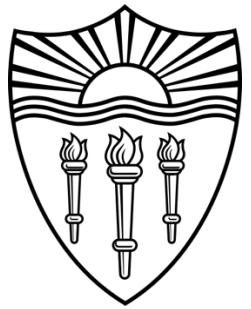

Combining active learning machine learning with transfer learning: Building better interatomic potentials for nuclear fuels

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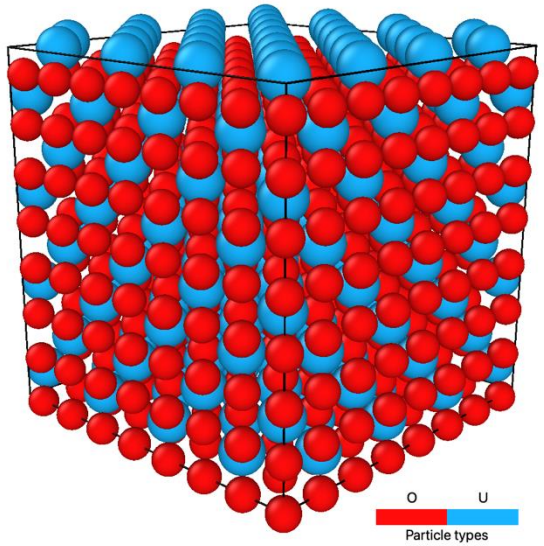
Background: Simulations of Nuclear Fuels

Why do we simulate nuclear fuels?

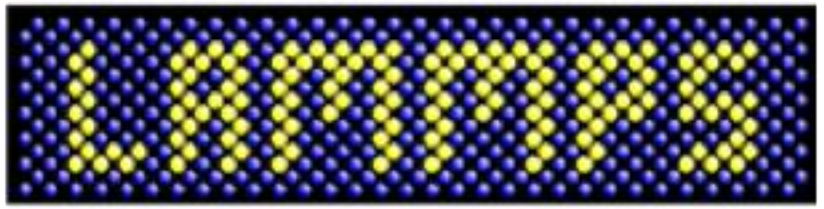
Allows a better understanding of nuclear fuels under varying conditions which can be difficult to investigate experimentally.

Simulations help predict nuclear fuel properties.

*Can we create a machine learning interatomic potential (**MLIP**) that can accurately predict values within reasonable error to experimental values?*



Uranium Dioxide (Nuclear Cell)



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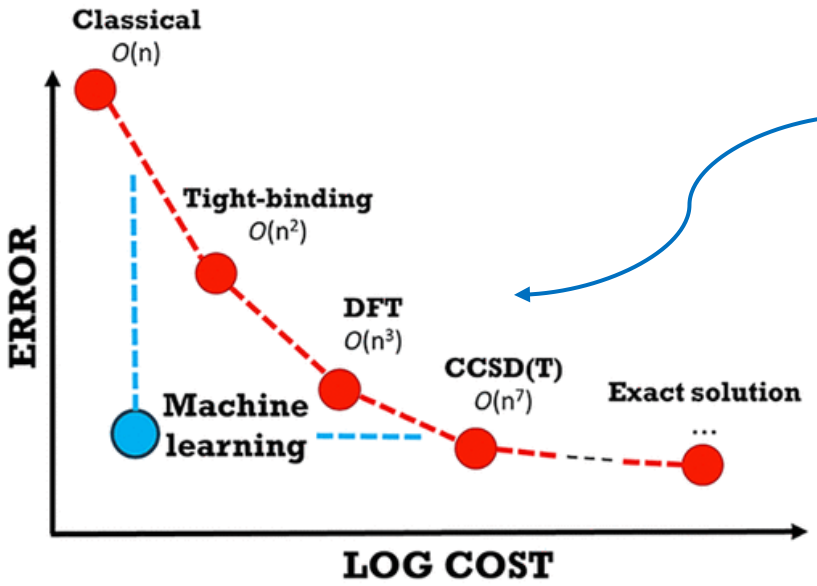
Background: Machine Learning

Why use machine learning?

