Machine Learning Vacancy Formation Energy in Nickel-Based Superalloys

Aditya Sundar

NETL Support Contractor

2025 MRS Spring Meeting & Exhibit

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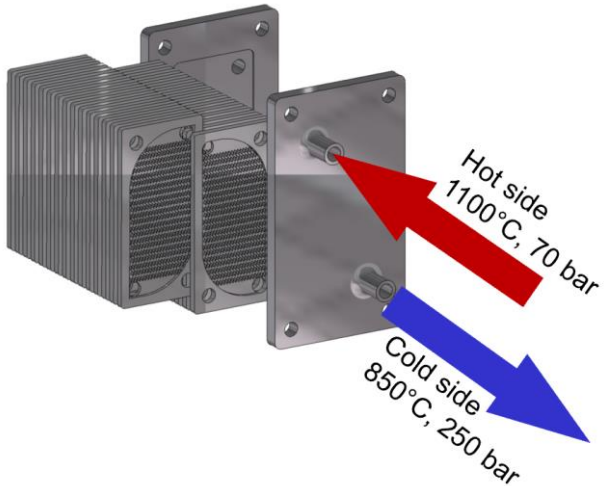
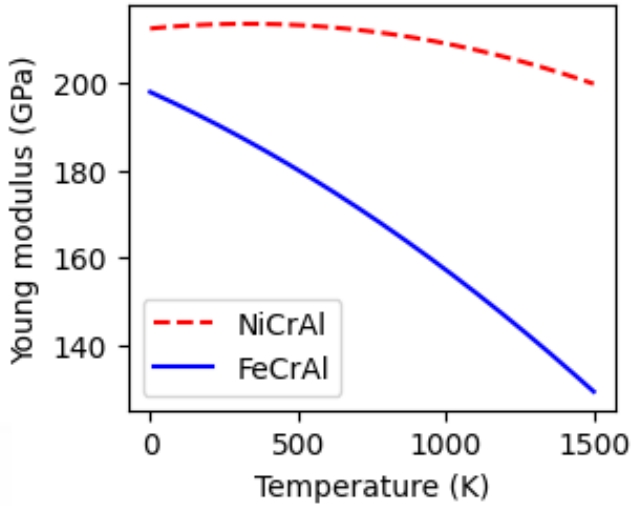
Introduction: Ni Superalloys and Applications

