Training Machine Learning Potentials for Nuclear Fuels

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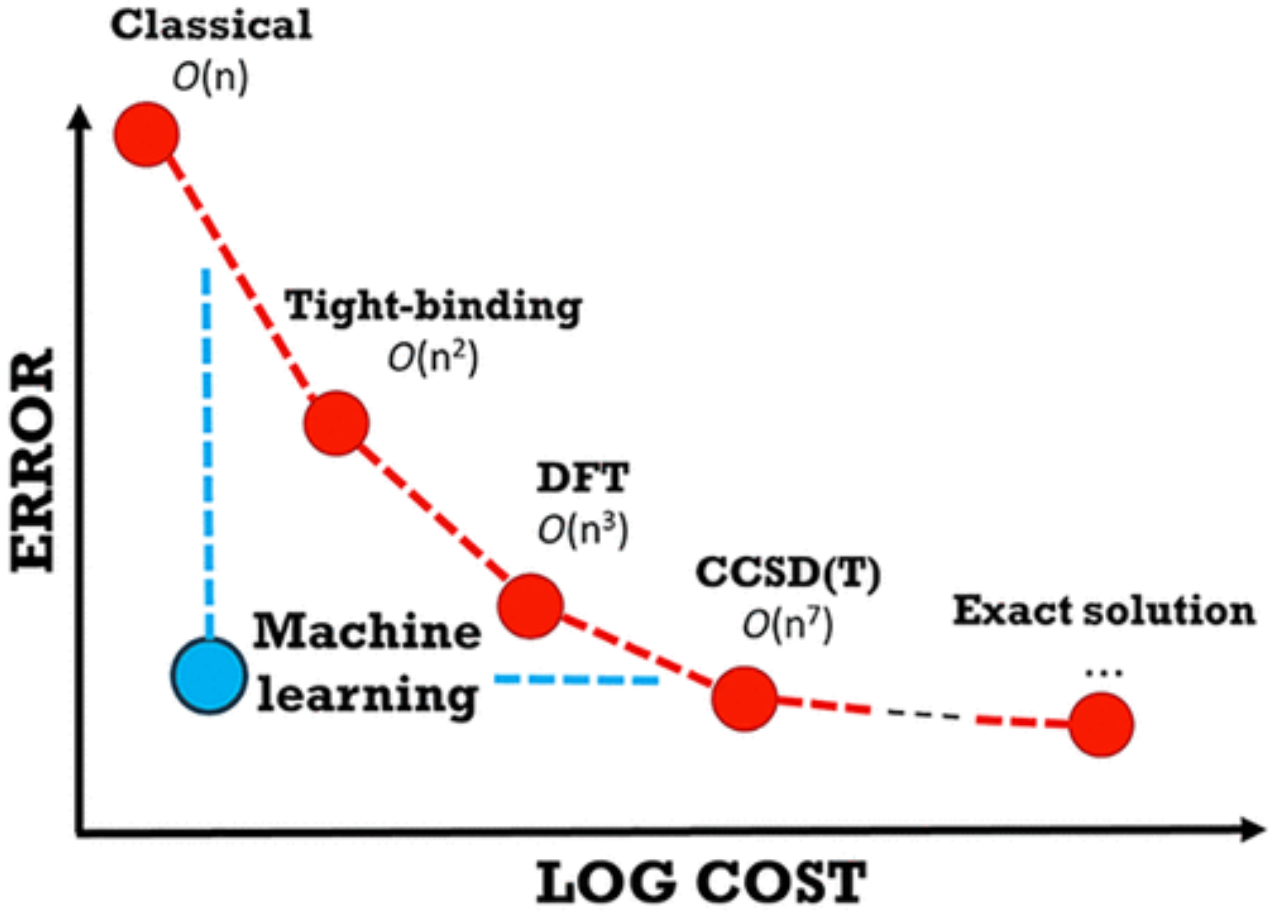
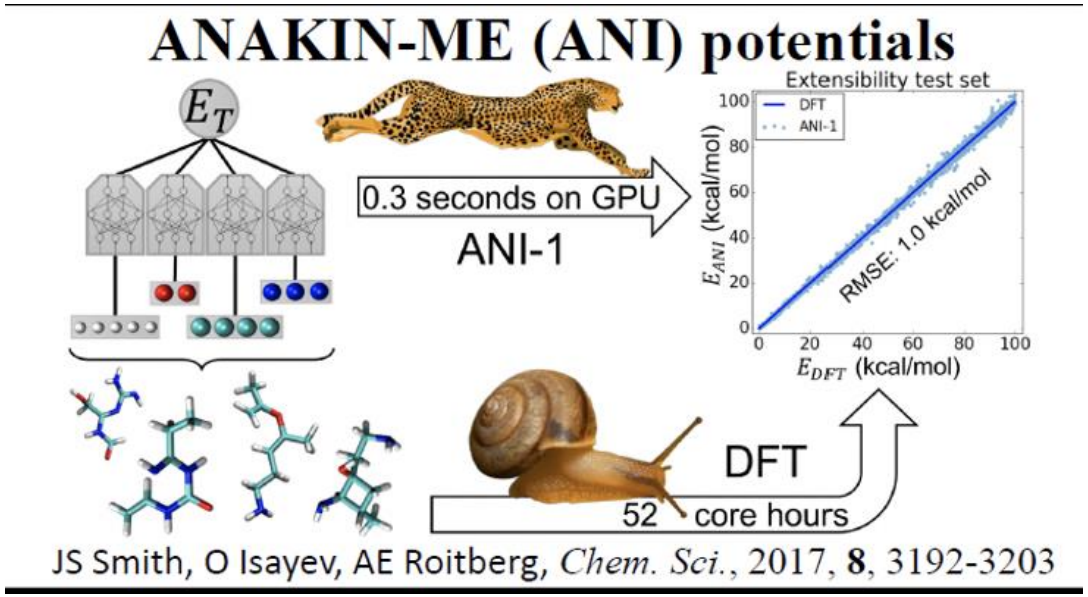
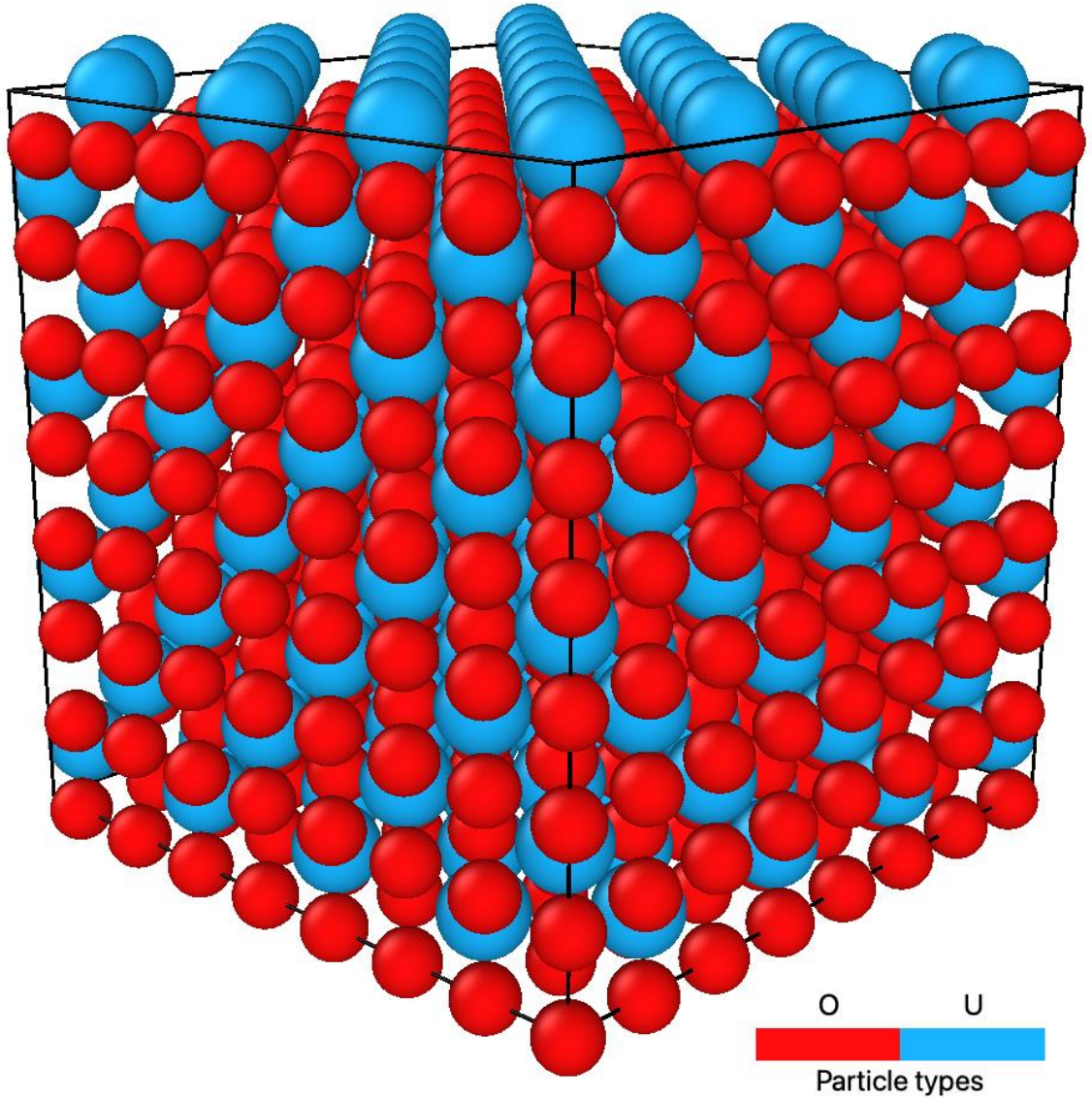
Machine learning (ML) potentials





