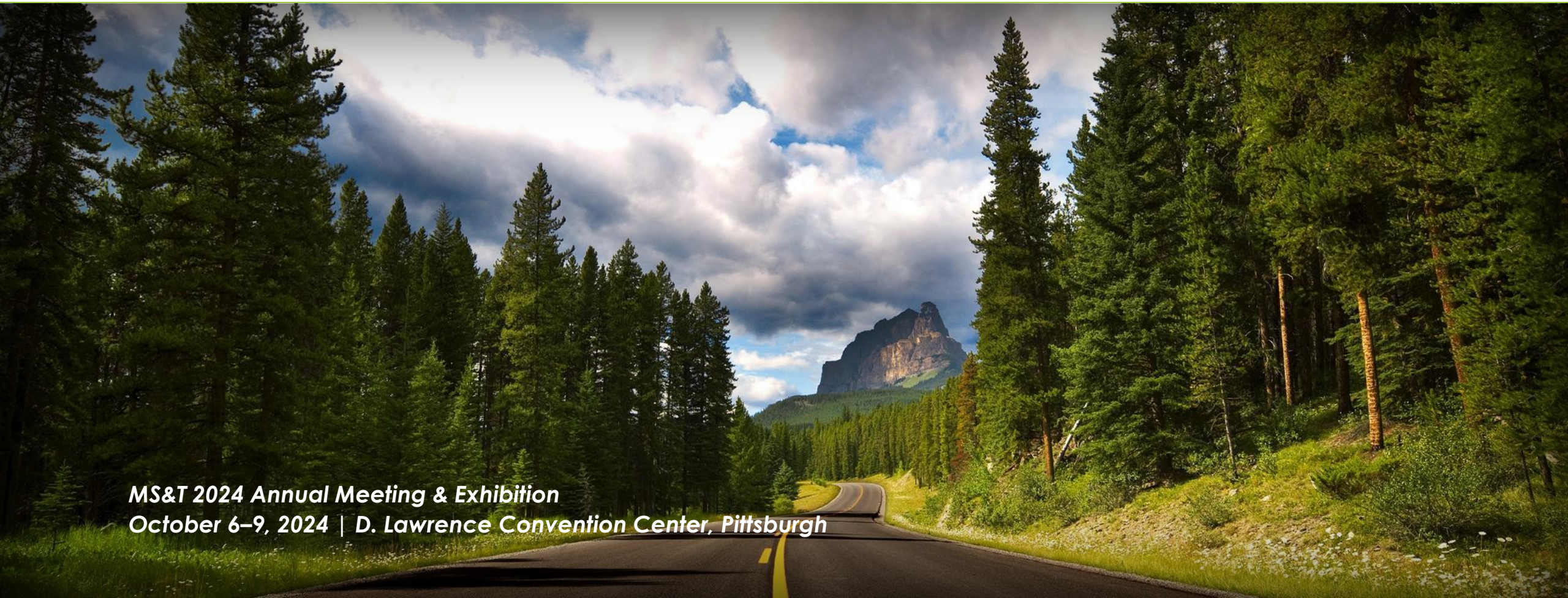
Revolutionizing Materials Design: The Intersection of Quantum Mechanics and Data Modeling

Wissam A. Saidi
Computational Materials Engineering Team, Head
National Energy Technology Laboratory



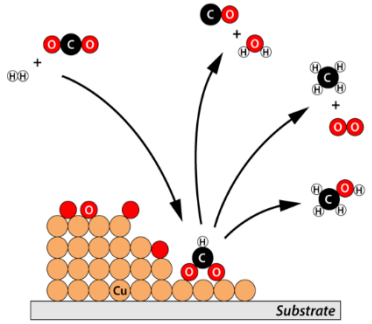
Saidi Lab

Department Chair

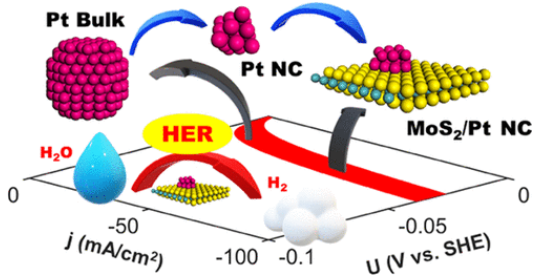
Department of Mechanical Engineering and

Materials Science

University of Pittsburgh

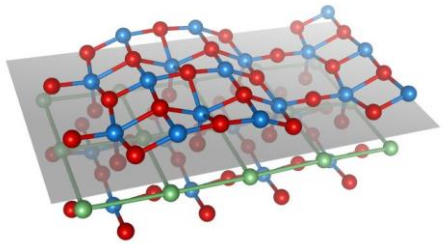


Catalysis

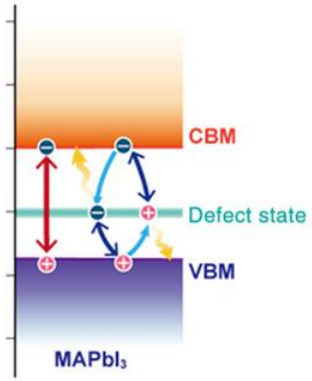


Electrocatalysts

Oxidation and Corrosion



Nano Materials



Solar cells

Materials Design for energy technologies through multiscale simulations

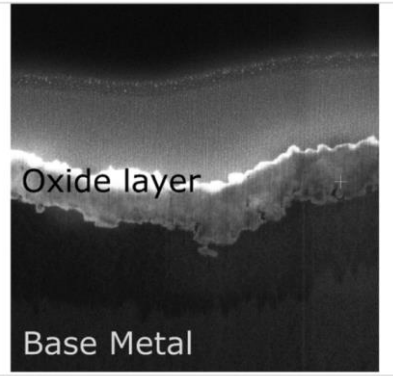
Industrial & Engineering Chemistry Research > Vol 58/Issue 3 > Article

Cite Share Jump to Expand

ARTICLE | December 24, 2018

Mechanism behind the Inhibiting Effect of CO_2 on the Oxidation of Al–Mg Alloys

Nicholas Smith*, Brian Gleeson, Wissam A. Saidi, Anne Kvithyld, and Gabriella Tranell



Effects of CO_2 Cover Gas and Yttrium Additions on the Oxidation of AlMg Alloys

Conference paper | First Online: 16 February 2019

pp 1025–1032 | Cite this conference paper

N. Smith, B. Gleeson, W. Saidi, A. Kvithyld & G. Tranell

National Energy Technology Laboratory (NETL)

One of 17 U.S. Department of Energy (DOE) national laboratories; producing technological solutions to America's energy challenges.

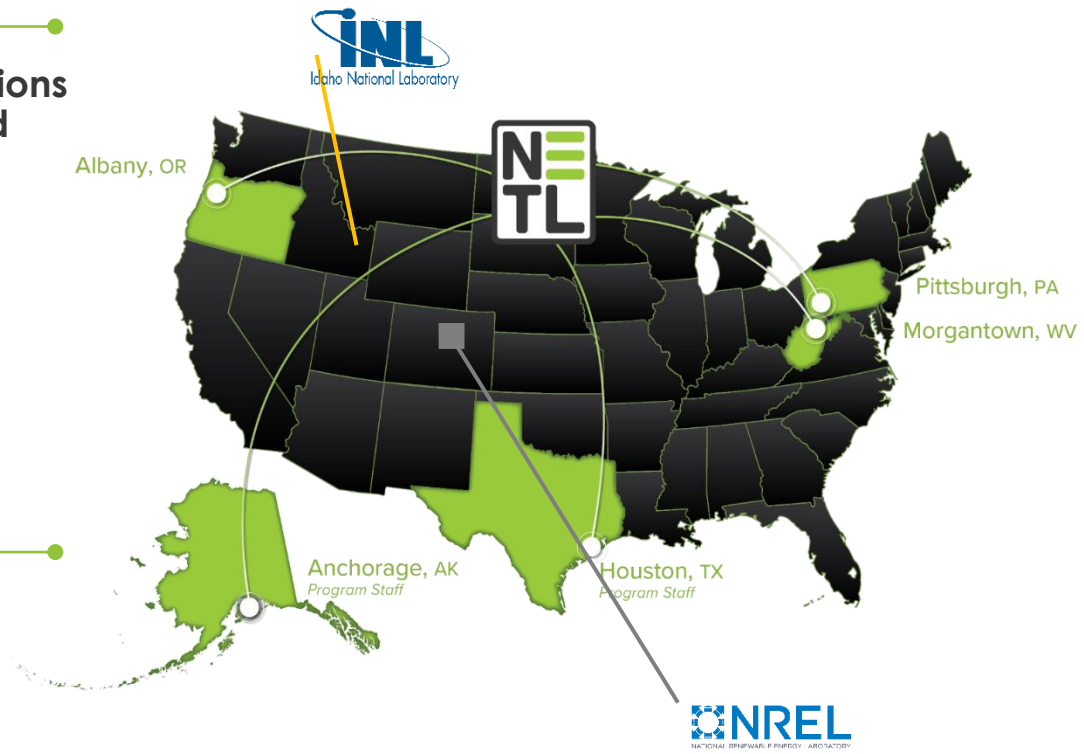
MISSION

Driving innovation and delivering solutions for an environmentally sustainable and prosperous energy future:

- Ensuring affordable, abundant and reliable energy that drives a robust economy and national security, while
- Developing technologies to manage carbon across the full life cycle, and
- Enabling environmental sustainability for all Americans.

VISION

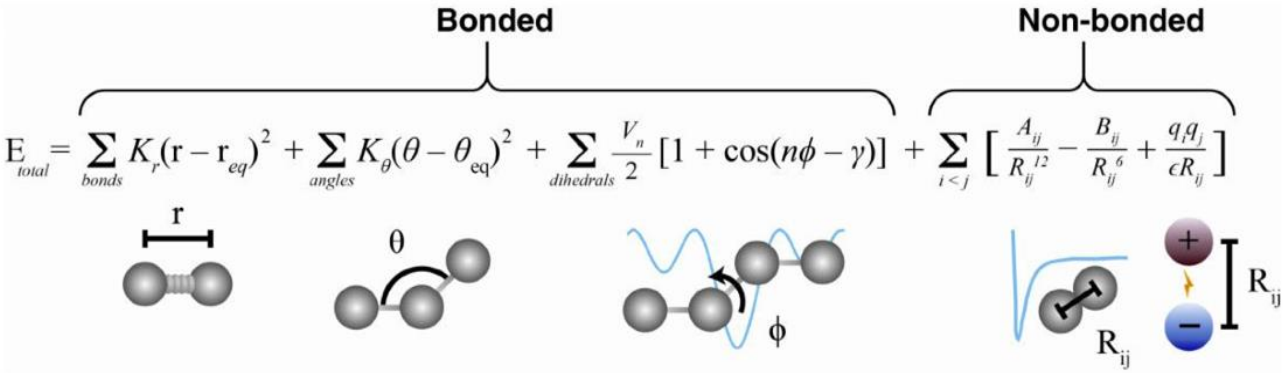
To be the nation's premier energy technology laboratory, delivering integrated solutions to enable transformation to a sustainable energy future.