Turbine blades

Heat exchangers

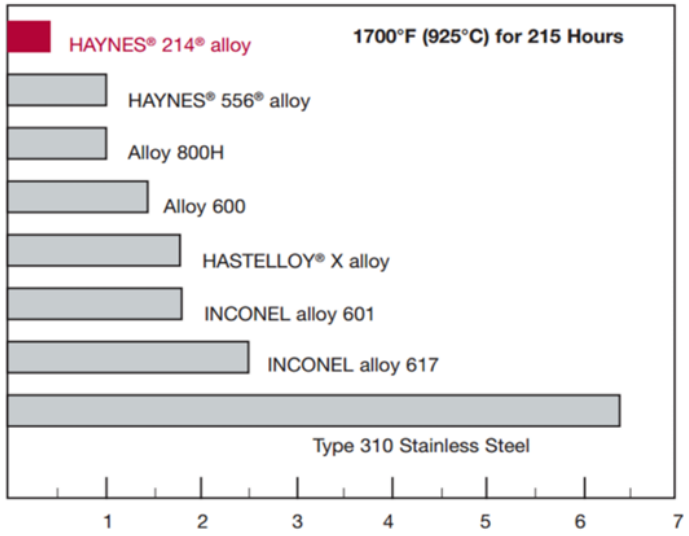


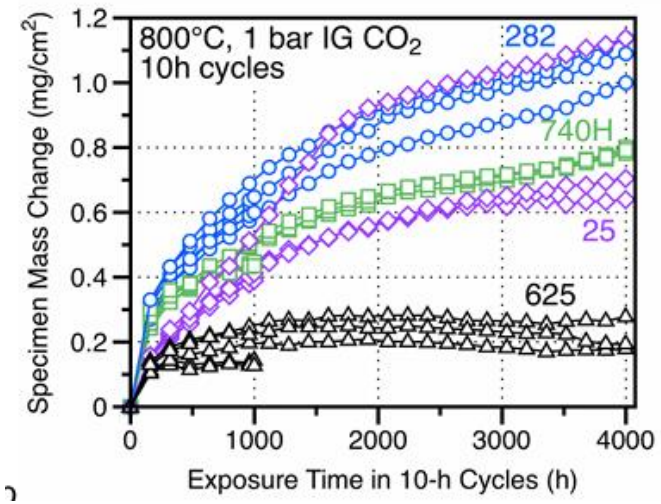
High strength



https://mooseframework.inl.gov
https://www.haynesintl.com/

Corrosion resistance

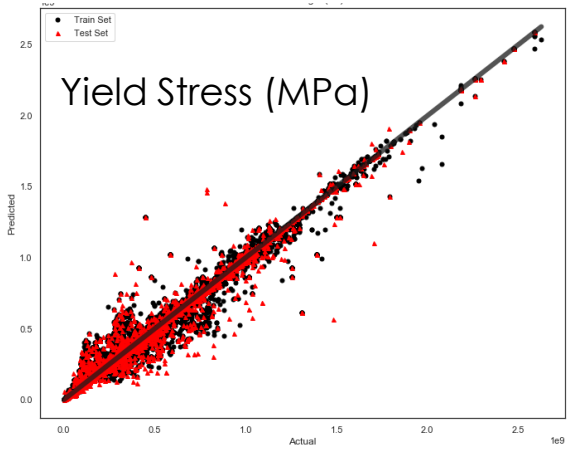


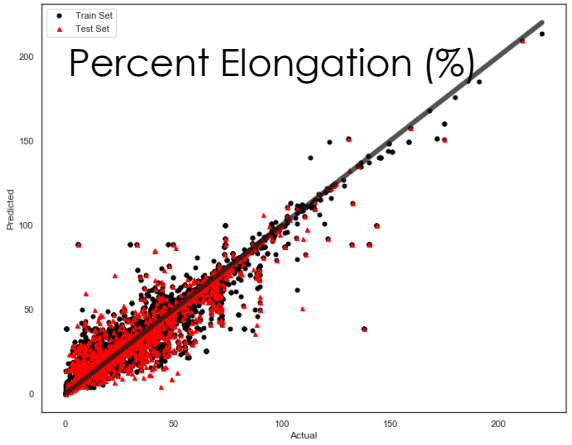


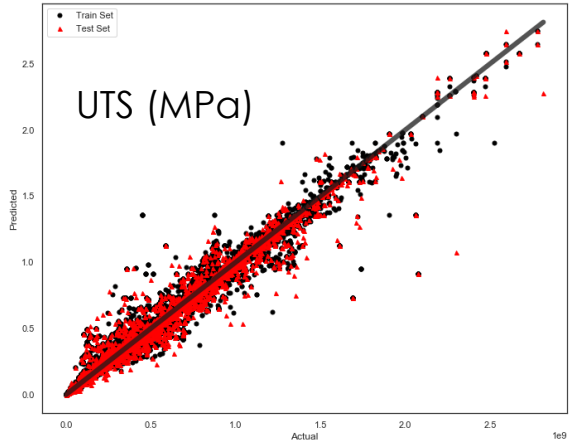
Pint and Pillai DE-EE0001556 Lifetime Model Development for sCO2 CSP (2019).