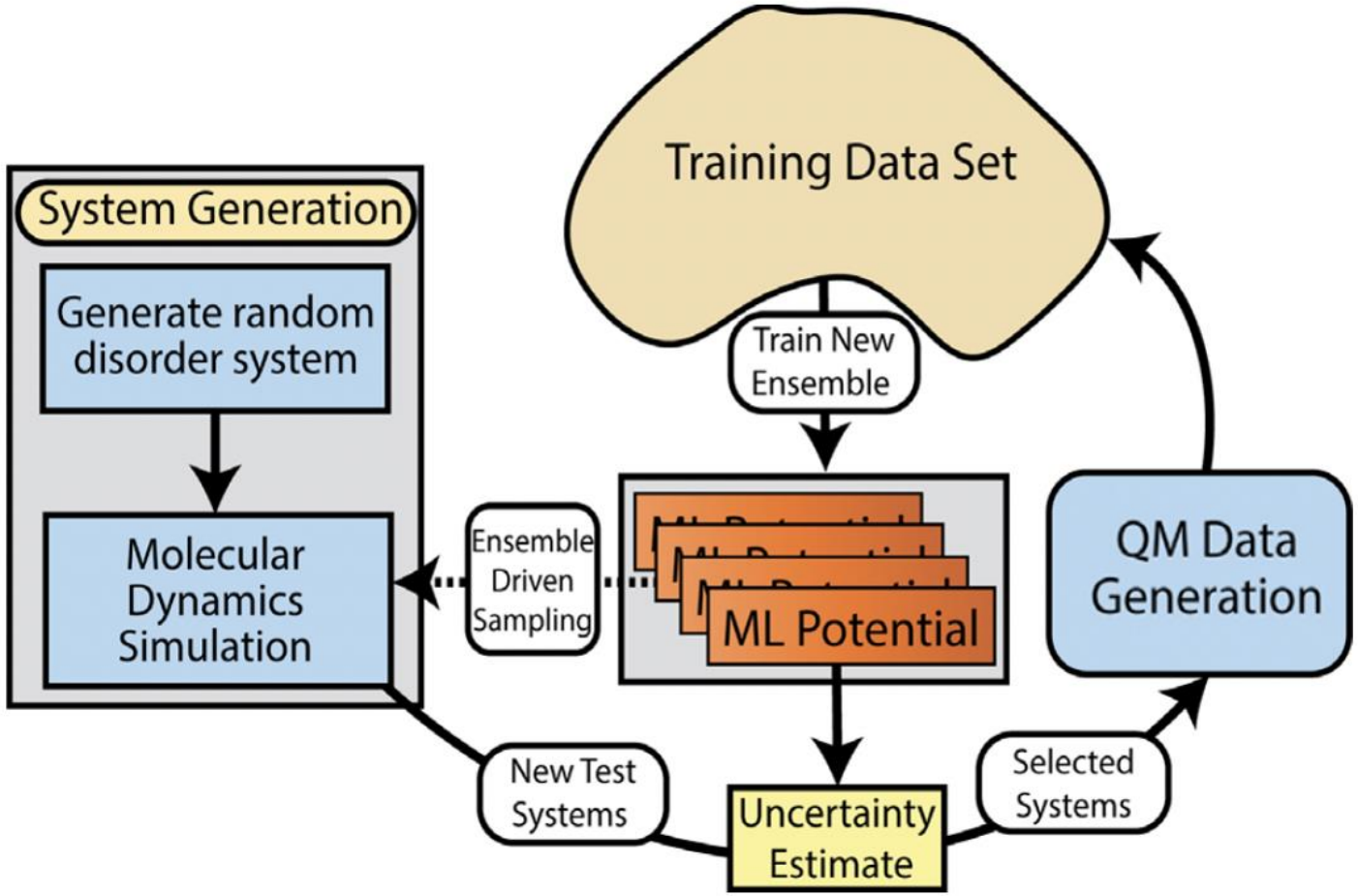
Kulichenko, M. et al. "The Rise of Neural Networks for Materials and Chemical Dynamics." *J. Phys. Chem. Lett.*, 2021,**12**,26,6227-6243



Active Learning Machine Learning

The ANI Potential

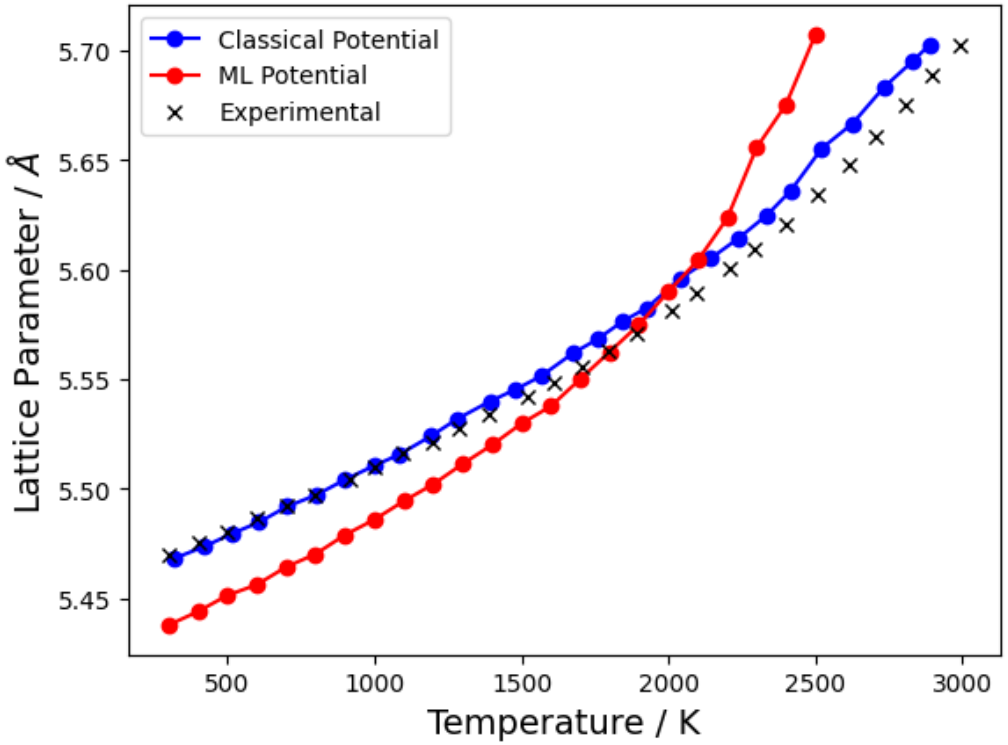
Active learning: An automated iterative approach to develop ML potentials

