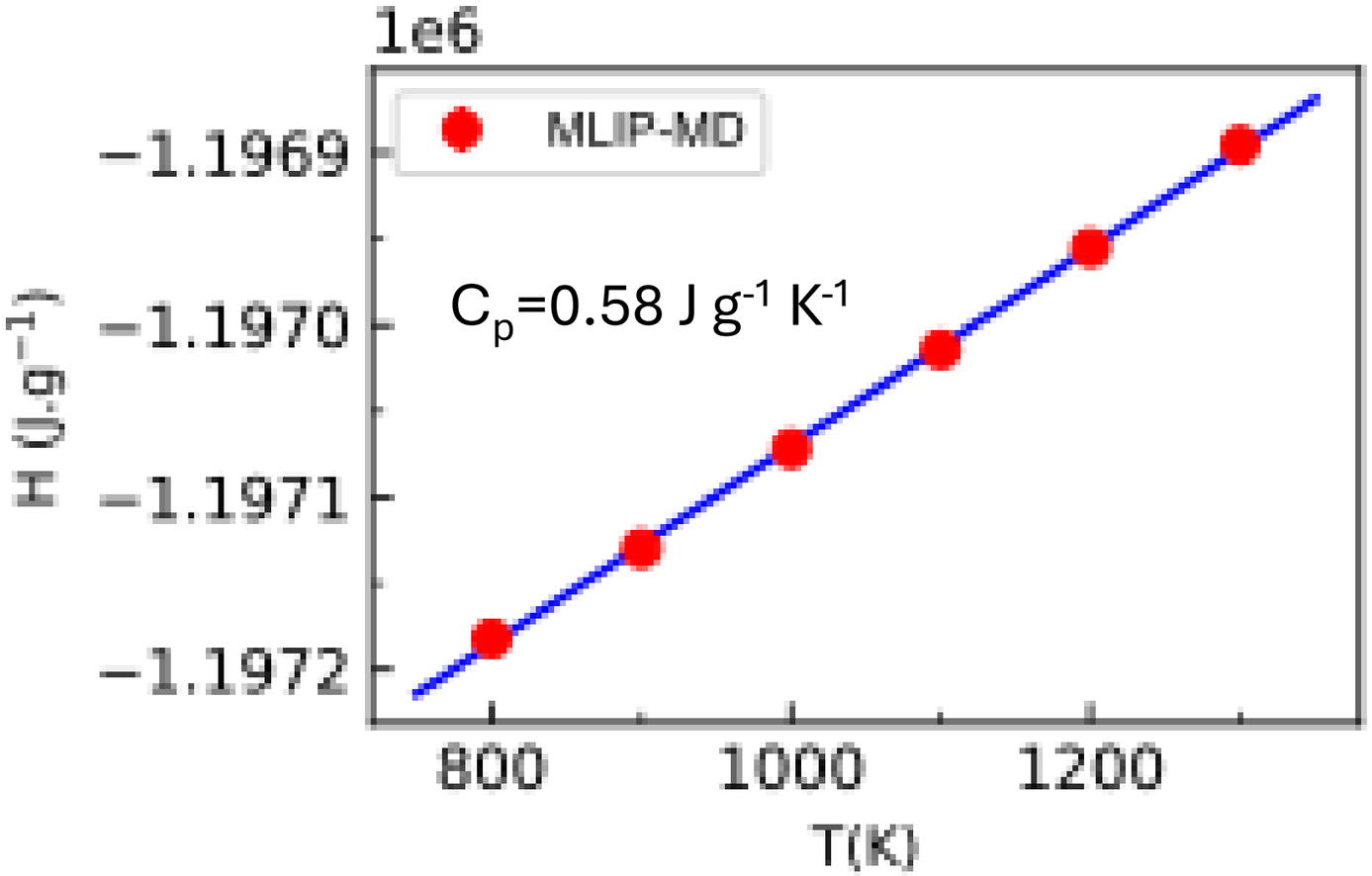
Thermal expansion coefficient was calculated through the density