- NETL has **three** research laboratories
- **One of three** applied research national labs
- Government owned & operated
- Leader in cutting-edge research in CO_2 conversion to higher-value products
- Only National Lab dedicated to carbon management research

Atomistic Force-Fields Indispensable Tool for Materials Design

Traditional Approach



Limitations of traditional forcefields:

- **Time-intensive (low training transparency)**
- **Limited accuracy and transferability**
- **Unavailable for many materials**

AI/ML

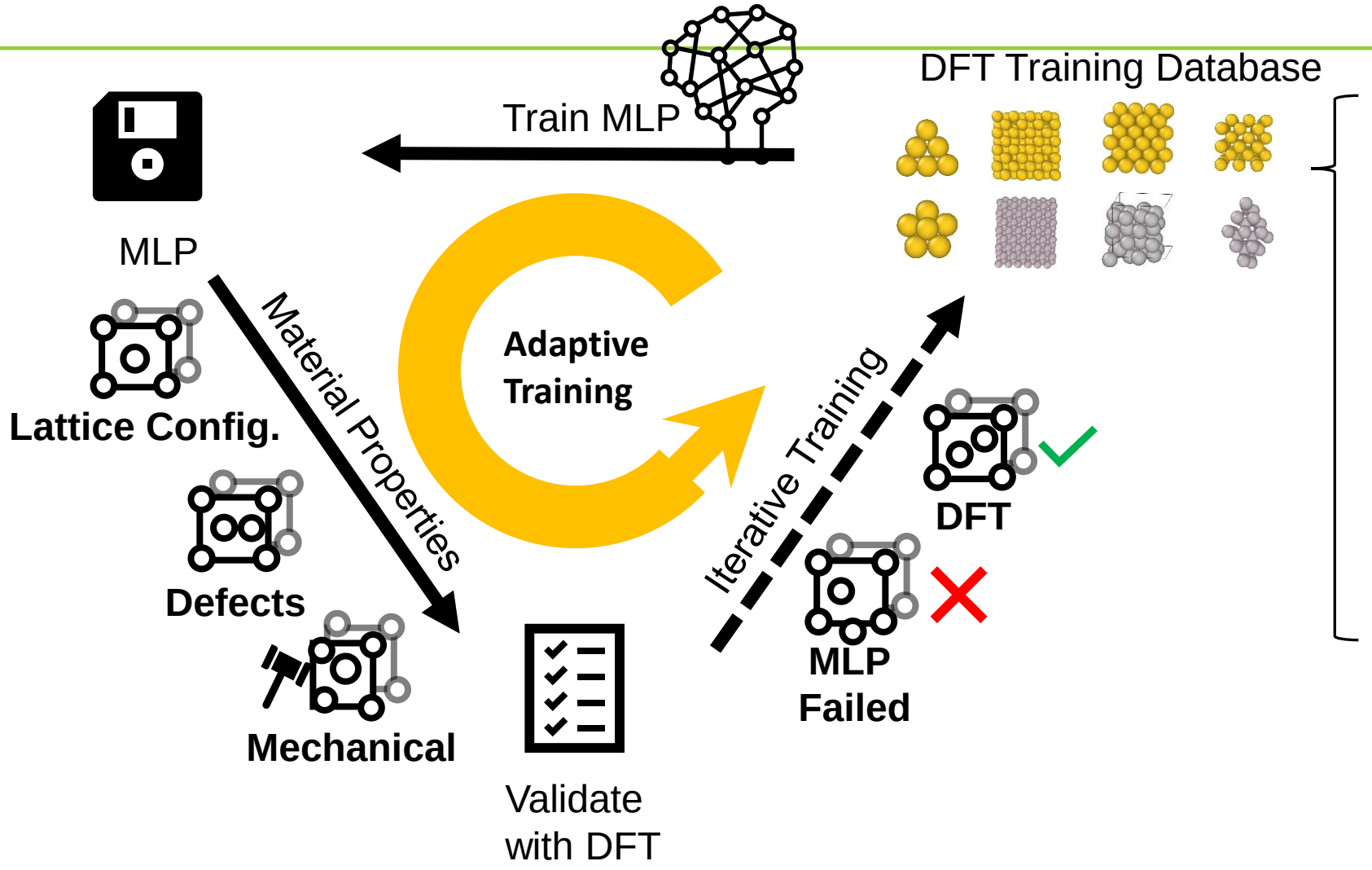
Big data

Noble prize 2024 in physics: Hopfield and Hinton

ML atomistic potentials accelerate DFT without comprising accuracy

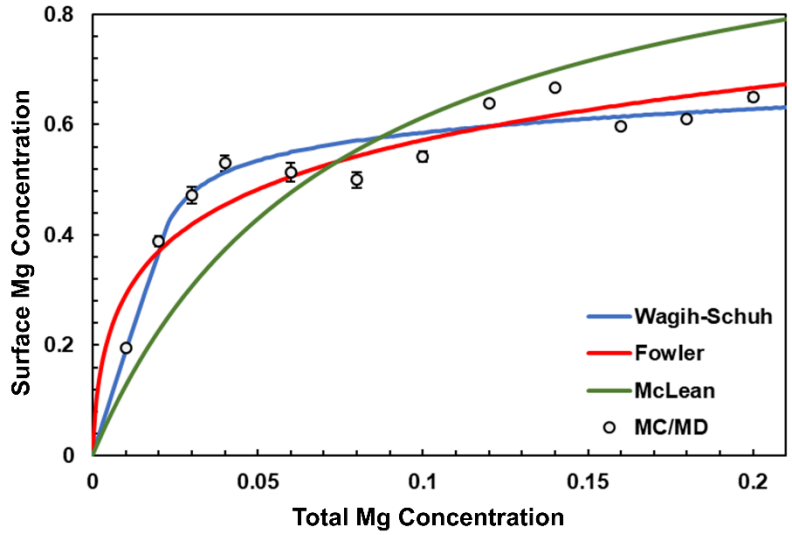
- **Melting**
- **Oxidation of binary alloy system**

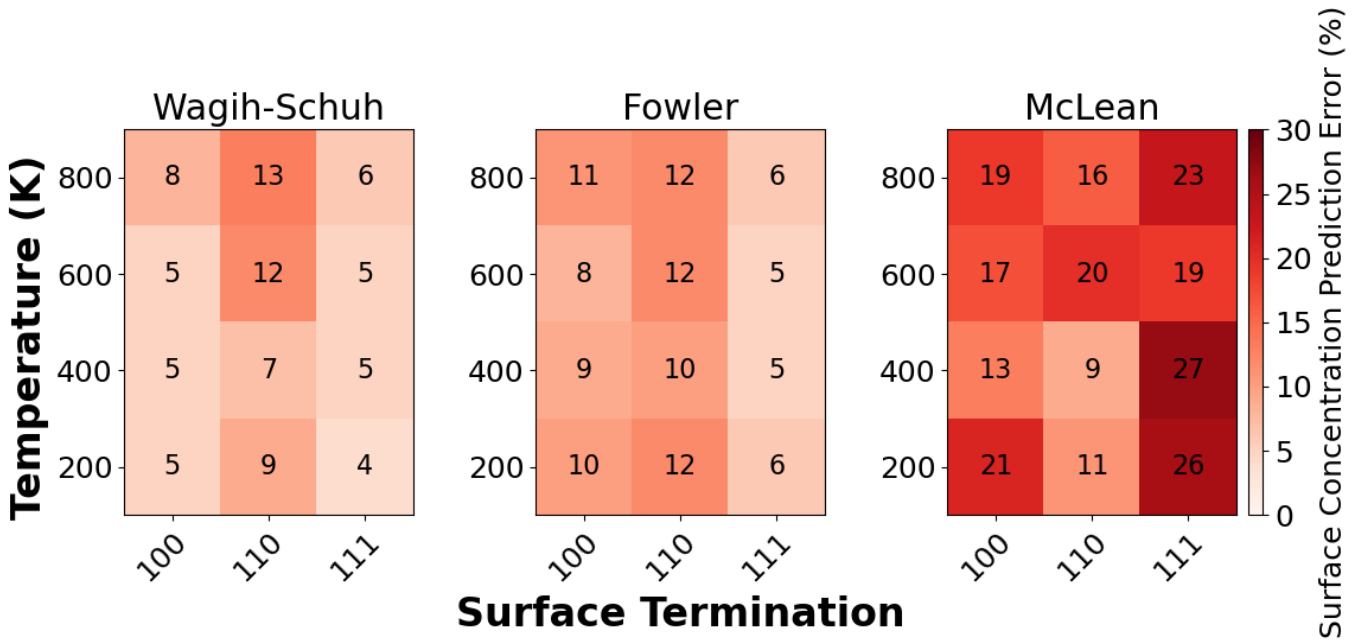
MLP Workflow: Training and Validation



- ❑ Crystalline systems
- ❑ Amorphous systems
- ❑ monometalics and intermetallics
- ❑ slab models
- ❑ alloy systems
- ❑ nanoparticles
- ❑ ….
- ❑ ….
- ❑ ….

Temperature Dependent Mg Surface Segregation AlMg Alloys: Isotherm Comparison





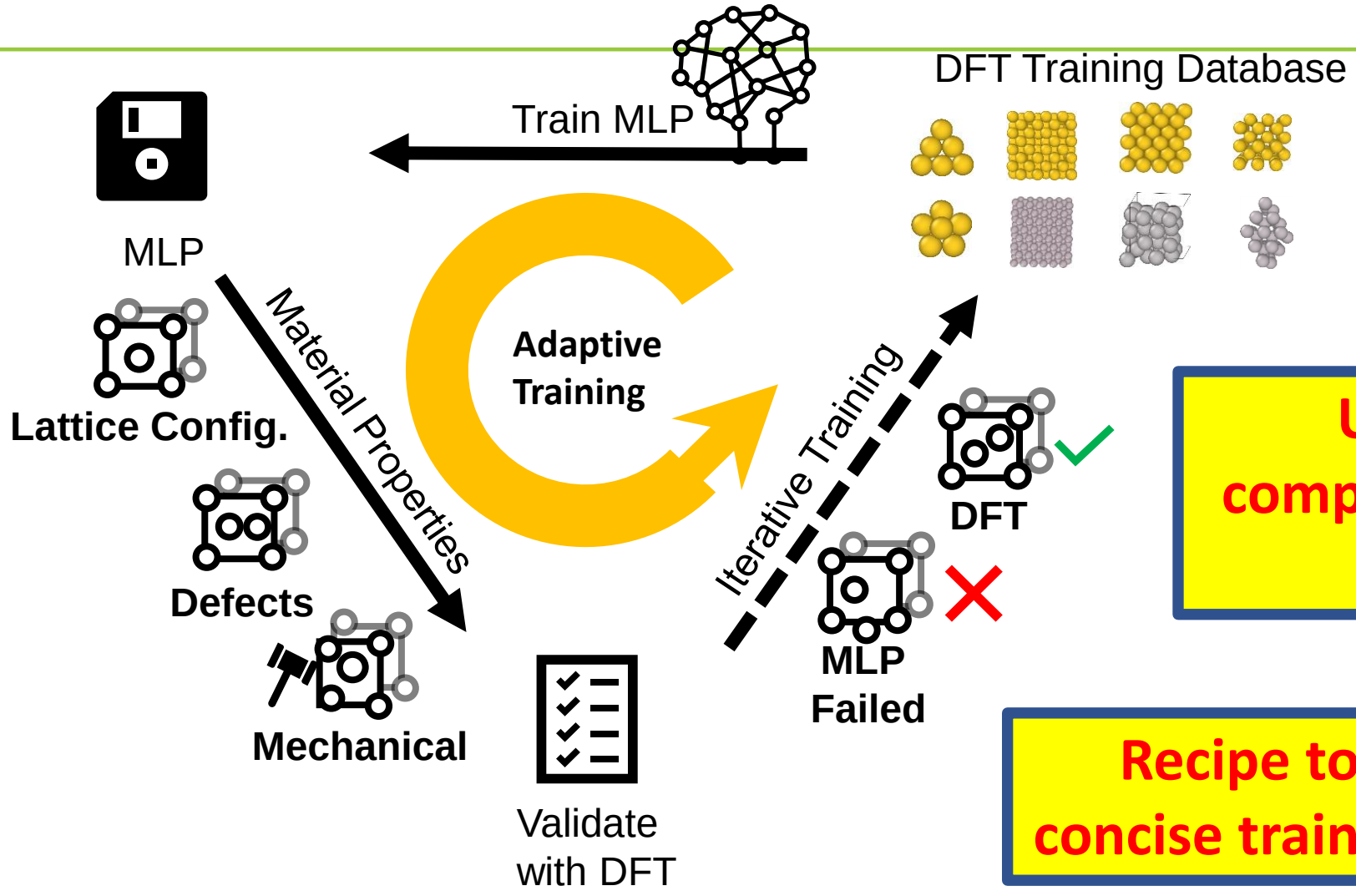
Wagih-Schuh Isotherm (Modified for Surfaces)

$$\Delta E^{\omega}(X_{surf}) = \begin{cases} 0 & X_{surf} \le X_0 \\ \omega(X_{surf} - X_0) & X_{surf} > X_0 \end{cases} \qquad X_{tot} = (1 - f_{surf})\, X_{bulk} + f_{surf}(A)^{-1} \qquad A = \left[1 + \frac{1 - X_{bulk}}{X_{bulk}} \cdot \exp\left(\frac{\Delta \bar{E}_{seg} + \Delta E^{\omega}}{kT}\right)\right]^{-1}$$