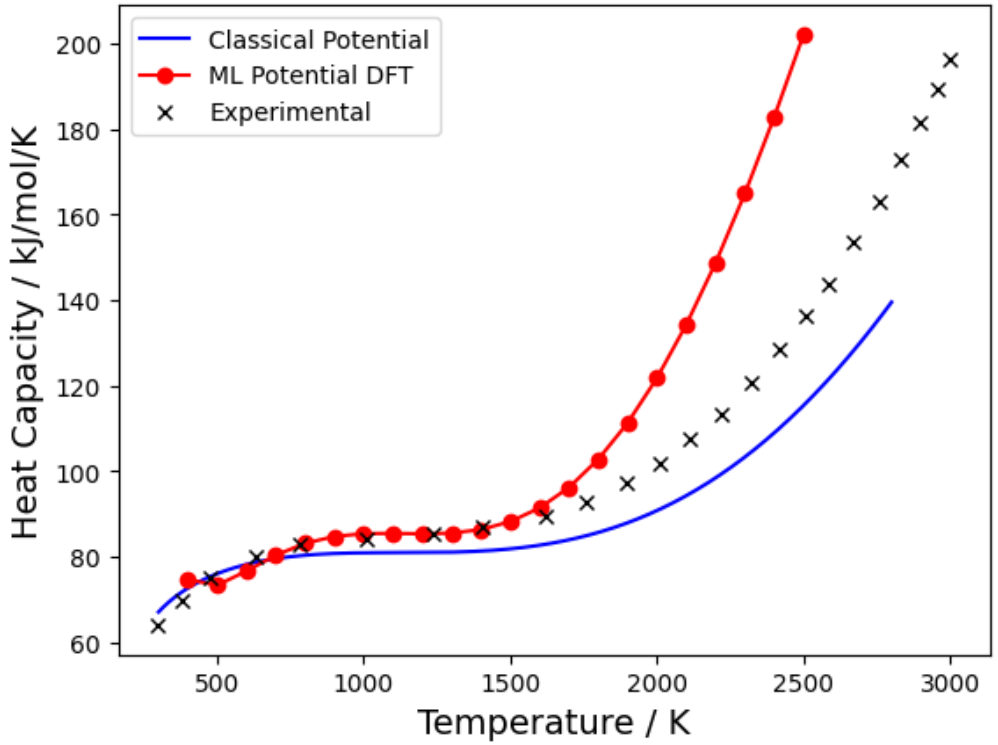


- Automated, iterative, and efficient approach to build dataset to train ML potential
- Structures generated from preliminary ML potentials
- DFT calculation in VASP
  - PBE functional
  - GTH pseudopotentials
  - 2x2x2 supercell

Smith, J. et al. "Automated discovery of a robust interatomic potential for aluminium." *Chem. Sci.*, 2017,**8**, 3192-3203.

Experimental Validation of the ANI Active Learning ML Potential





- 4x4x4 supercell (768 atoms)
- Calculations performed in LAMMPS

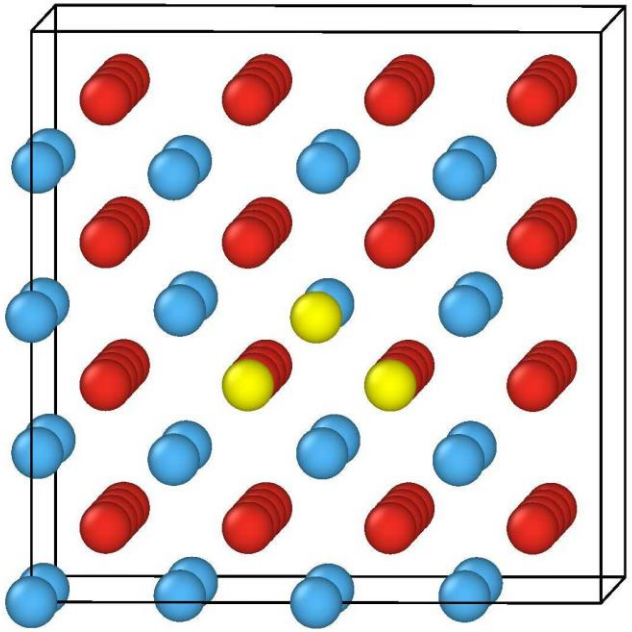
Cooper, M. et al. “A Many-Body Potential Approach to Modelling the Thermomechanical Properties of Actinide Oxides.” *J. Phys.: Condens. Matter* **2014**, *26* (10), 105401.

Defect Energies

Red: oxygen
Blue: uranium
Yellow: vacancy
Dark Blue: interstitial

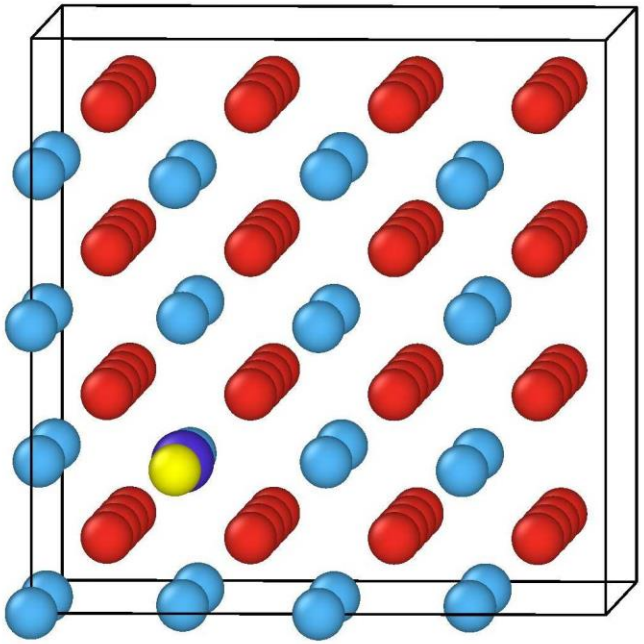
Schottky (SD)

- The removal of one UO2 unit



Frenkel Pair (FP)