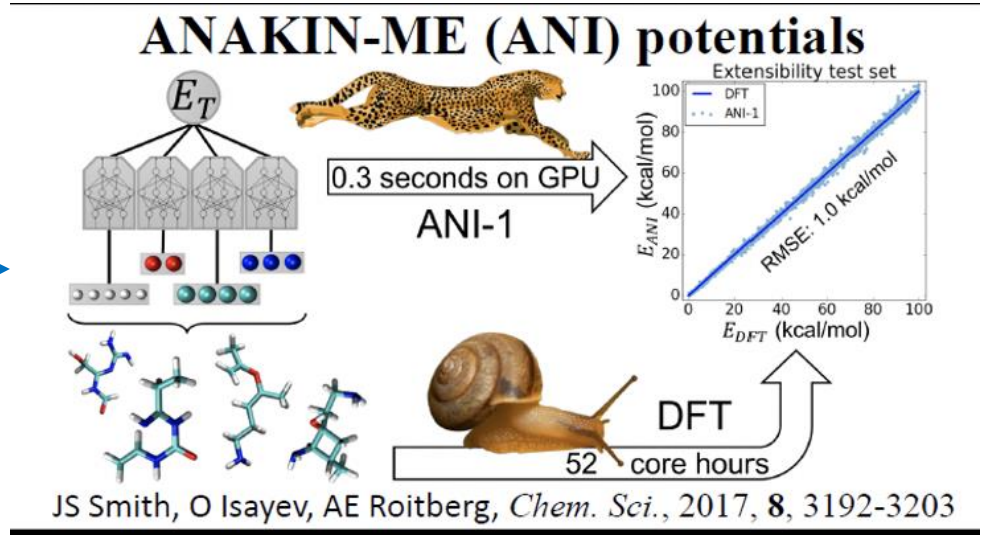
1. Active Learning

2. Transfer Learning



Computational affordability without sacrificing accuracy

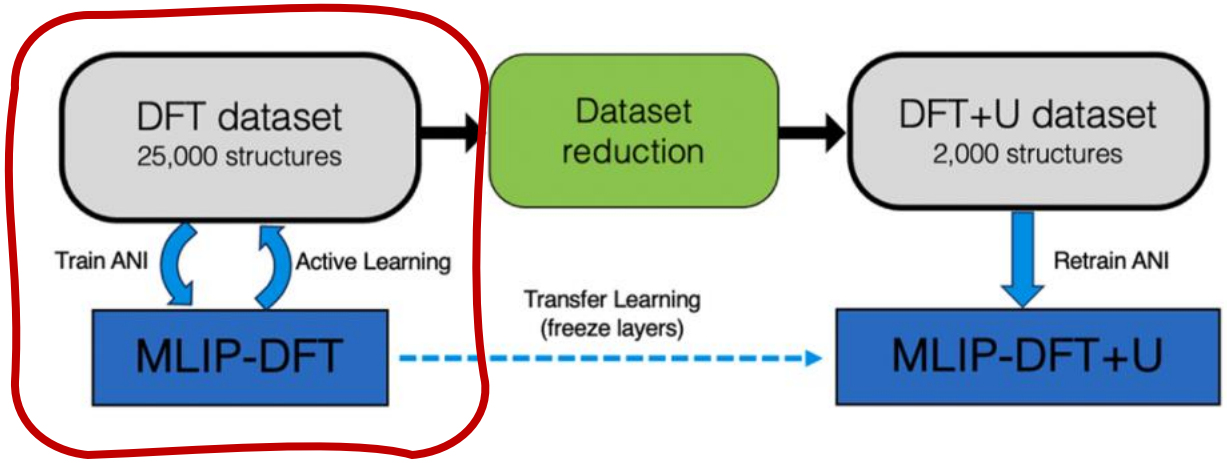
Computational speed



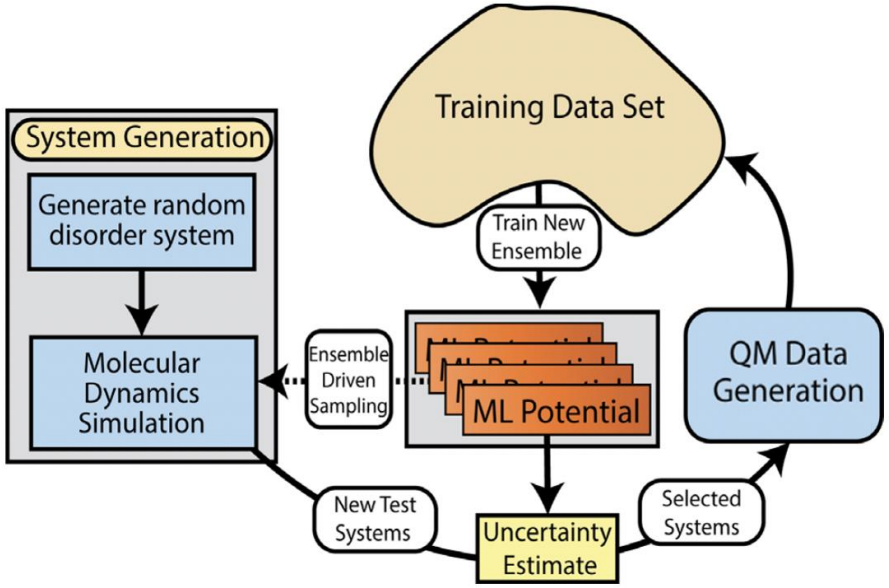
Kulichenko, M. et al. "The Rise of Neural Networks for Materials and Chemical Dynamics." *J. Phys. Chem. Lett.*, 2021,**12**,26,6227-6243

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

Methods: Building a Machine Learning Potential