MLP results match most precisely with Wagih-Schuh Isotherm

C. Andolina, J. Wright, N. Das, and W. A. Saidi, *Phys. Rev. Mater.* 2021, *5*, 083804.

MLP Workflow: Training and Validation

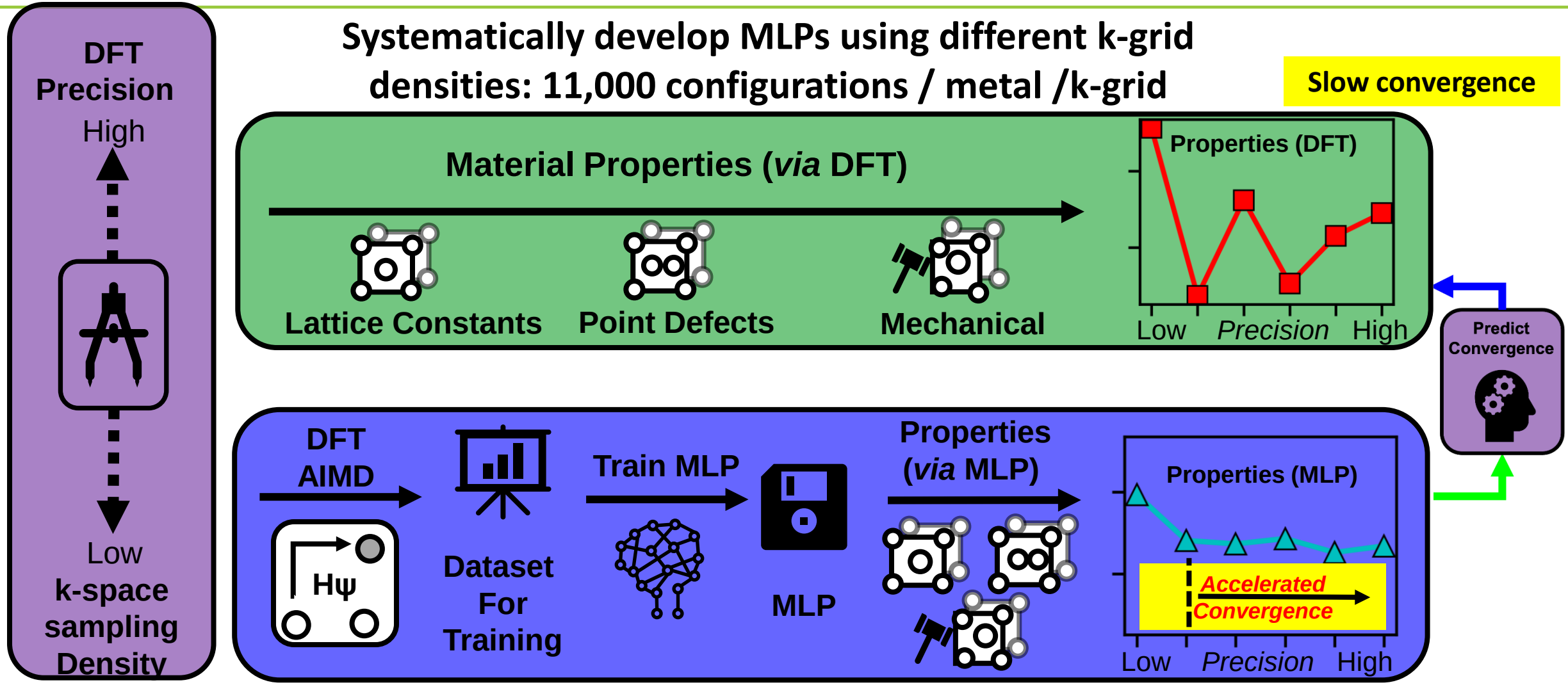


Thousands of configurations ....

Utilize low-accuracy computational framework to devleop MLP

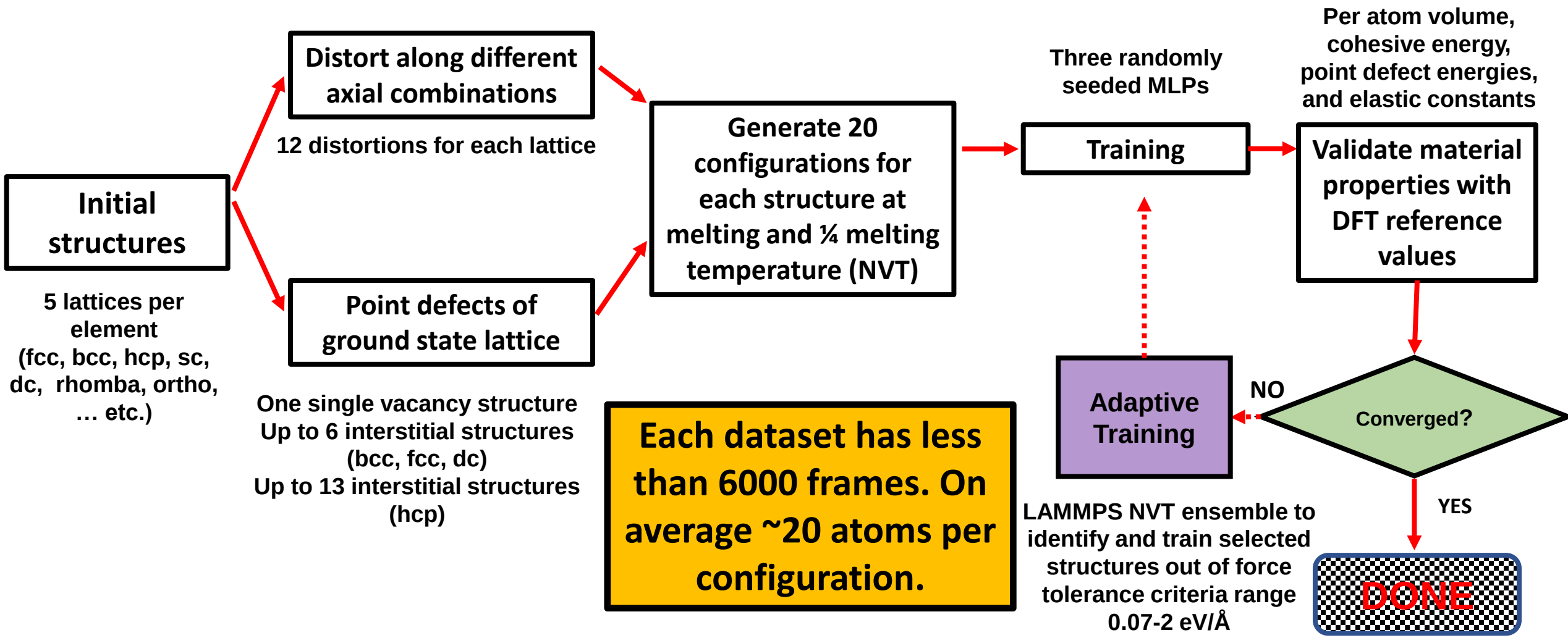
Recipe to generate concise training database

Computational Experiments on Al, Cu and Mg



D. Bayerl, C. Andolina, S. Dwarknath and W. A. Saidi, Digital Discovery, 1, 61-69 (2022)

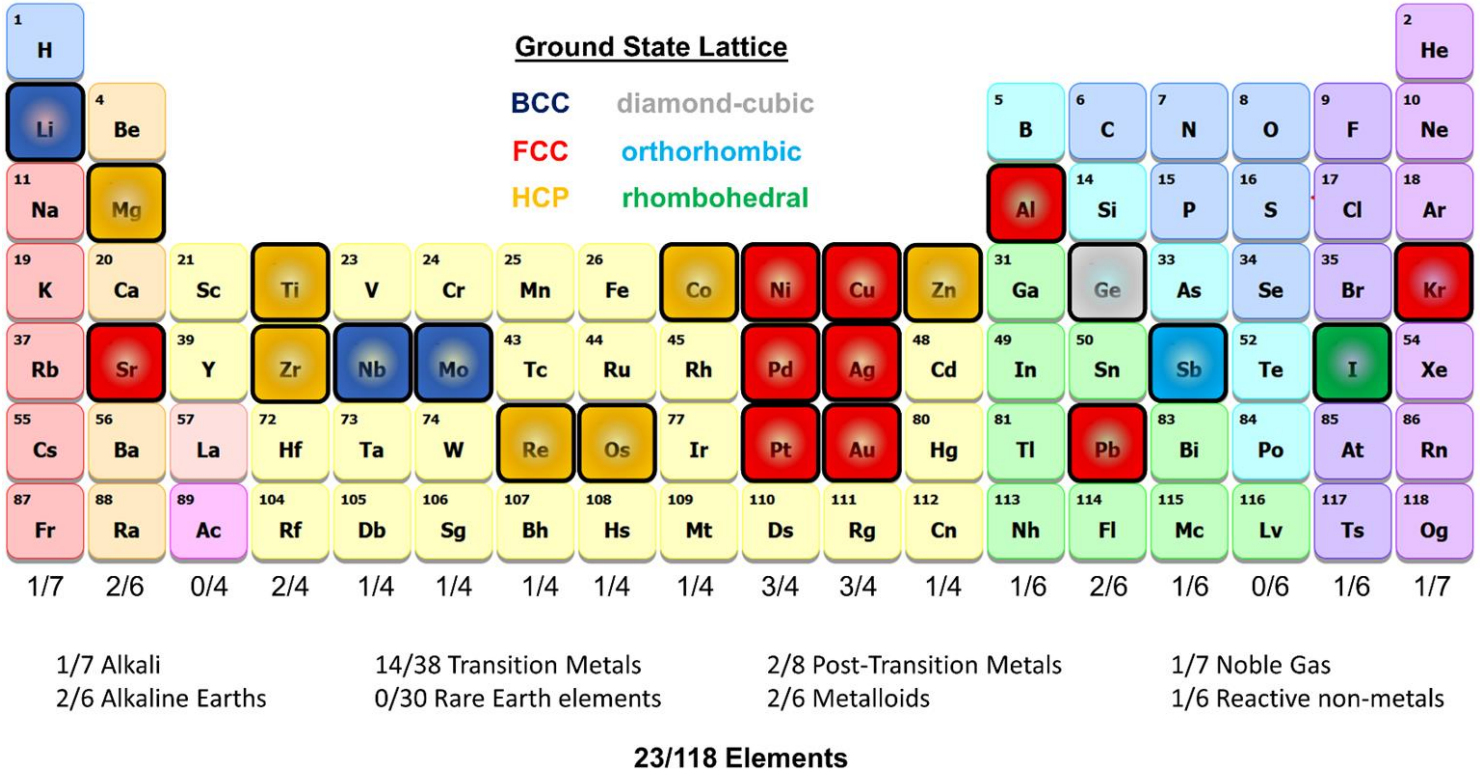
Recipe for concise dataset….