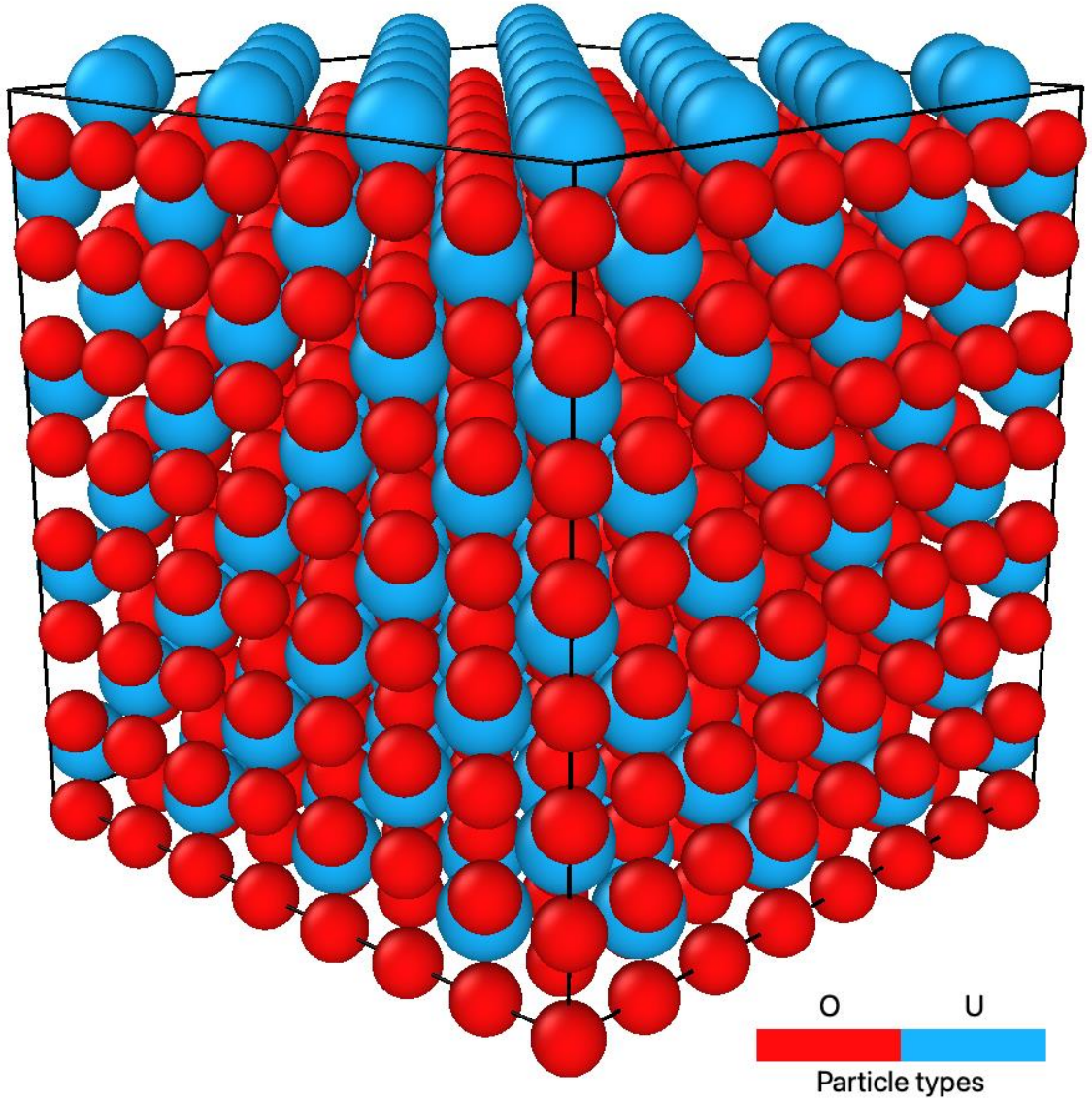
- The removal of an atom and an inclusion of an interstitial atom



Defect Energies

Defect Energy (eV)	ANI-ML	Classical (EAM)	DFT (Lit.)	Defect Energy (eV)	ANI-ML	Classical (EAM)	DFT (Lit.)
SD (isolated)	5.26	10.64	5.6-10.6	U-FP (isolated)	9.40	15.47	10.6-17.2
SD1	4.09	6.18		U-FP1	6.83	11.09	
SD2	3.95	5.27		O-FP (isolated)	5.86	5.73	2.6-5.77
SD3	3.92	5.05		O-FP1	3.94	5.37	
				O-FP2	3.95	4.94	

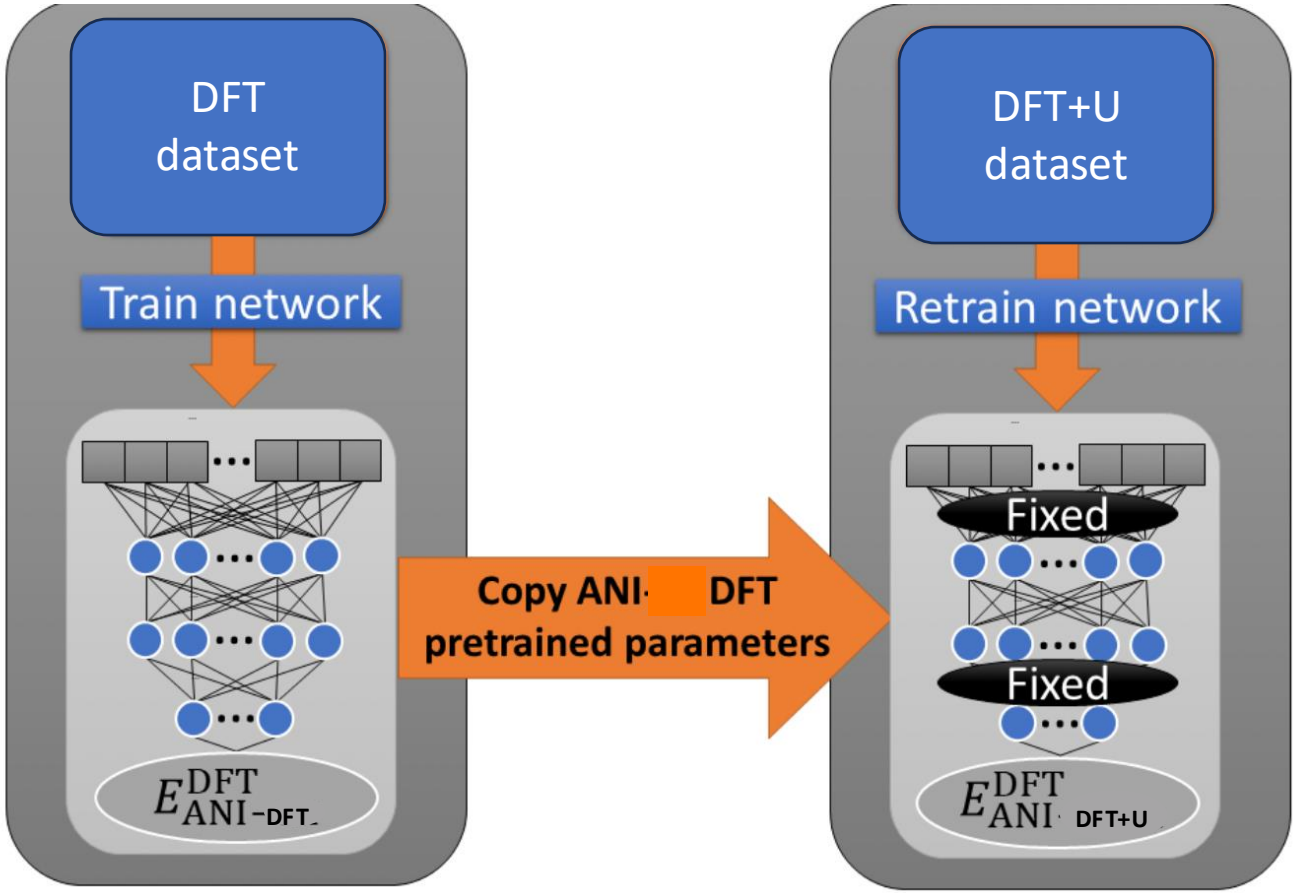
Cooper, M. et al. “A Many-Body Potential Approach to Modelling the Thermomechanical Properties of Actinide Oxides.” *J. Phys.: Condens. Matter* **2014**, *26* (10), 105401.