DFT *Density Functional Theory*
MLIP *Machine Learning Interatomic Potential*
DFT+U *DFT plus Hubbard Parameter*

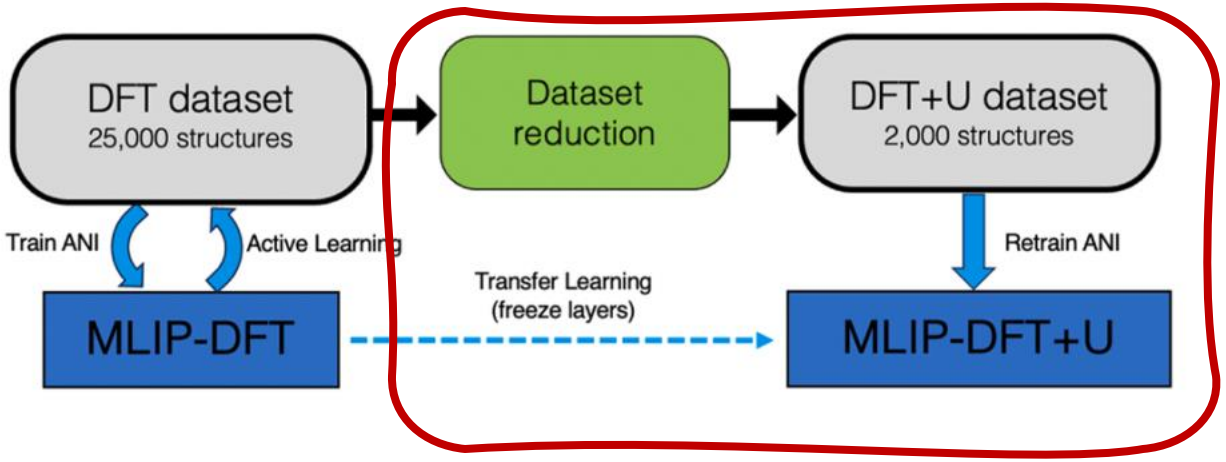


Active Learning ML

An iterative method that automatically trains the MLIP

1. Sampling
   - Atomic coordinates are generated and added to training dataset
2. Labeling
   - Energies and forces for each atom (in each system) assigned
3. Training
   - Fit the MLIP to the training dataset, described by steps 1 and 2