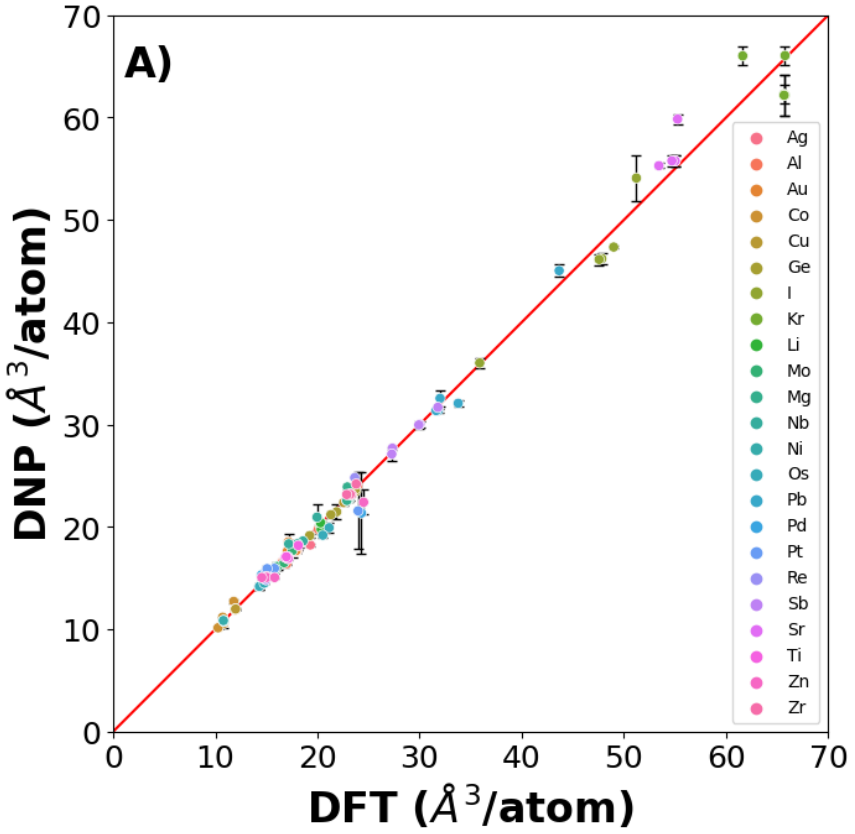


***No Pristine Lattices used in training ***

21

Single Element MLPs





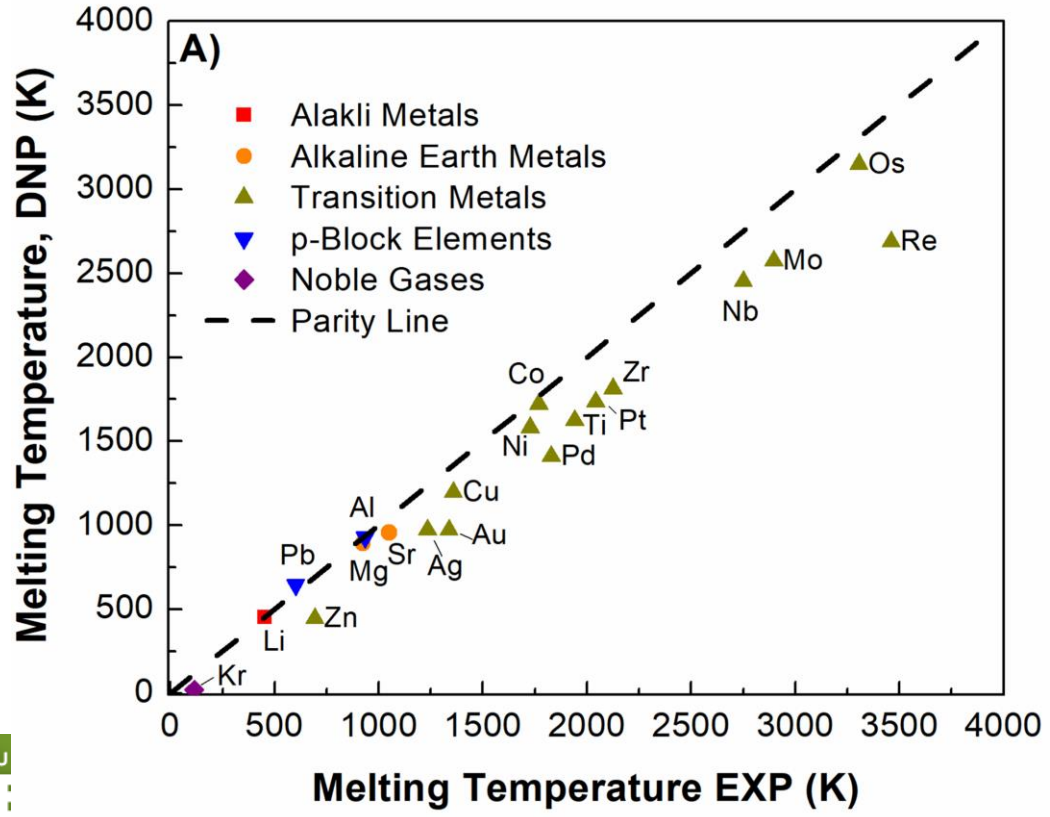
Good predictive accuracy of benchmark material properties using small datasets for 23 elements

C. M. Andolina and W. A. Saidi, *Digit. Disc.* ***2***, *1070, 2023*

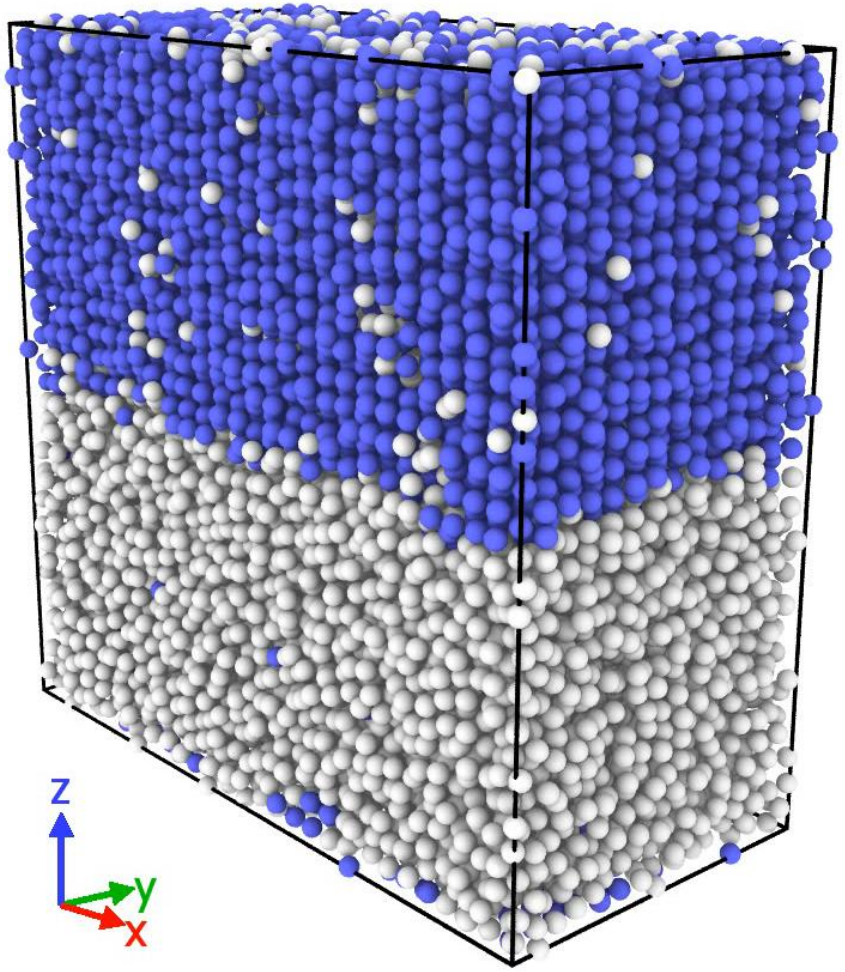
Understanding Material Melting

- **Two-Phase Coexistence (TPC) approach is the "gold-standard" ….. but requires system sizes in excess of 10,000 atoms!**

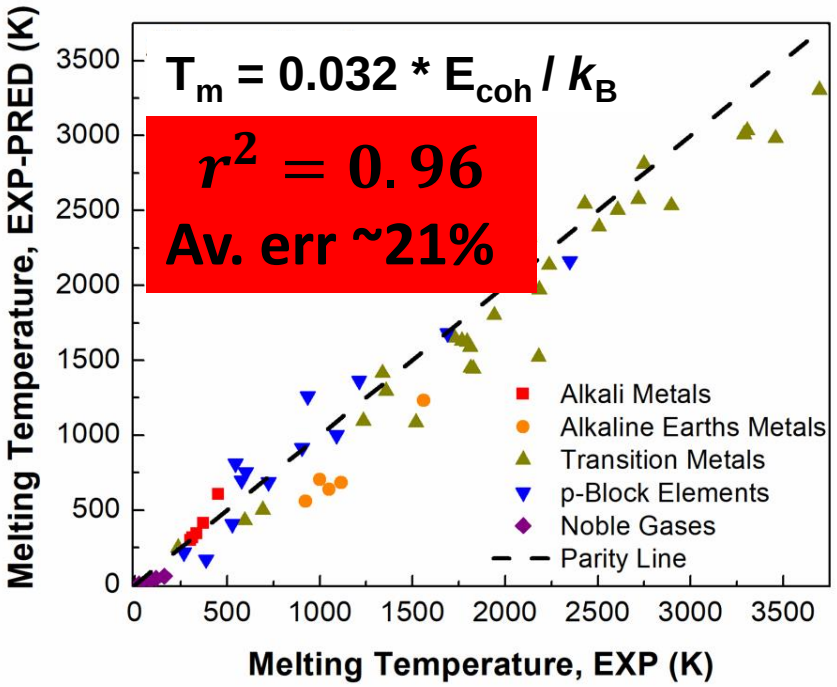
Applied MLP to compute T_m of 20 elements



Mo
BCC



Estimate T_m from Cohesive Energy



Using group-specific trends

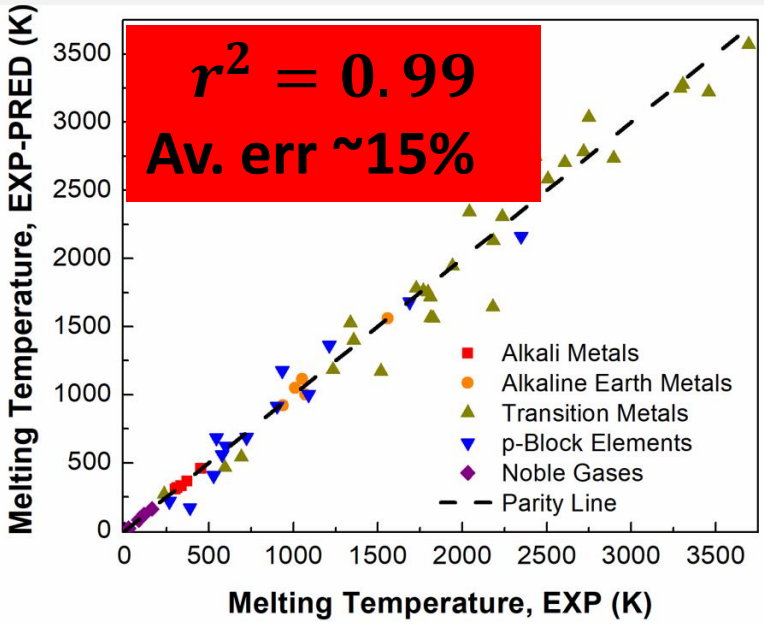
T_m = (340 K/eV * E_{coh}) + 430 K (Alkali Metals)

T_m = (181 K/eV * E_{coh}) + 162 K (Alkaline Earths)

T_m = 401 K/eV * E_{coh} (Transition Metals)

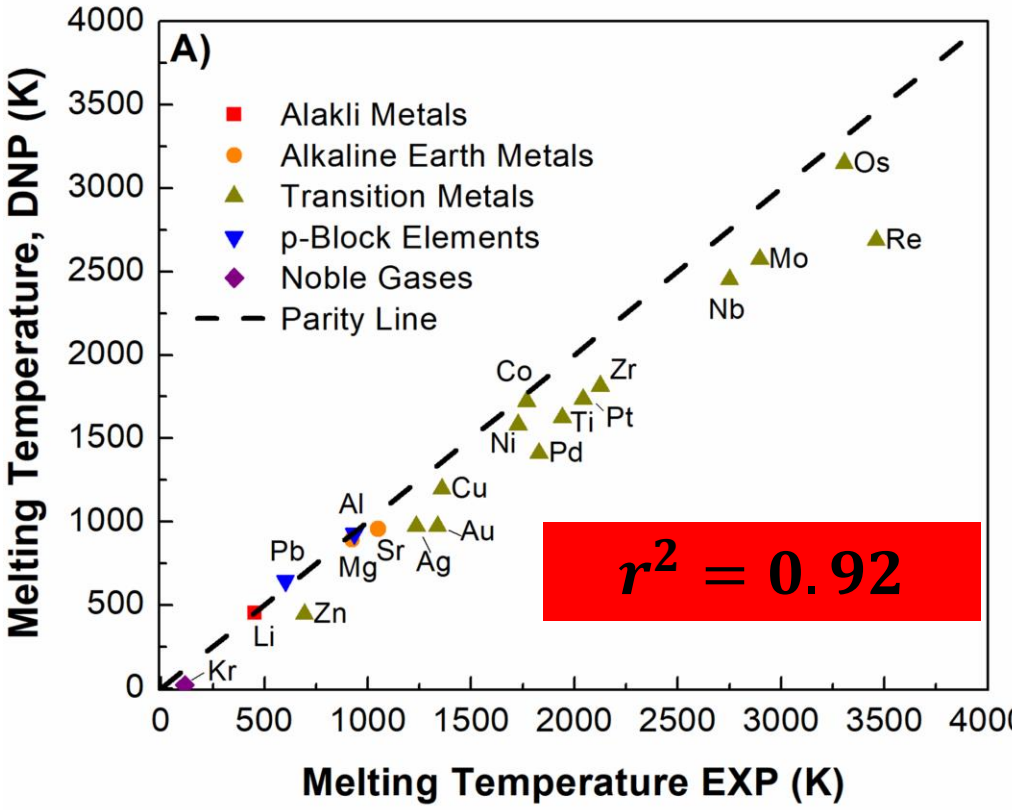
T_m = (407 K/eV * E_{coh}) – 204 K (p-block)

T_m = 1012 K/eV * E_{coh} (Noble Gases)