$$\beta = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_p$$

- Very small
- Increased with temperature
- $\beta \approx \beta_0 + A \times T$

Constant pressure heat capacity C_p was predicted through enthalpy

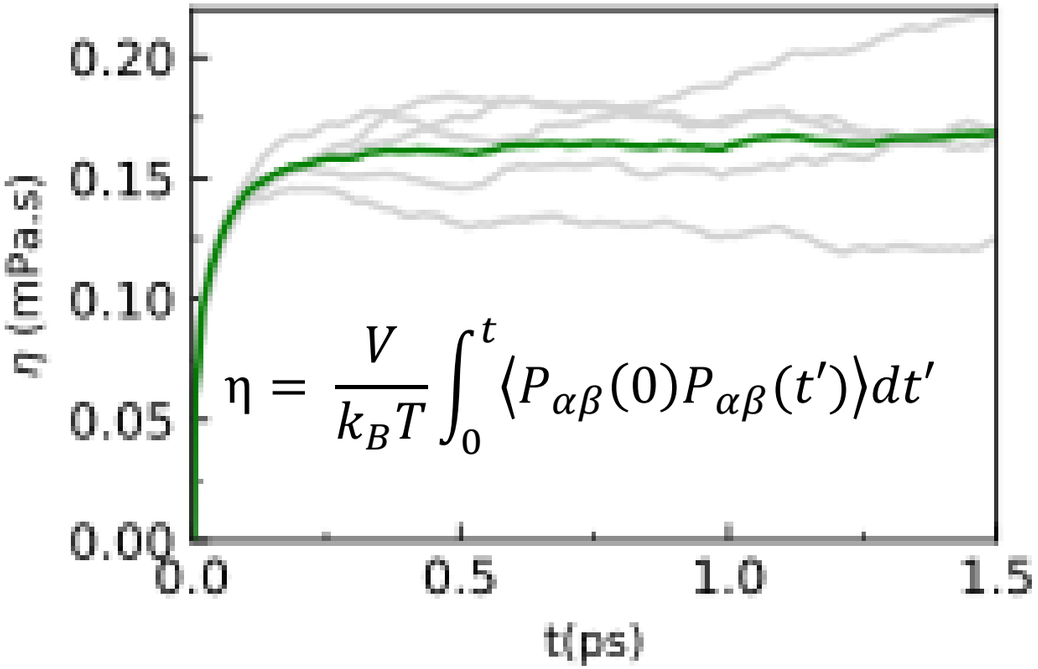


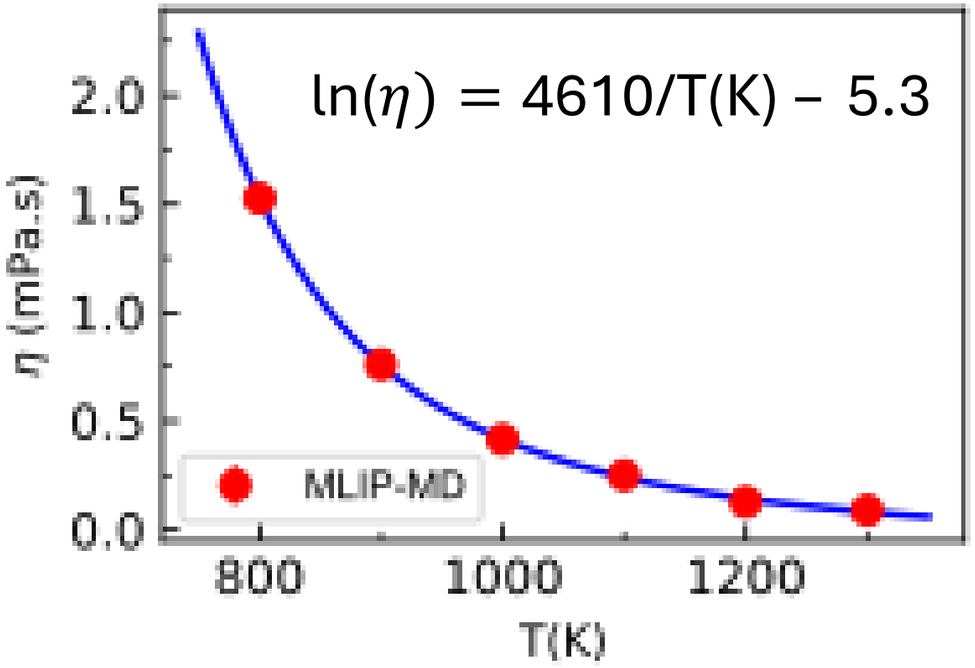
$$C_p = \left(\frac{\partial H}{\partial T}\right)_p$$

H = total energy + V*P

- Increased with temperature
- $H \propto C_p \times T$

Viscosity was calculated using the Green-Kubo formalism





- Multiple simulations for 1 data point
- Decreased with temperature
- Arrhenius relationship

Publications and Presentations

***Peer-reviewed journals*:**

- Manh-Thuong Nguyen, Michael Woods, Juliano Schorne-Pinto, Nick Erfurth, Toni Karlsson, “Thermophysical Properties of NaCl-UCl_3-$PuCl_3$ Molten Salts: A Combined Computational and Experimental Study”, *ACS Applied Energy Materials (accepted)*

Presentations:

- “Structural and Transport Properties of Actinide-Containing Molten Salts: Machine Learning -based Studies”, invited talk at XXth Rencontres du Vietnam Materials Informatics: Accelerating Materials Research and Design with Artificial Intelligence. August 2024. Quynhon, Vietnam & Online, Vietnam.
- “Machine Learning -based Studies of Actinide Molten Salts”, poster at 2024 Molten Salt Reactor Workshop, November 2024. Knoxville, TN 37902, USA.
- “Exploring Liquid Complexity: Electronic Structure Theory, Statistical Mechanics and Data Science –based Modeling of Molten Salts”, seminar at Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA. November 2024.

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