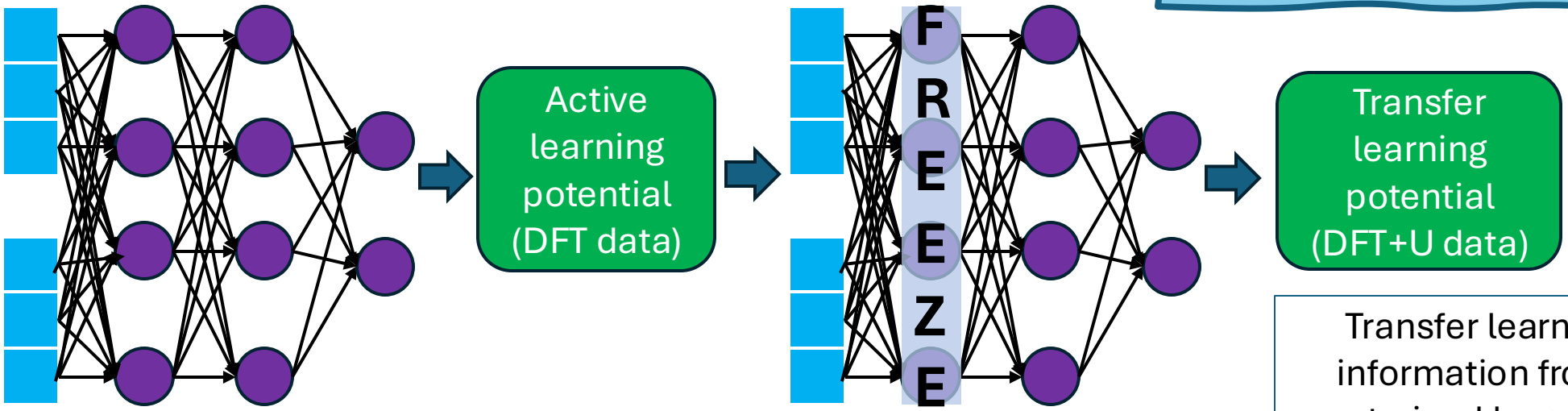
Methods: Building a Machine Learning Potential



Question: Why did we add DFT+U structures?

Answer: More accurate! DFT+U includes antiferromagnetic properties important in the ground state of uranium and other actinides

Tradeoff: High cost for high accuracy



Transfer learning retains information from the pre-trained larger dataset

Results: Temperature Dependent Properties

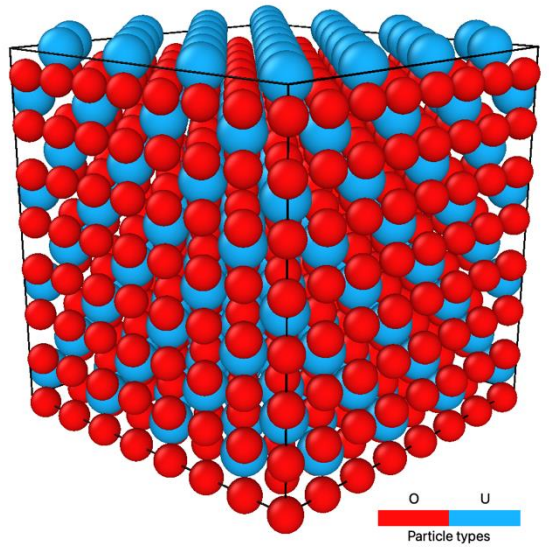
Two different machine learning interatomic potentials (MLIPs):