The "Universal" equation can be improved by sorting elements by periodic group

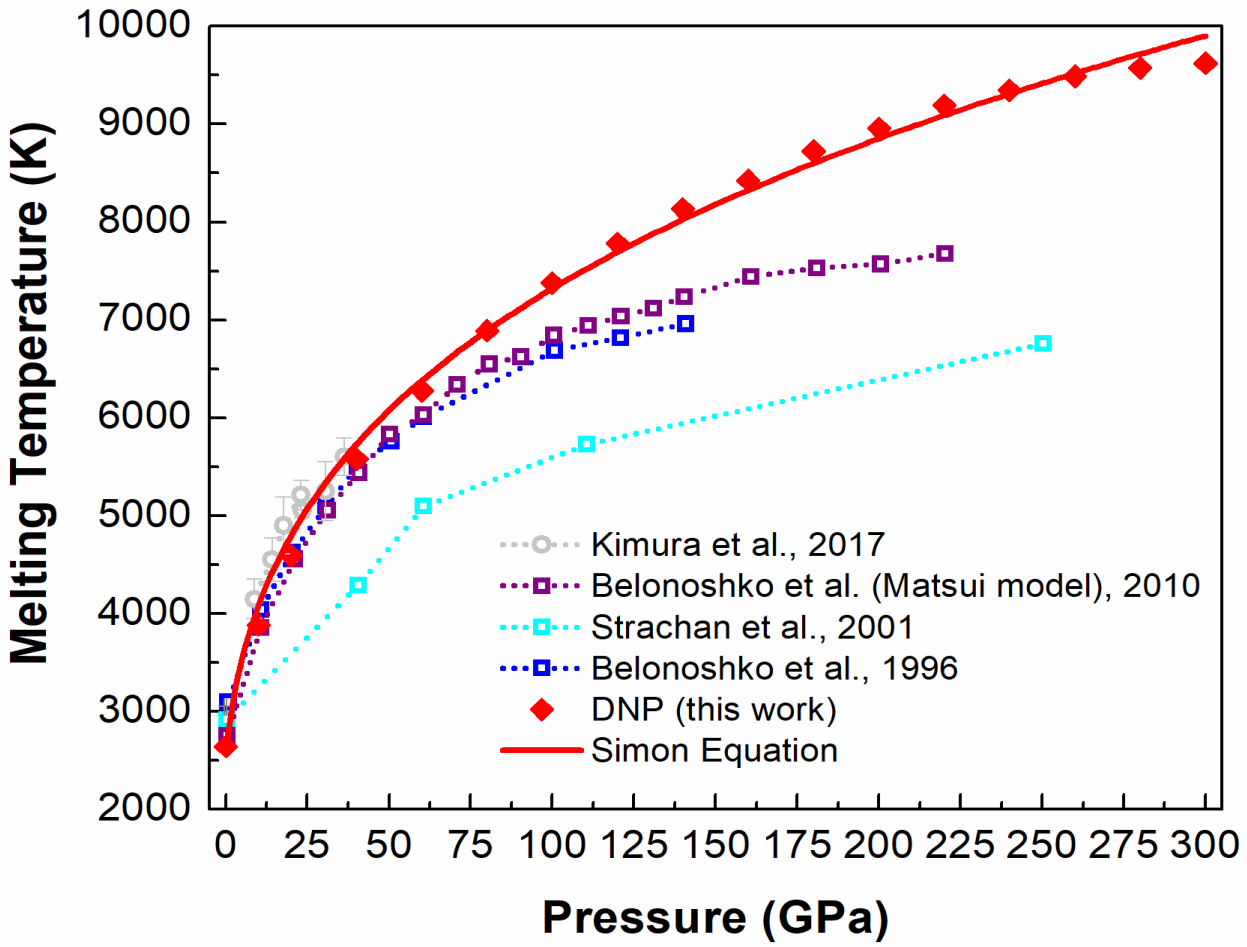
Comparing TPC-MLP to Experiment and DFT-Predicted



DFT/MLP underestimates experimental T_m

Improved agreement MLP and DFT-predicted.

MgO Solid-Liquid Phase Transitions at Mantle Conditions



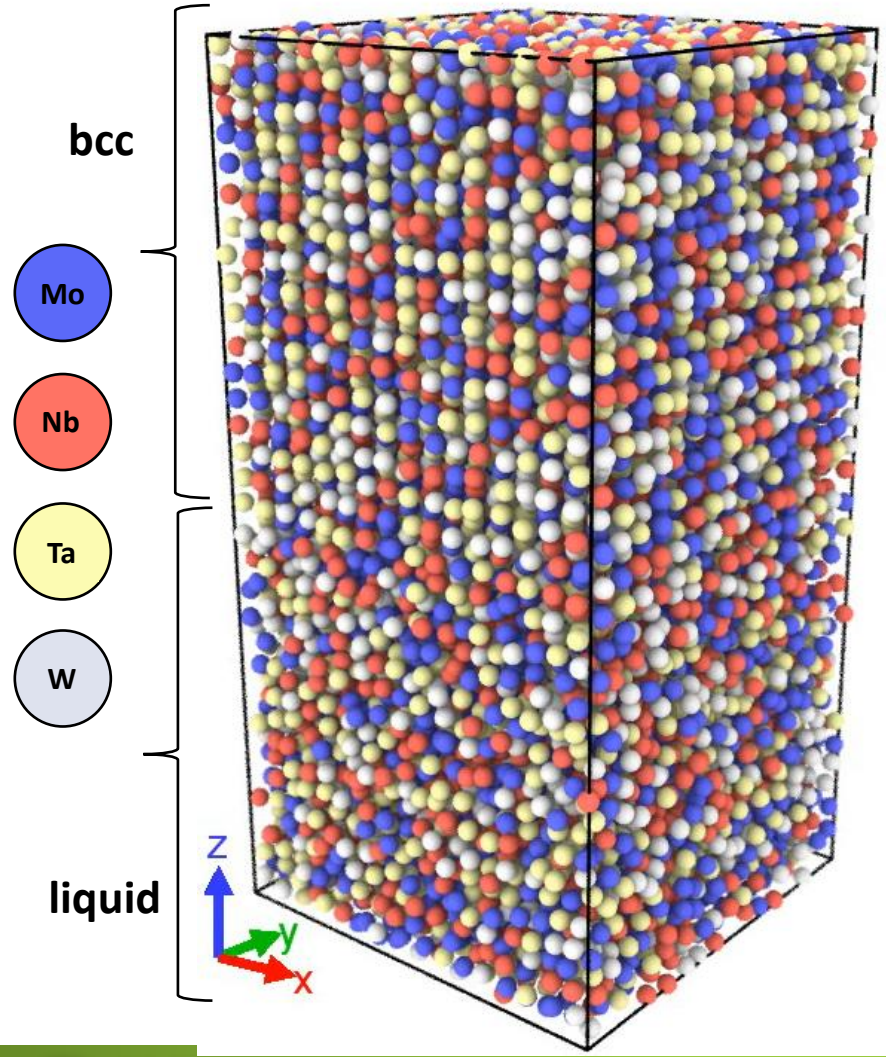
We can describe solid-liquid phase changes for large supercells over long time scales and extreme pressures ~300 GPa with DFT accuracy

P. Wisesa, C. M. Andolina, and W. A. Saidi, *J. Phys. Chem. Lett.* **14**, 468, 2023

Preliminary Results: MoNbTaW HEA

NATIONAL ENERGY TECHNOLOGY LABORATORY

MD MLP-TPC

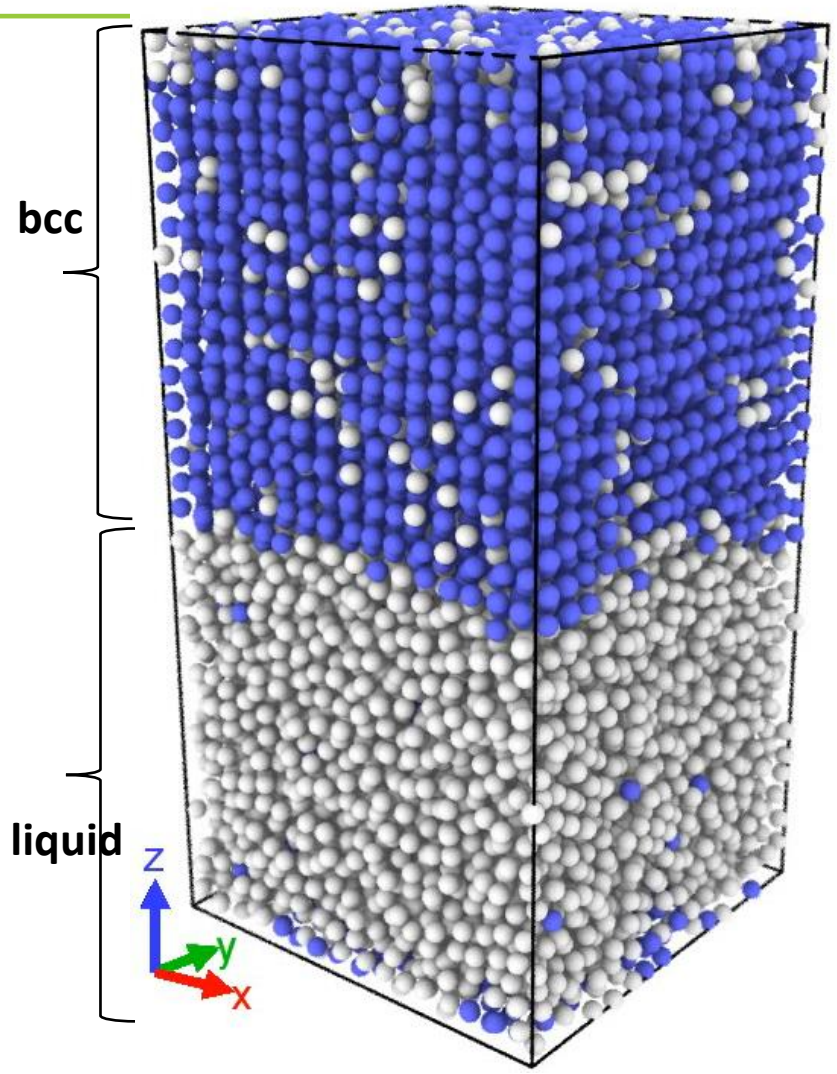


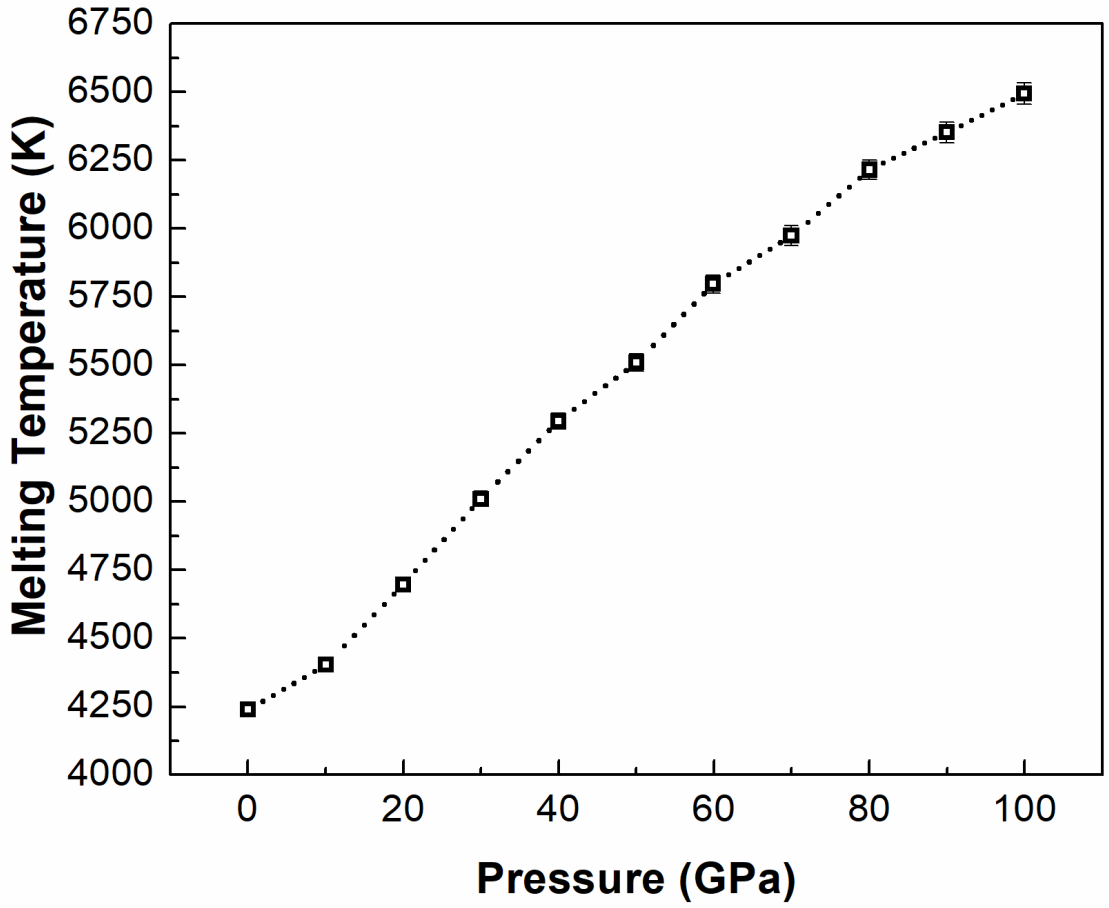
MoNbTaW Composition	E_{coh} (eV)	T_m-PRED (K)	T_m TPC-MLP (K)
Mo256.Nb256.Ta256.W256-r1	-10.211	4100	4216±28
Mo256.Nb256.Ta256.W256-r2	-10.210	4099	4221±21
Mo256.Nb256.Ta256.W256-r3	-10.208	4099	4217±22
Mo256.Nb256.Ta256.W256-r4	-10.210	4100	4220±21
Mo128.Nb384.Ta128.W384	-10.153	4077	4267±22
Mo128.Nb128.Ta384.W384	-9.837	3950	4251±20
Mo128.Nb384.Ta384.W128	-10.229	4107	4241±22
Mo384.Nb128.Ta128.W384	-9.477	3805	4117±23
Mo384.Nb128.Ta384.W128	-10.154	4077	4320±19
Mo384.Nb384.Ta128.W128	-9.622	3864	4215±28