Transfer Learning Machine Learning

The ANI Potential

Transfer Learning Algorithm

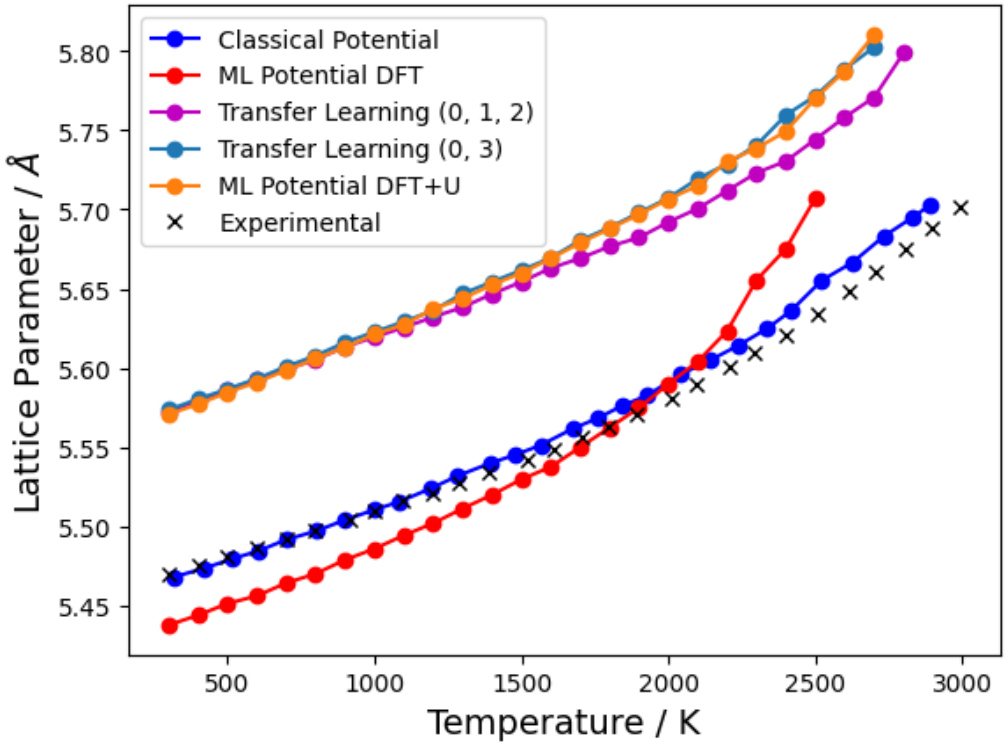


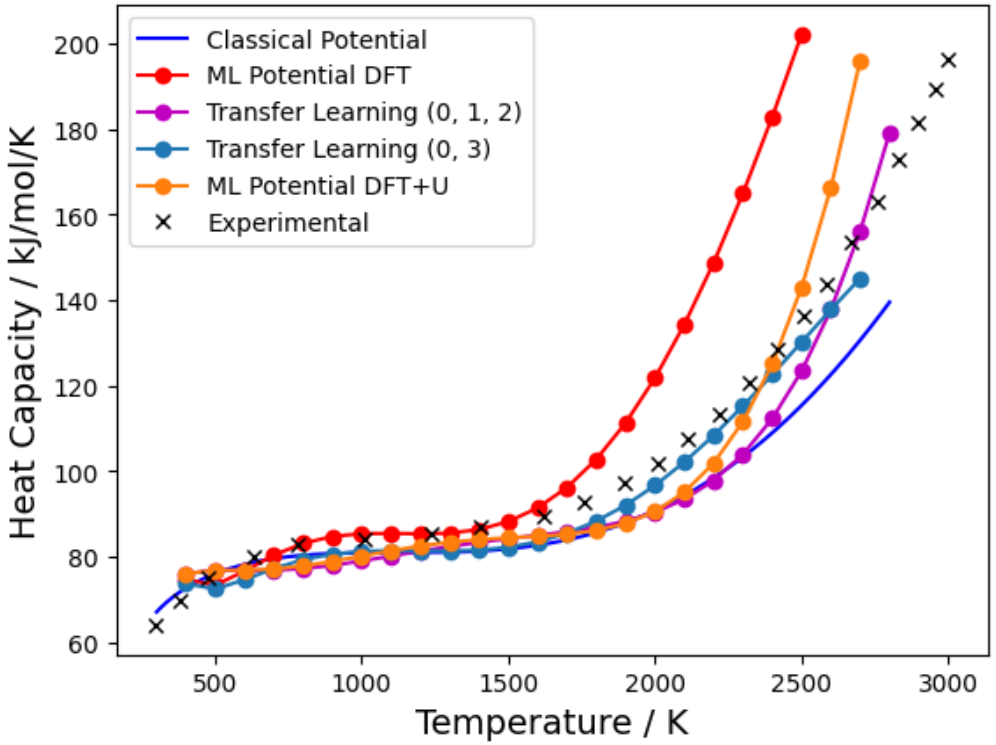
- Train using active learning and a DFT data set
- Subsample structures and recalculate with DFT+U
- Retrain the ML potential with a reduced DFT+U dataset, freezing specific layers

- DFT data set: 25,000+
- DFT+U data set: 1,000

Smith, J. et. al. “Outsmarting Quantum Chemistry Through Transfer Learning.” *ChemRxiv* July 6, 2018. https://doi.org/10.26434/chemrxiv.6744440.v1.

Experimental Validation of the ANI Transfer Learning ML Potential





Cooper, M. et al. “A Many-Body Potential Approach to Modelling the Thermomechanical Properties of Actinide Oxides.” *J. Phys.: Condens. Matter* **2014**, *26* (10), 105401.

Defect Energies: Transfer Learning (0, 1, 2 Layers Frozen)

Defect Energy (eV)	ANI-ML	ANI (TL)	DFT (Lit.)	Defect Energy (eV)	ANI-ML	ANI (TL)	DFT (Lit.)
SD (isolated)	5.26	6.18	5.6-10.6	U-FP (isolated)	9.40	10.76	10.6-17.2
SD1	4.09	4.45		U-FP1	6.83	7.71	
SD2	3.95	4.12		O-FP (isolated)	5.86	6.19	2.6-5.77
SD3	3.92	4.04		O-FP1	3.94	4.71	
				O-FP2	3.95	4.65	

Cooper, M. et al. “A Many-Body Potential Approach to Modelling the Thermomechanical Properties of Actinide Oxides.” *J. Phys.: Condens. Matter* **2014**, *26* (10), 105401.

Conclusions and Continuations

The active learning ML potential has been effective at reproducing values for lower temperatures (<2,300K)

Transfer learning is a promising method for generating results that better reflect experimental data as well as data generated via classical FF

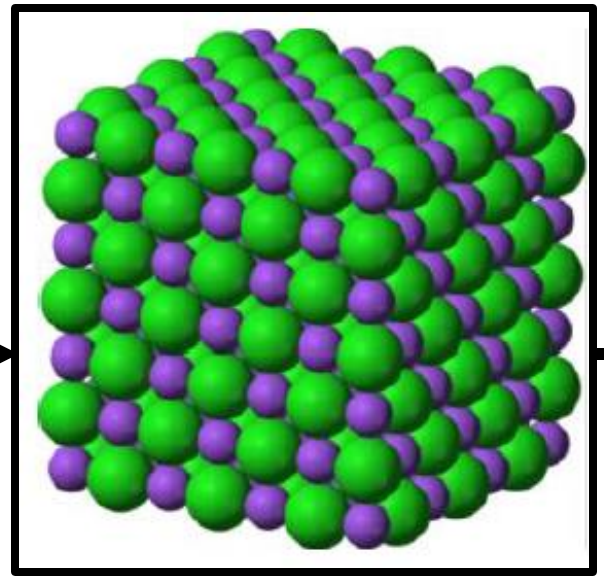
Continuations

To better improve results, the transfer learning method is going to be retrained with more DFT+U data