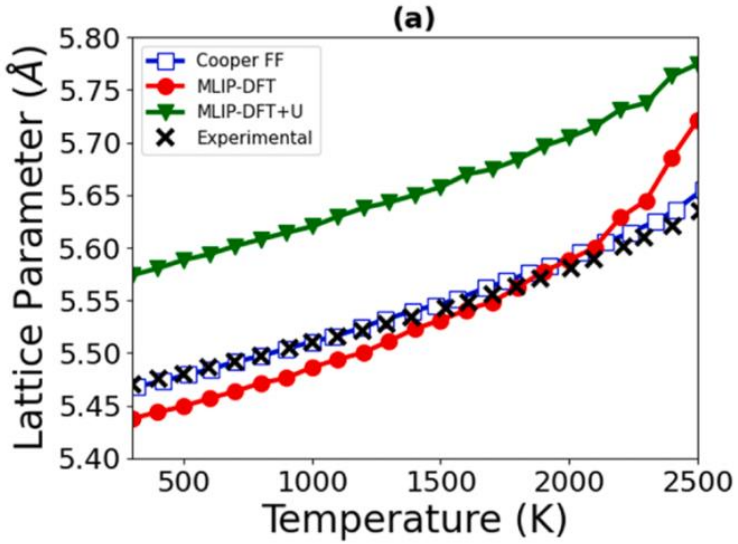
1. Sole DFT data
2. Mixture of DFT+U and DFT data

Dataset consists of:

- 96 atom supercell
- MD simulations from 300 – 2,500 K
- Both zero pressure and non-zero pressure systems
- Structures containing point defects

4x4x4 supercell
768 atoms
UO_2





MLIP-DFT+U overestimates lattice parameters by ~0.10Å

Qualitatively, MLIP-DFT+U follows a similar trend to experiment

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.

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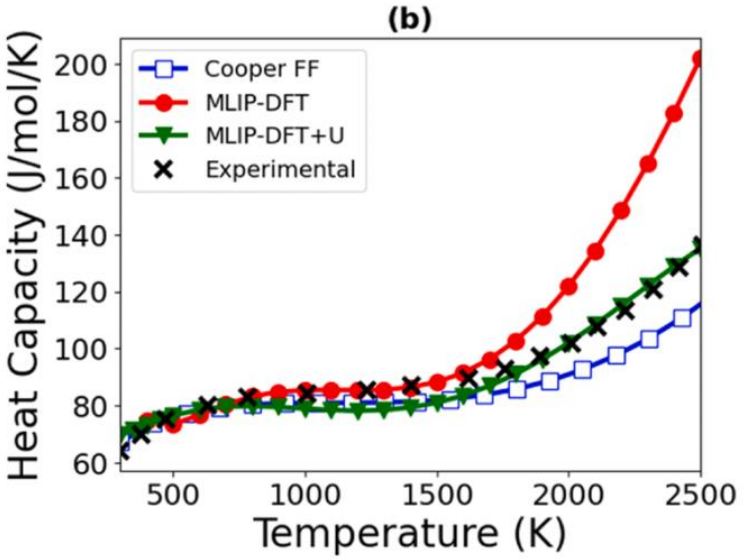
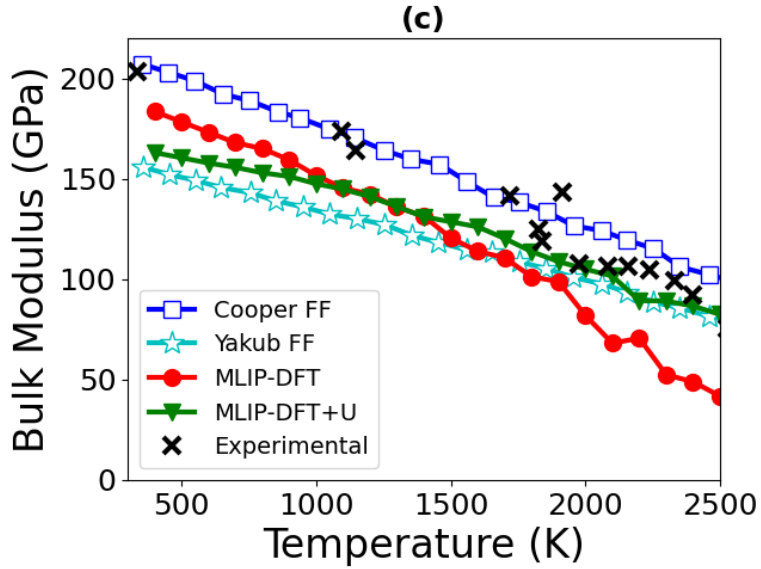
Results: Temperature Dependent Properties