- **Similar T_m over narrow compositional changes.**
- Predicted Tm from E_{coh} agrees well with MLP-TPC T_m

Preliminary Results: MoNbTaW HEA

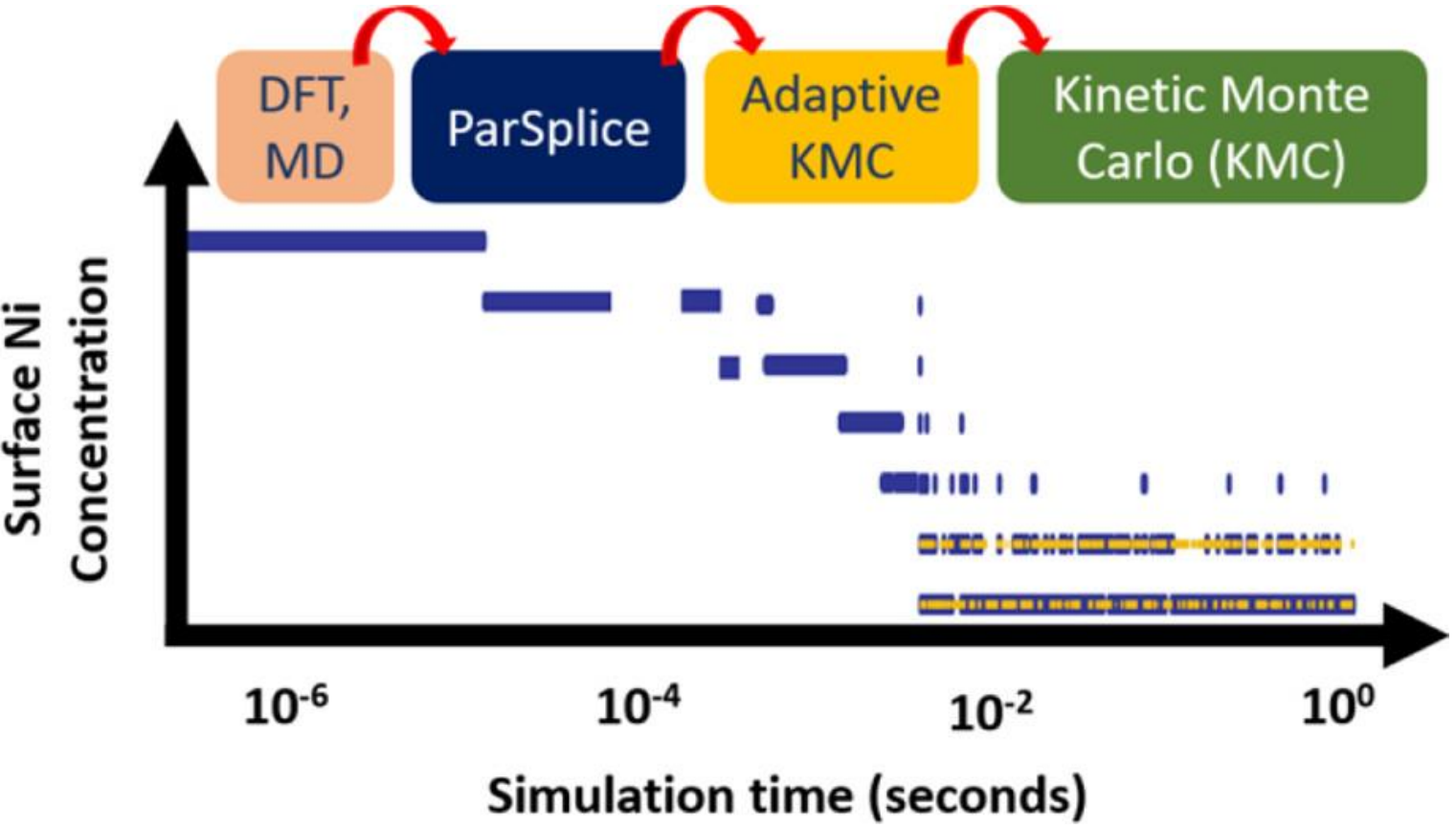
MD MLP-TPC





MLP describes T_m behavior well at elevated pressure

The trend for Cu-Ni Alloy Segregation



CuNi dynamics from ns → s

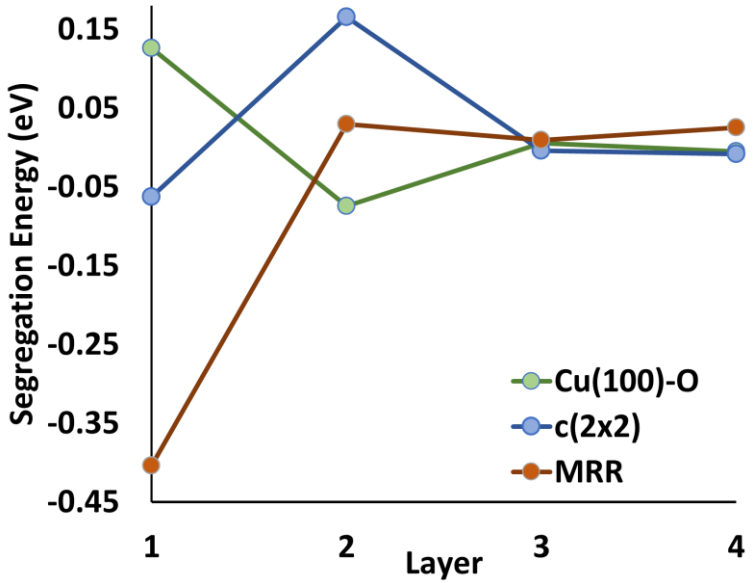
Cu segregates to the surface under reducing conditions

What happens under oxidizing conditions?

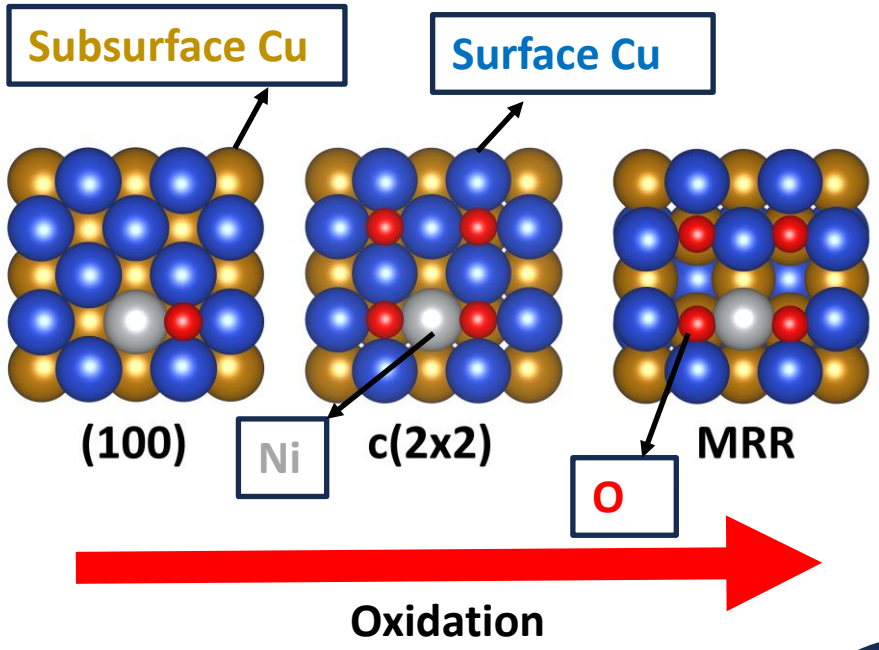
Competition between surface reconstructions and segregation tendencies

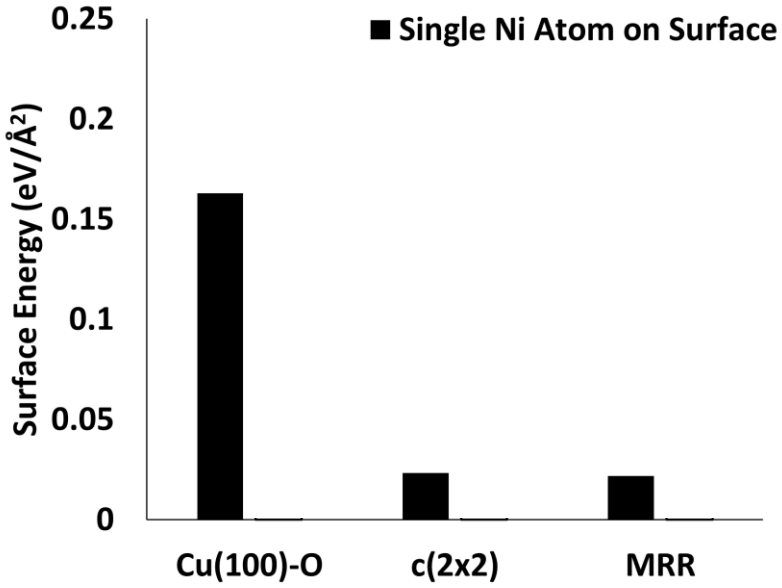
Garza, Lee, Nguyen, Garmon, Perez, Li , Yang, Henkelman, and Saidi, *JCTC* 2022, *18*, 4447

DFT segregation preferences on Cu (100) reconstructions:



←Surface | Bulk →





MRR is preferred over c(2x2) for small Ni doping and c(2x2) becomes preferred as Ni increases.

MLP to address the complexity of the system

Wisesa, Li, Curnan, Han, Yang, and Saidi, *arxiv.org 2308.11867*

Segregation at finite time – MC/MD Simulation

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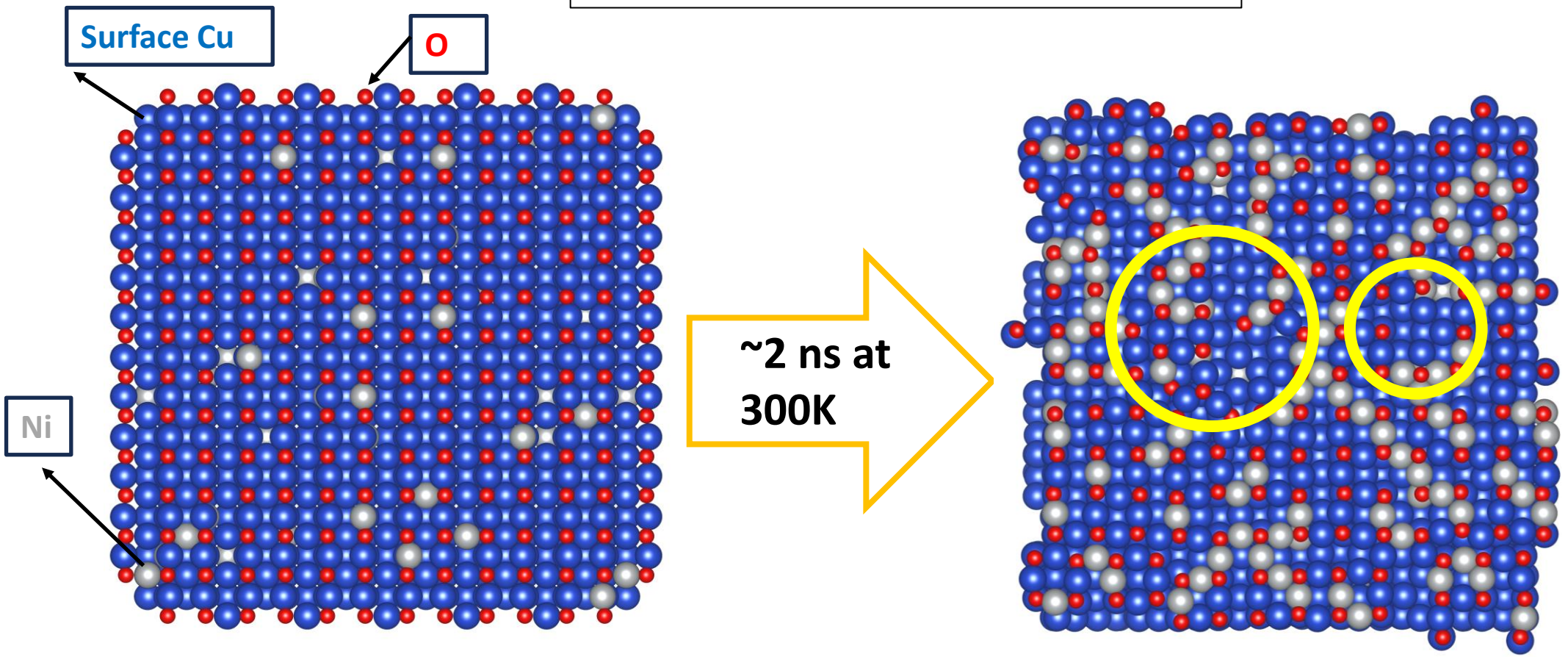
MRR with random 5% Ni