Extra Slides

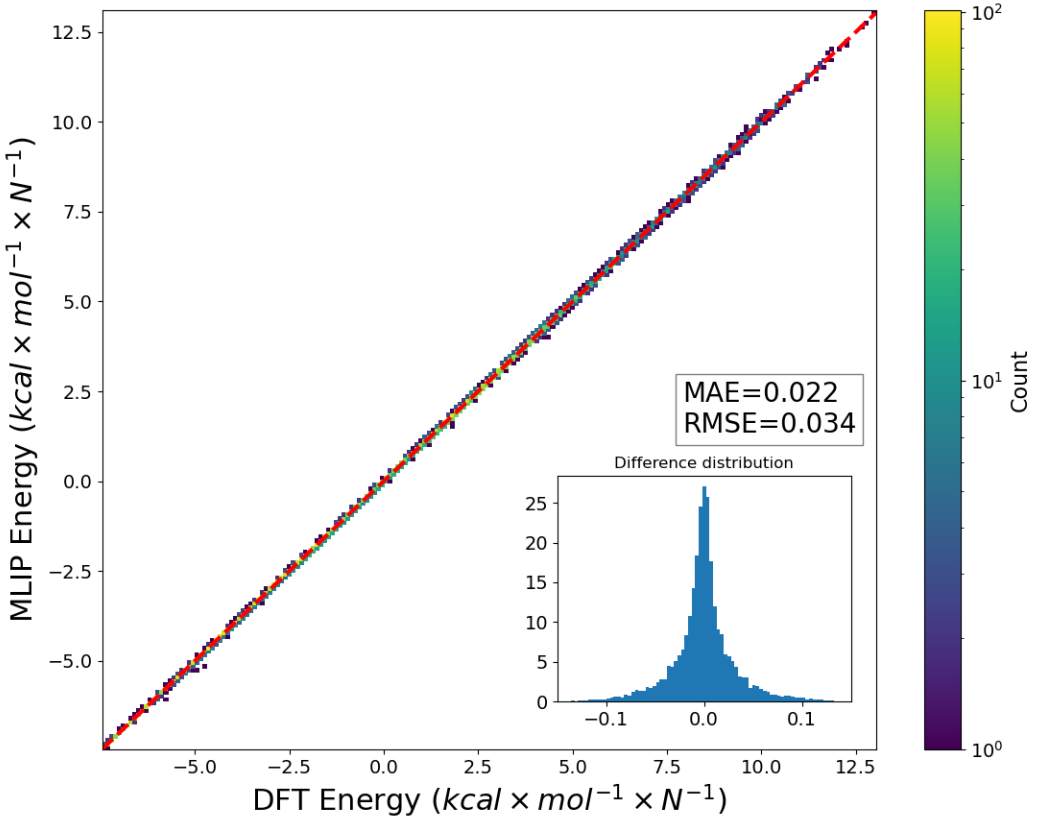
Molecular Dynamics: An Overview of Methods

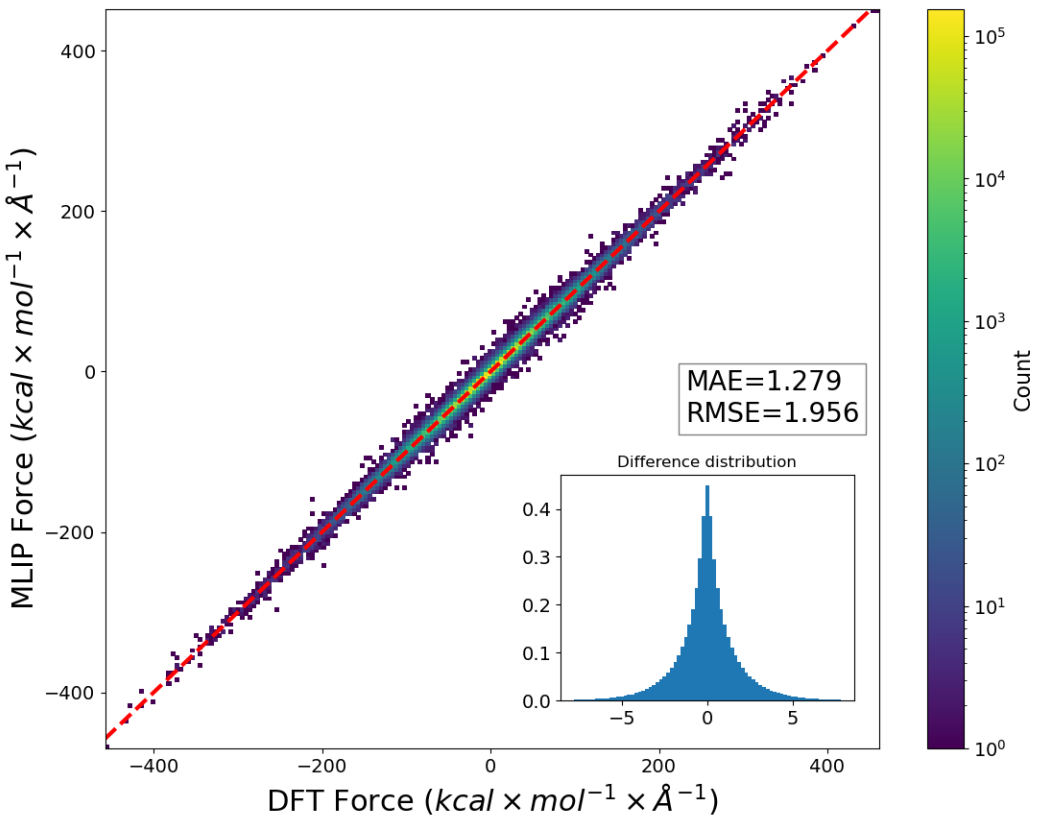
- Density functional theory (DFT) / Kohn-Sham DFT
  - Plane wave (PW)
  - Pseudopotentials
- Classical potentials (force fields):
  - Embedded atom method (EAM)
  - Lennard-Jones 12-6
- Machine-learning (ML) potentials:
  - Neural networks



- Structural properties:
  - Radial distribution function
- Thermophysical properties:
  - Density
  - Melting temperature
- Transport properties:
  - Diffusivity
  - Thermal conductivity