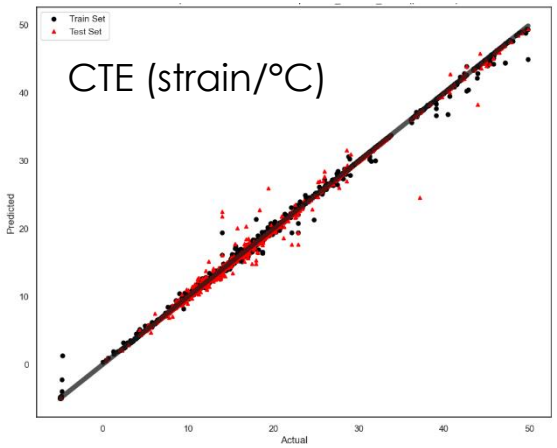
Alloy Modeling Effort at NETL

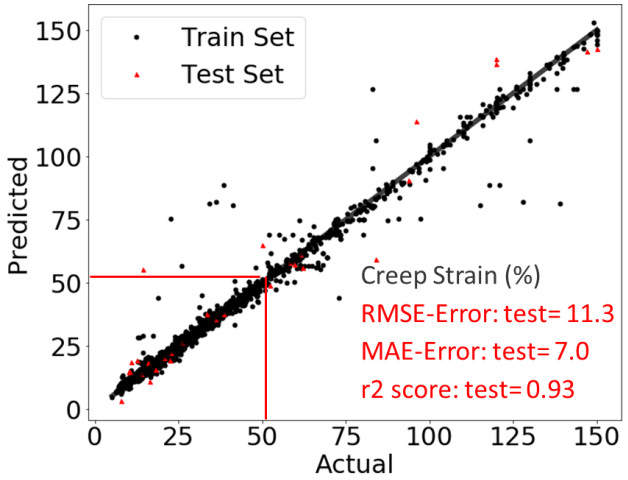
NETL's In-house Multi-Objective Machine Learning Effort