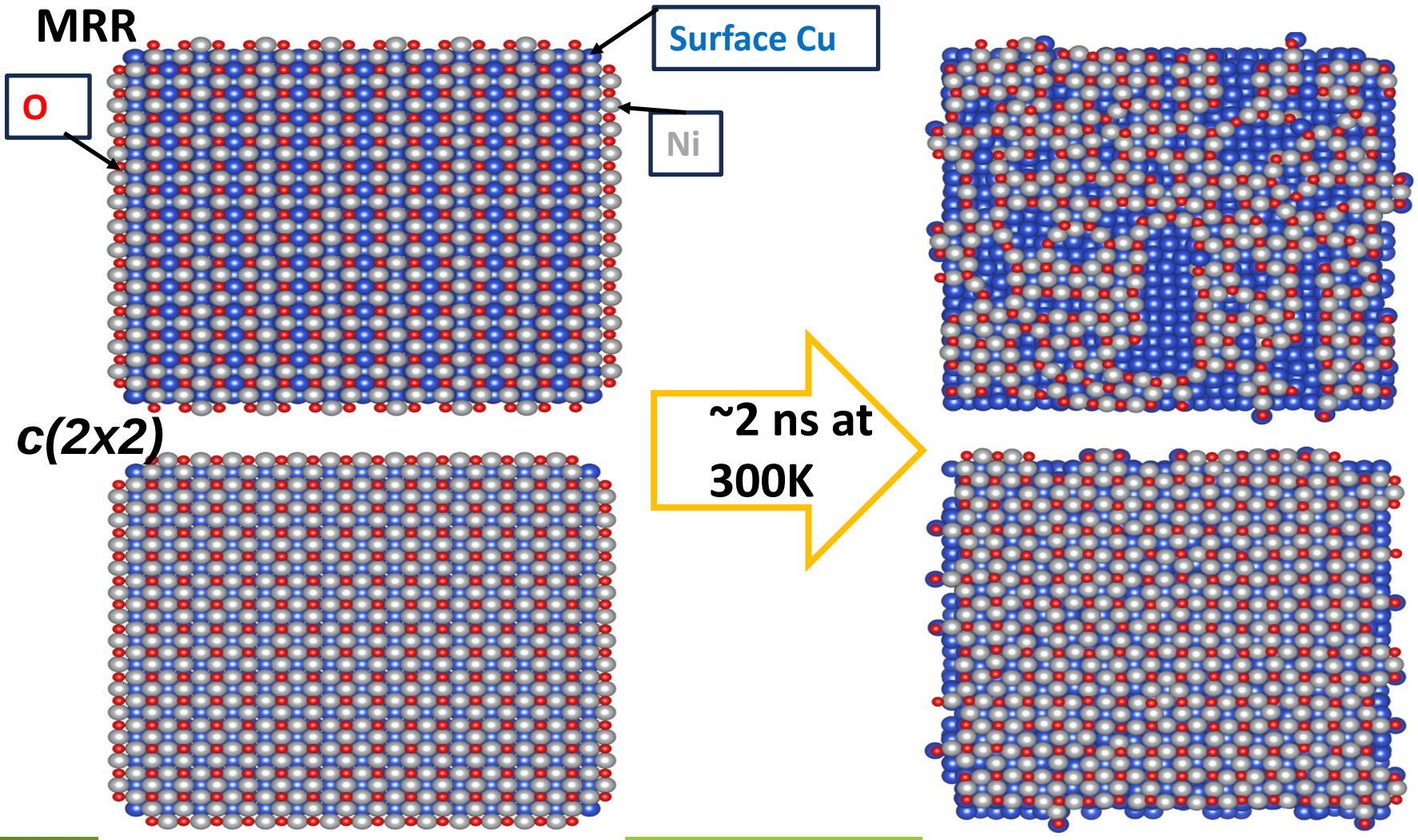


Ni segregation and surface rearrangement to form Ni-O bonds

Ni segregate to the surface and induce Ni-O nucleation

Simulations made possible by MLP

Stability of full Ni monolayer … MC/MD simulation

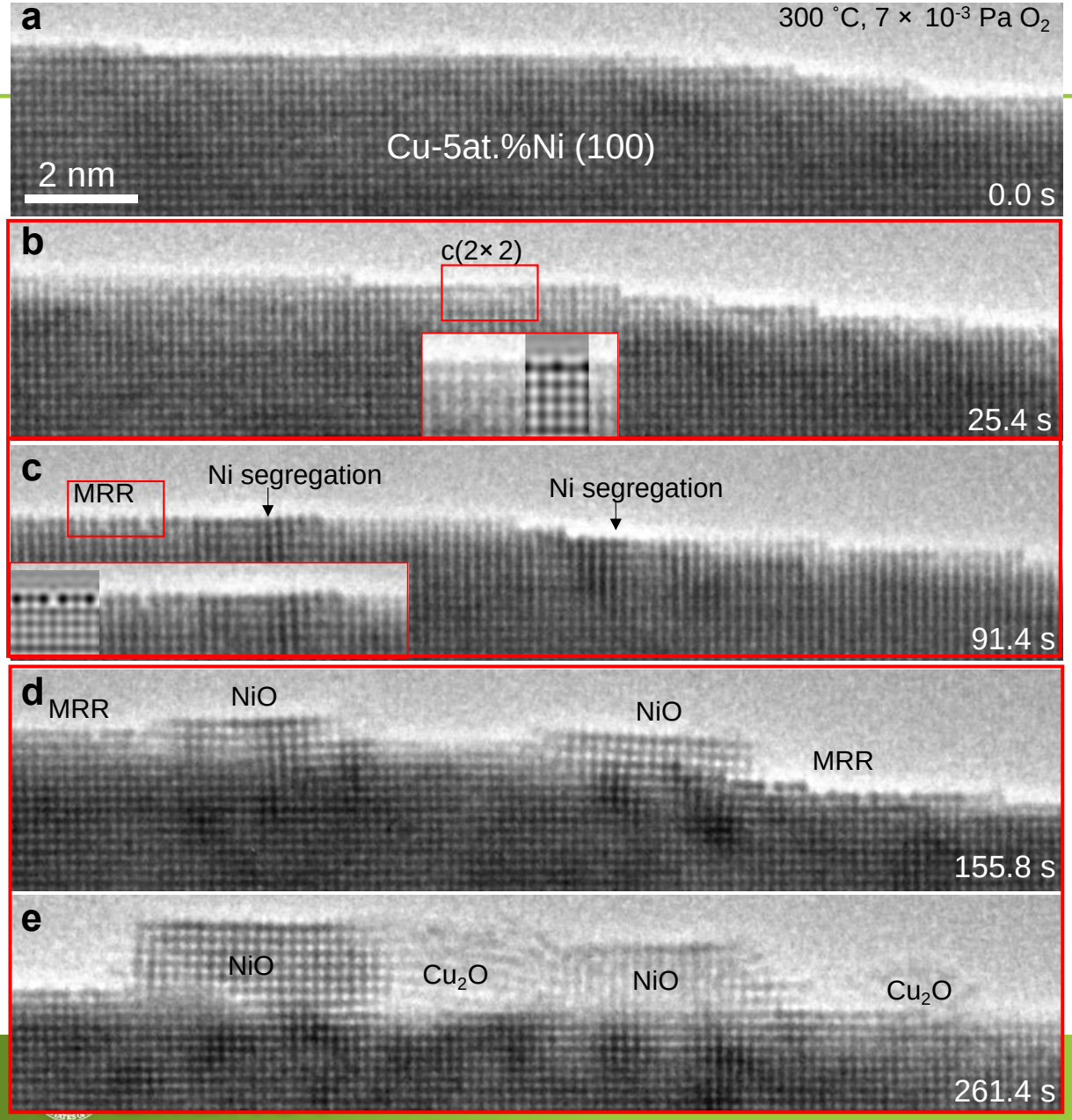


c(2x2) surface is more stable due to the NiO arrangement.

Simulations made possible by MLP

Experimental Verification

Collaboration with Li and Yang BNL



ETEM 5% Ni (300 °C, $7x10^{-3}$ Pa O2)

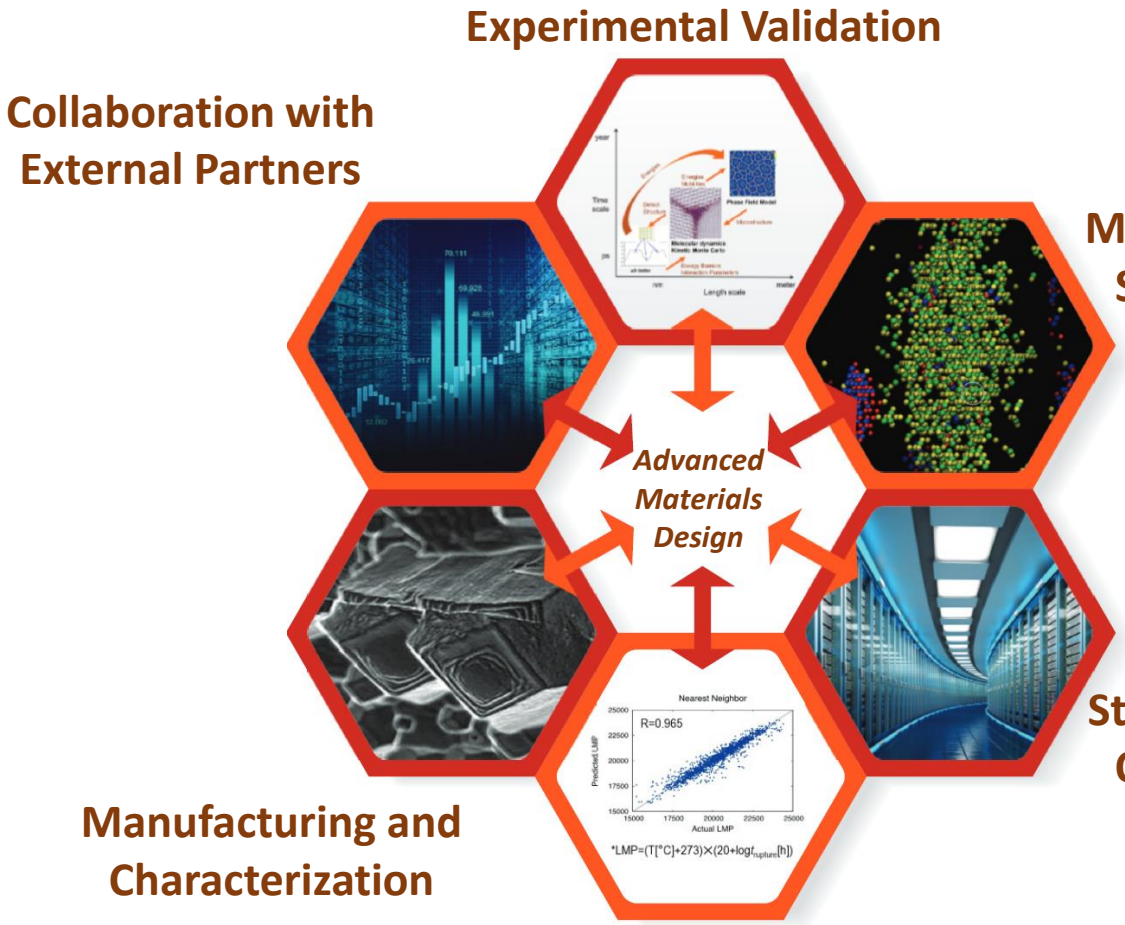
- **(b) shows the formation of c(2x2)**
- **(c) Ni segregates to the c(2x2), as highlighted by strain due to the difference between the lattice constant of Cu and Ni.**
- **At the same time MRR is forming from a region free of Ni.**
- **The c(2x2) with Ni eventually forms NiO, while the MRR forms Cu_2O.**

DFT and MLP results are verified by experimental observation.