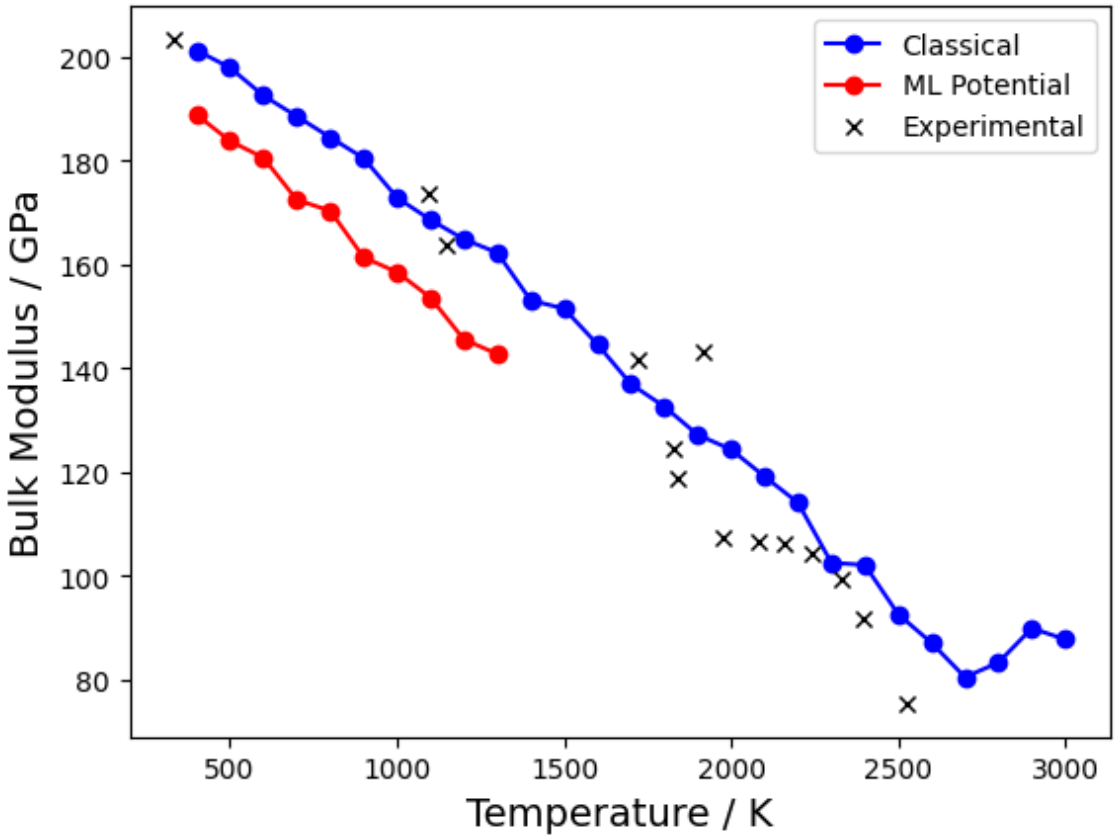
Validation of the ML Potential





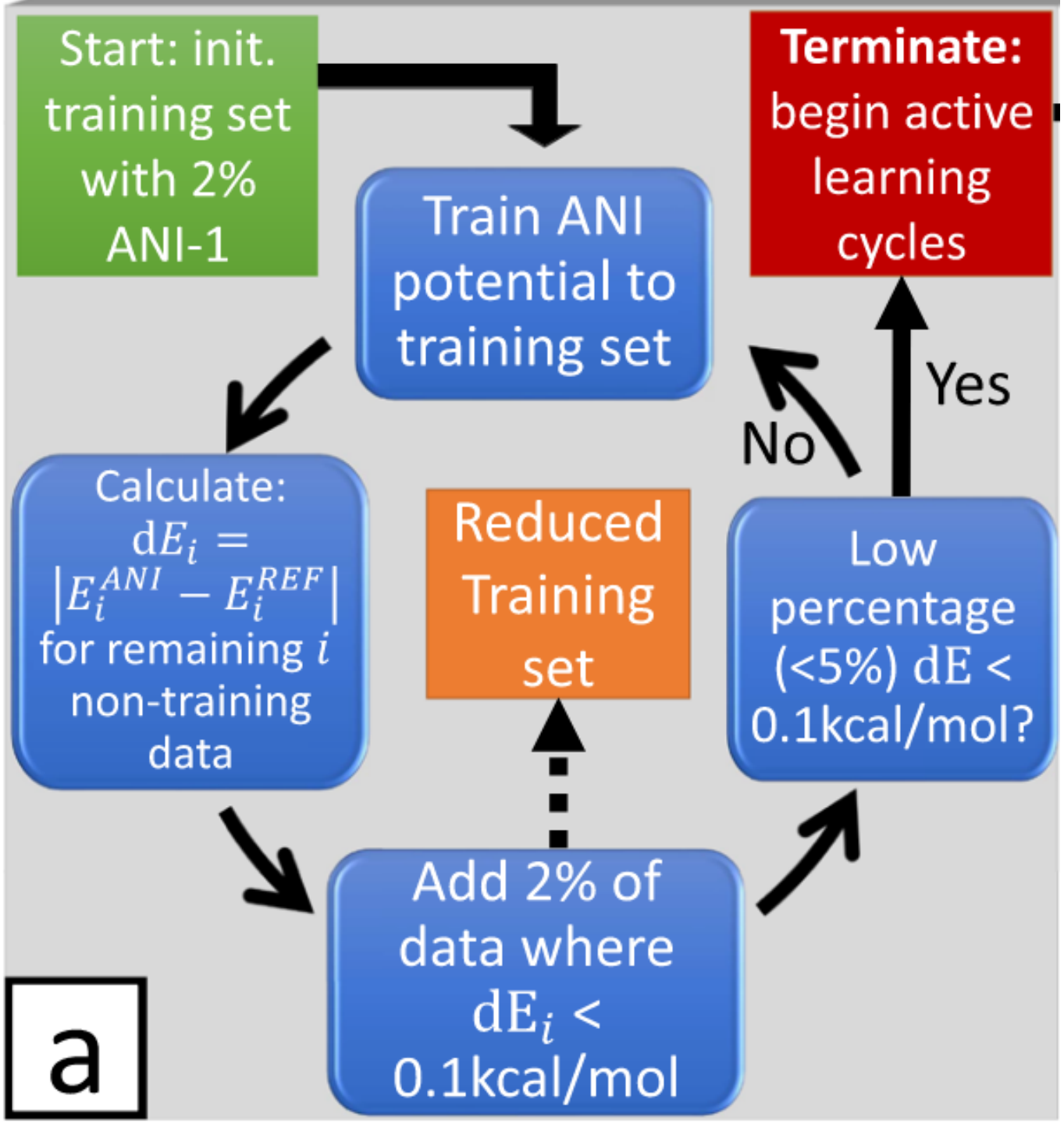
- Dataset of 25,000+ structures

Comparing the ANI (ML) Potential and the Classical EAM Potential

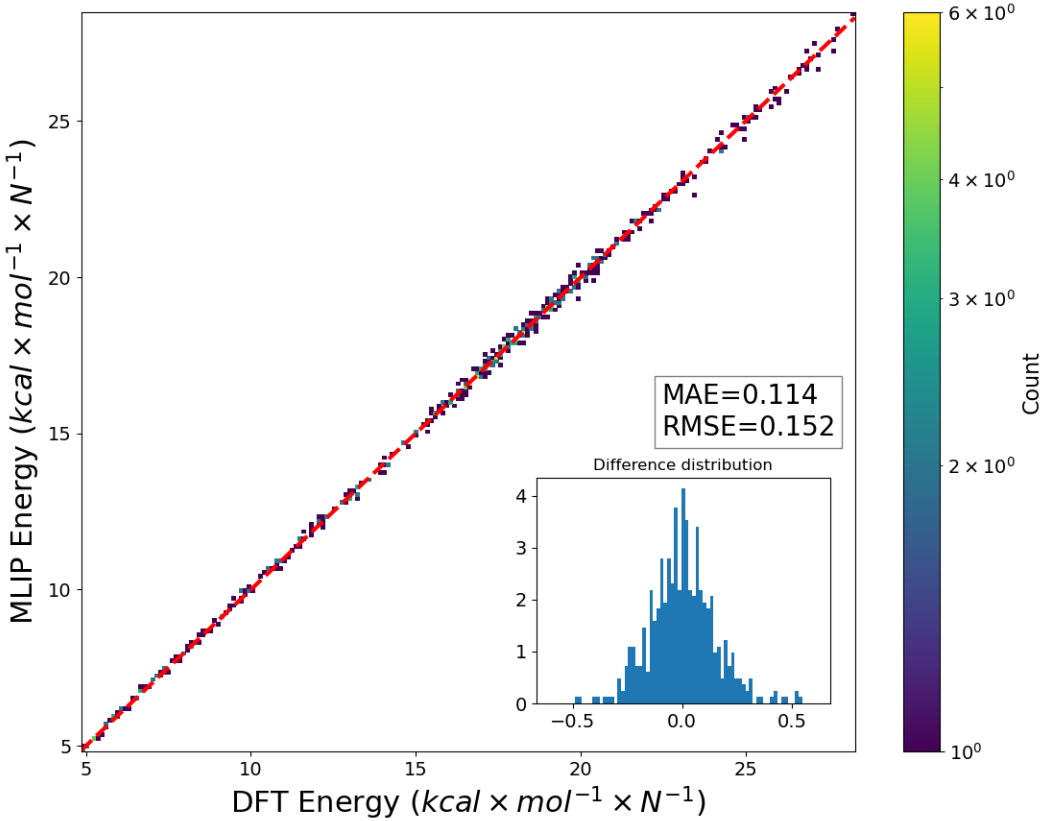


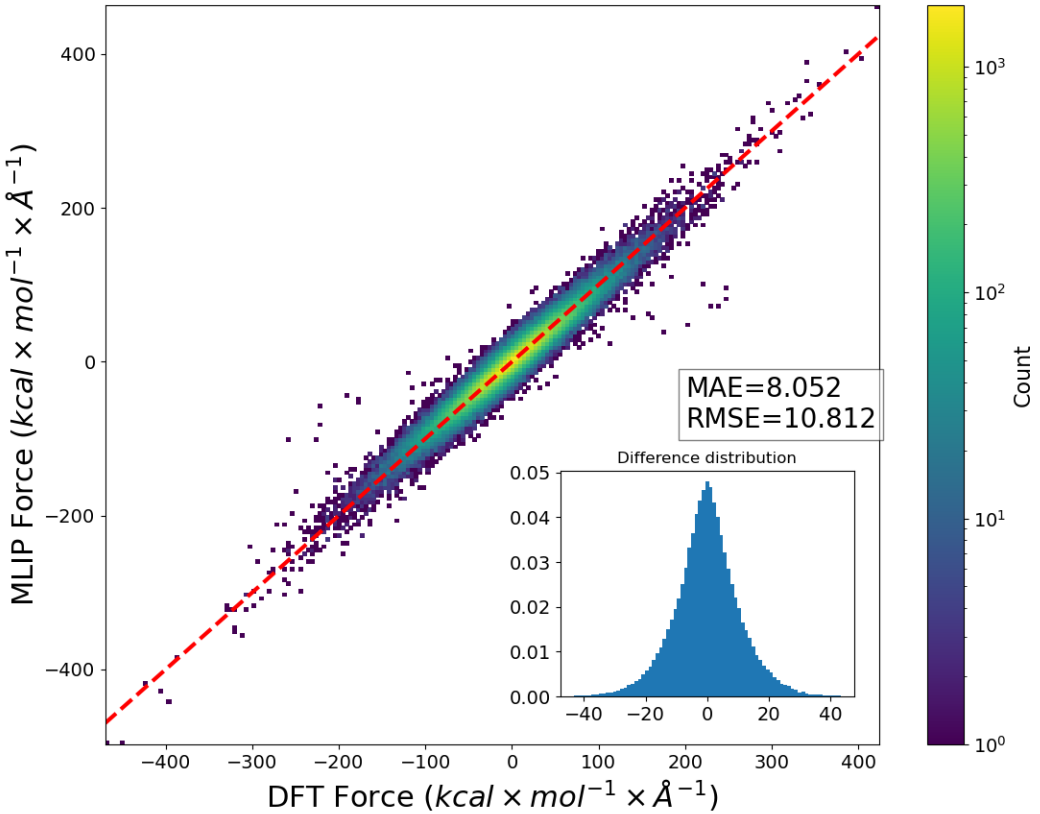
Property	ANI-ML	Classical (EAM)	DFT	Experimental
C_{11} (GPa)	394.5	406.3	371.7	389.3
C_{12} (GPa)	118.3	124.7	117.5	118.7
C_{44} (GPa)	76.3	63.89	66.3	59.7
B (GPa)	207.1	218.6	202.9	208.9

Transfer Learning Algorithm (Cont.)