MLIP-DFT+U provides great accuracy compared to experiment at high temperatures

MLIP-DFT provides great accuracy compared to experiment at lower temperatures

$$c_p = \frac{1}{n}\left(\frac{\partial H}{\partial T}\right)_p$$



Qualitatively, MLIP-DFT follows a similar trend to experiment

MLIP-DFT+U performs better than the Yakub force field

$$K = -V\left.\frac{dP}{dV}\right|_{V=V_0}$$

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Results: Zero Temperature Properties

Property	MLIP-DFT	MLIP-DFT+U	FF	DFT	DFT+U	Exp.
a (Å)	5.45 (-0.42%)	5.51 (+0.68%)	5.45	5.42	5.54	5.473*
C_{11} (GPa)	389.47 (+0.04%)	344.75 (-11.44%)	406.3	371.7	393.8	389.3
C_{12} (GPa)	121.21 (+2.04%)	118.20 (-0.42%)	124.7	117.5	114.7	118.7
C_{44} (GPa)	77.98 (+30.62%)	37.19 (-37.71%)	63.89	66.3	63.9	59.7
B (GPa)	207.80 (-0.53%)	190.98 (-8.58%)	218.6	202.9	197	208.9

Lattice Parameter

Elastic Constants

Similar to the temperature dependent lattice parameter, DFT+U overestimates the zero-temperature lattice parameter

The MLIP-DFT shows agreement with experimental values

**Value at ambient temperature*

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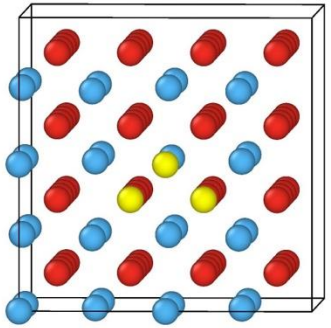
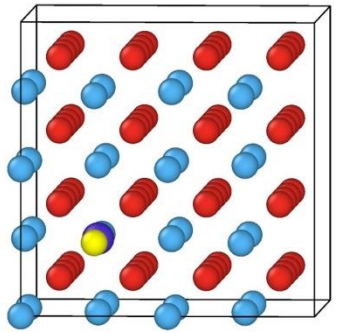
Results: Defect Energies

Schottky (SD)
The Removal of one UO_2 unit



Frenkel Pair (FP)
The removal of an atom and an inclusion of an interstitial atom



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

MLIP-DFT+U performs better than classical force fields when compared to experiment

Defect energy (eV)	MLIP-DFT	MLIP-DFT+U	FF	DFT (Lit.)	DFT+U (Lit.)	Exp.
$SD_{isolated}$	5.26	6.31	10.64	5.6-10.6	4.2-11.8	6.0-7.0
SD_1	4.09	4.30	6.18			
SD_2	3.95	3.96	5.27			
SD_3	3.92	3.86	5.05			
$U\text{-}FP_{isolated}$	9.40	10.15	15.47	10.6-17.2	9.1-16.5	9.5
$U\text{-}FP_1$	6.83	7.30	11.09			
$O\text{-}FP_{isolated}$	5.86	5.25	5.73	2.6-5.77	2.4-7.0	3.0-4.0
$O\text{-}FP_1$	3.94	4.36	5.37			
$O\text{-}FP_2$	3.95	4.26	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.

Conclusions

The active learning MLIP paired with transfer learning was successful in reproducing important characteristics of uranium oxide.

By including DFT+U data, the accuracy of the MLIP increased, showing the importance of antiferromagnetism in nuclear fuels.

Moving Forward

This model can be improved by training the MLIP using experimental data.

This model can be expanded to include other novel nuclear fuels, including UN and UC.

Acknowledgements

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