Accelerating Material Design for Energy Applications

Integrating experimental, simulation and data-driven methods for material development

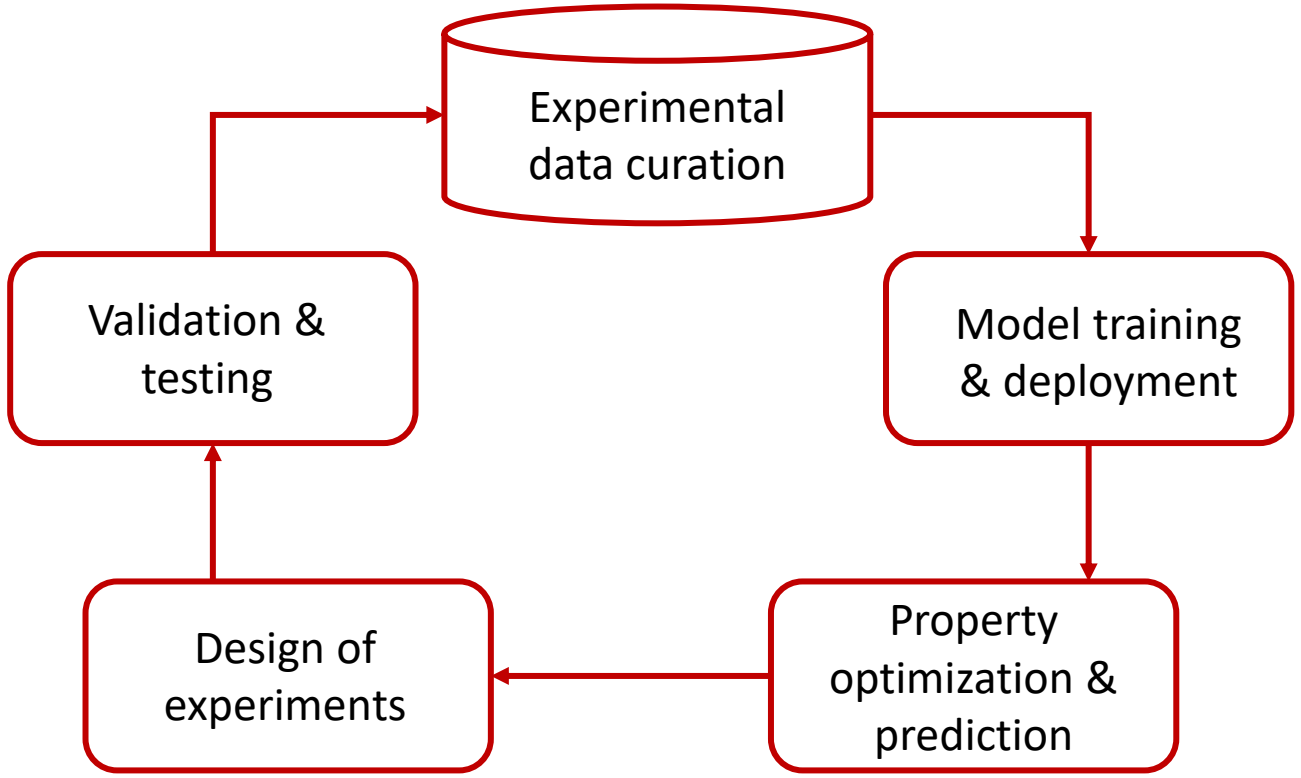
- **Accelerate development** and **deployment** of new materials for energy applications
- **Predict** long-term materials performance in fossil energy power plants of any cycle
- **Advance technologies** including High entropy alloys; structural alloys; hydrogen embrittlement; oxidation; catalysts; sorbents



Integration into materials design & testing

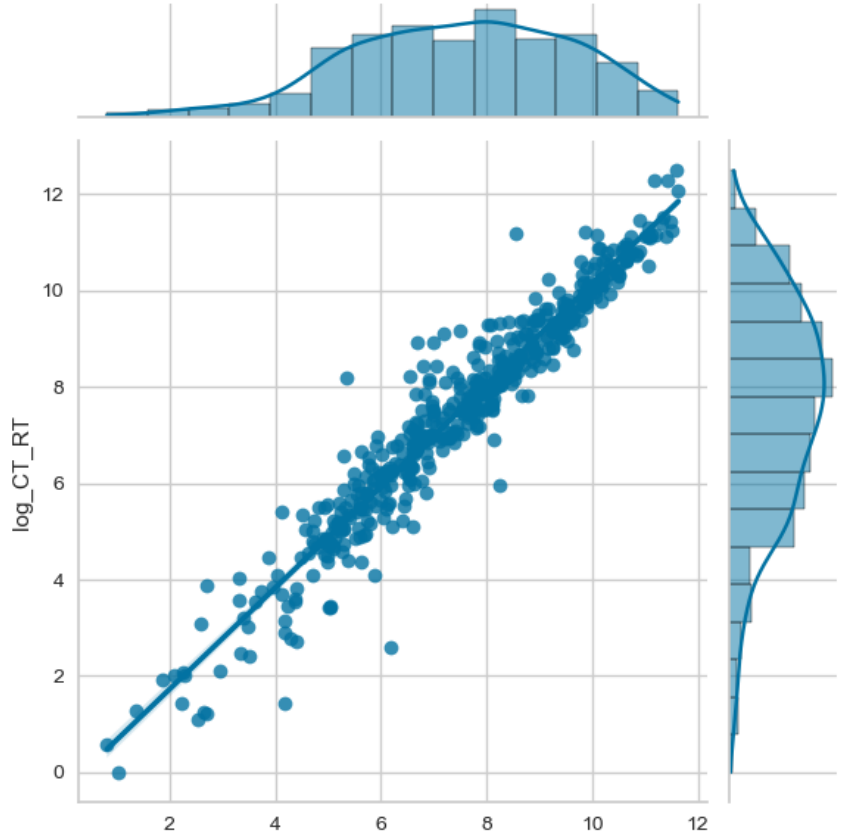
Creep Rupture Time Prediction

- Leveraging property prediction in material design loop
- Improving efficiency of experimentation through prediction of long-term properties
- Optimization of material performance



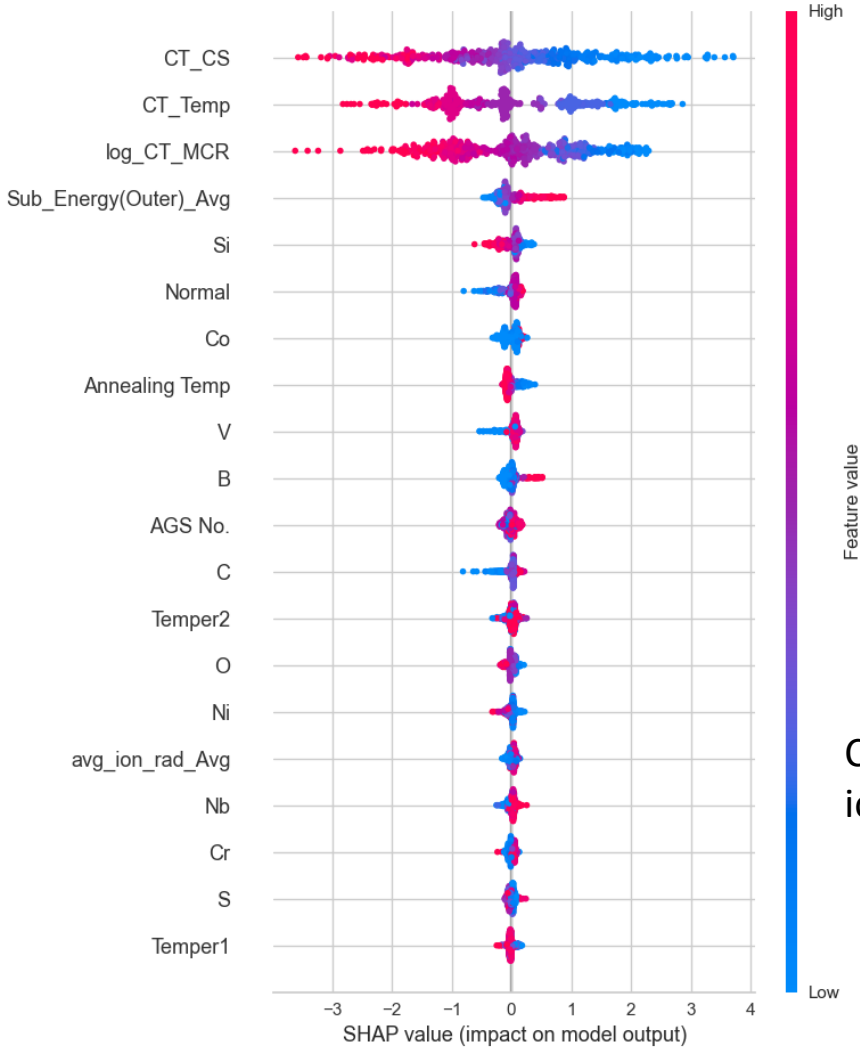
Creep rupture time prediction with physical features

CatBoost Regression of log Creep Rupture Time



Train R^2: 0.9185 +/- 0.0051

Hold-out test R^2 : 0.9282



Rank order value of outermost subshell energy level

Calculated average ionic radius

Addition of physical features

Aiding generalizability, interpretation

- Derived from Rule of Mixtures using at.% composition
- Adding physically relevant features related to mechanical behavior of alloys
  - Atomistic & physical properties
- Calculating 76 average and standard deviation values

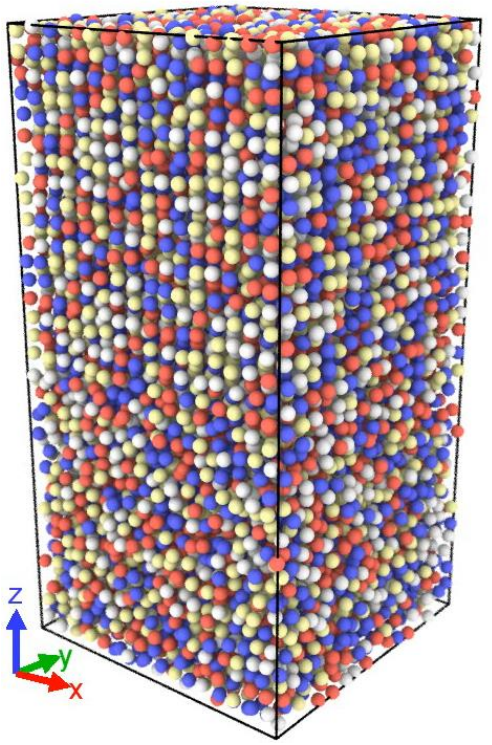
Physical Properties
Coefficient of linear thermal expansion (/K)
Electrical conductivity (106/cm Ω)
Thermal conductivity (W/cmK)
Density (g/cc @ 300K)
Bulk modulus (Gpa)
Rigidity Modulus (Gpa)
Youngs Modulus (Gpa)
Enthalpy of atomization (kJ/mole @ 25°C)
Enthalpy of fusion (kJ/mole)
Enthalpy of Vaporization (kJ/mole)
Brinell hardness (MN m^-2)
…

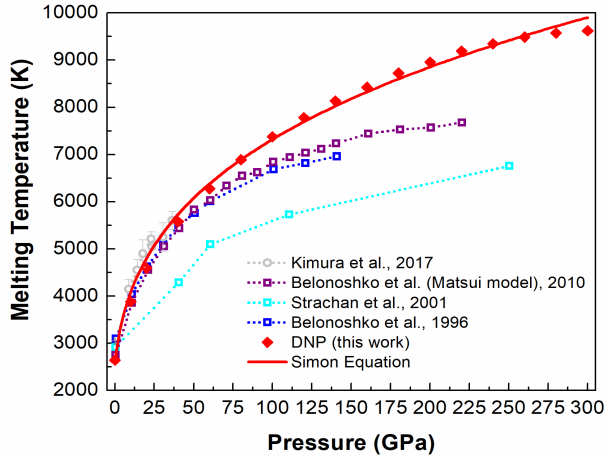
Atomistic Properties
Number of electron vacancies
Number of valence electrons
Number of electrons in outermost subshell
Electron capacity of outermost subshell
Electron Vacancies in the outermost subshell
Rank order value of outermost subshell energy level
Number of electrons in 2nd outermost subshell
Electron capacity of 2nd outermost subshell
Electrochemical Equivalent (g/amp-hr)
Electron work function (V)
Pauling electronegativity
…

Trehern, W., Ortiz-Ayala, R., Atli, K.C., Arroyave, R. and Karaman, I., 2022. Data-driven shape memory alloy discovery using artificial intelligence materials selection (AIMS) framework. *Acta Materialia*, *228*, p.117751. https://doi.org/10.1016/j.actamat.2022.117751

Summary

- **MLPs provide a good balance of accuracy and computational cost, and allows bridging the modeling gap with experiment**
- **Various examples including melting and early stages of oxidation of a binary alloy**
- **Creep rapture time prediction using experimental data and simple features**





Database and MLPs are available
https://github.com/saidigroup

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