Validation of the Transfer Learning ML Potential





- Dataset of 1,000 structures
- Layers frozen: 0, 1, 2

Defect Energies: Transfer Learning (0, 1, 2 Layers Frozen)

Defect Energy (eV)	ANI-ML	ANI (0, 1, 2)	Classical (EAM)	Defect Energy (eV)	ANI-ML	ANI (0, 1, 2)	Classical (EAM)
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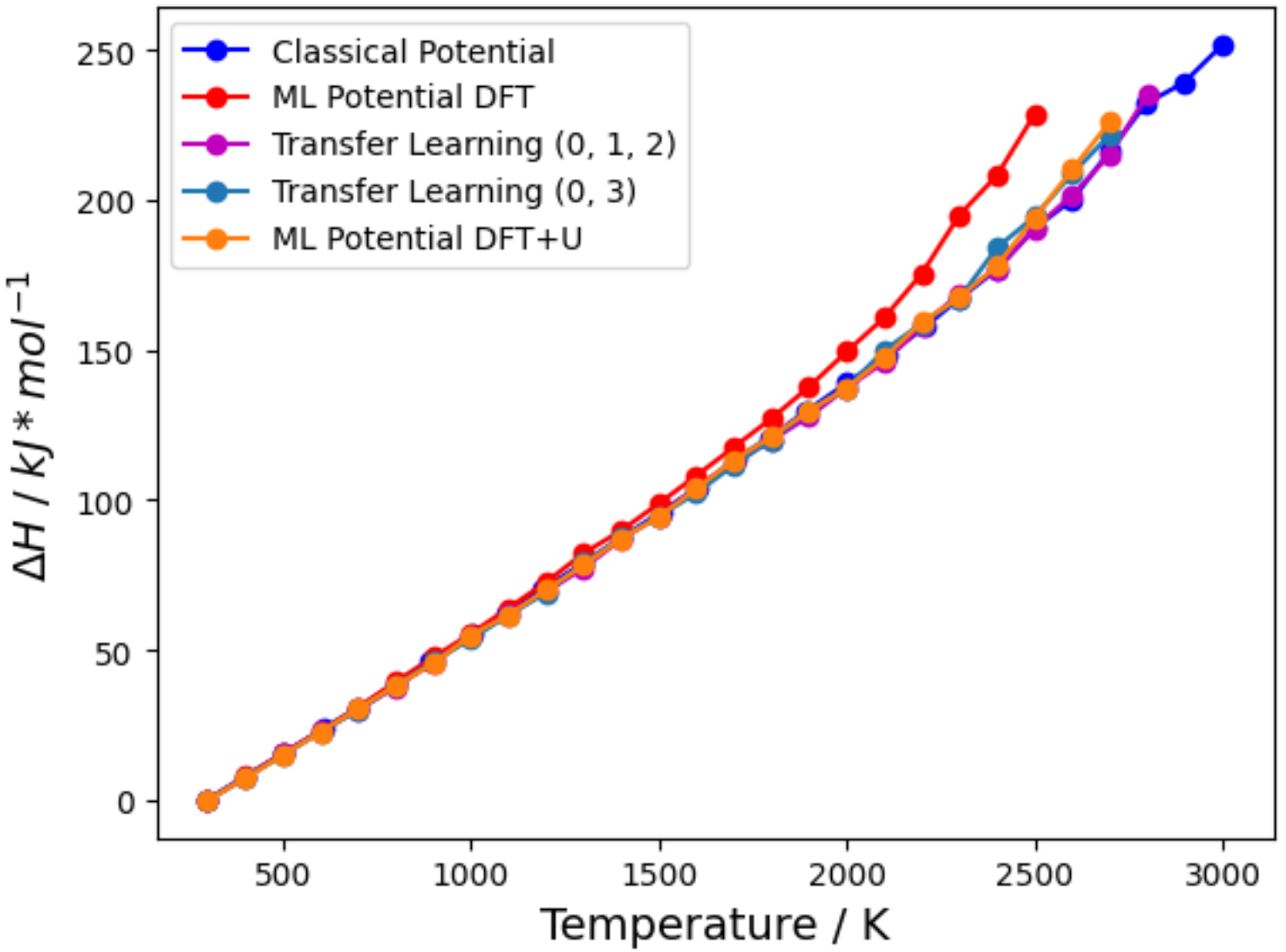
Enthalpy


Classical Potential
ML Potential DFT
Transfer Learning (0, 1, 2)
Transfer Learning (0, 3)
ML Potential DFT+U
250
200
150
100
50
0
500
1000
1500
2000
2500
3000
ΔH / kJ * mol⁻¹
Temperature / K