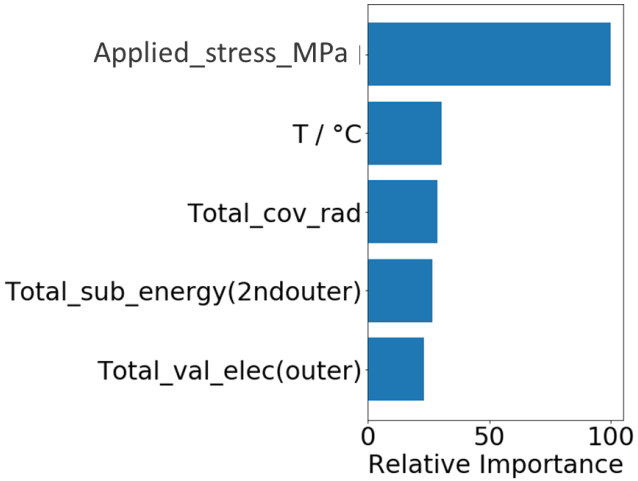






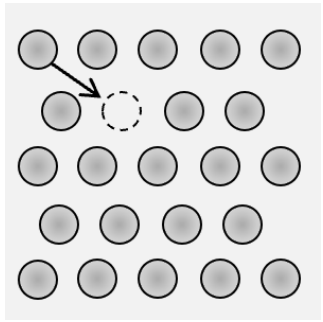


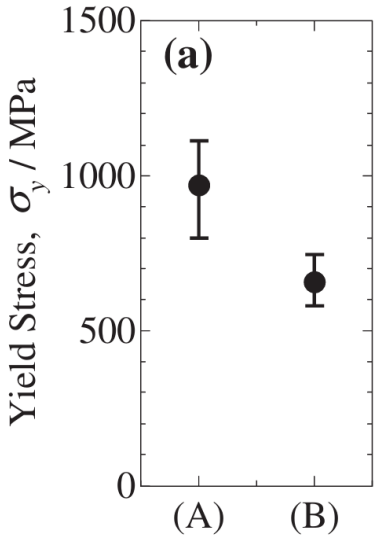


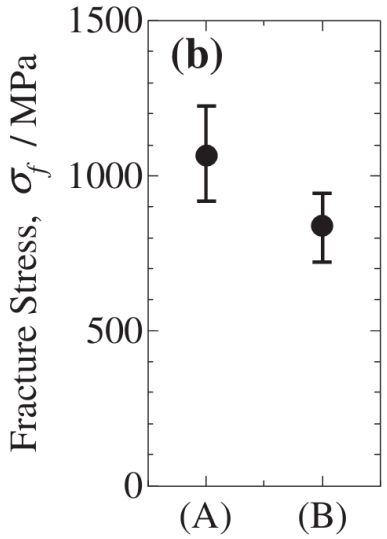


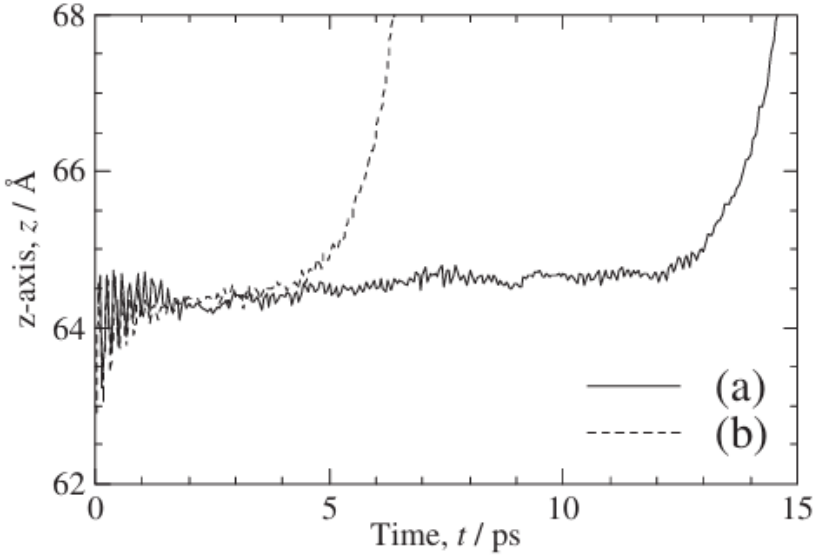
- Mechanical properties
  - **Yield strength**
  - **Ultimate tensile strength (UTS)**
  - **Tensile elongation**
  - **Creep performance**
  - **Fatigue (to be added)**
- Coefficient of thermal expansion (CTE)
- Oxidation (ongoing)
- Hydrogen embrittlement (ongoing)

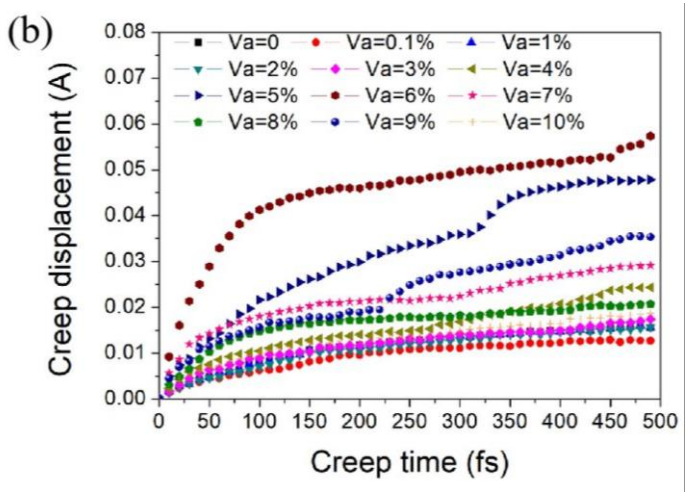
Introduction: Impact of Vacancies on Creep Properties

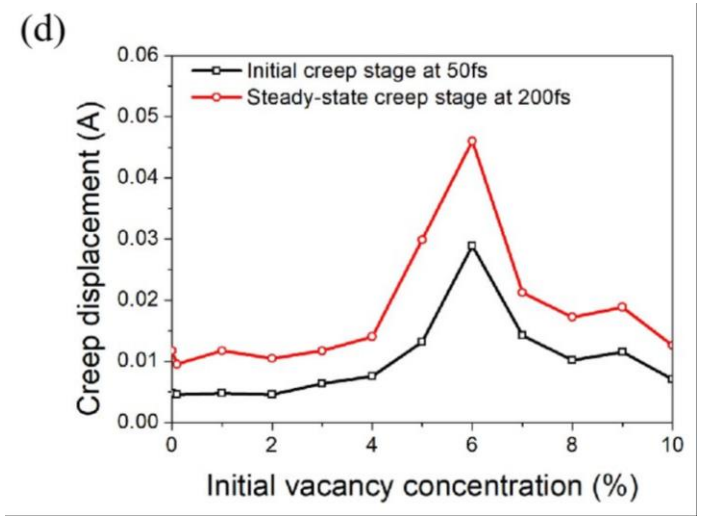












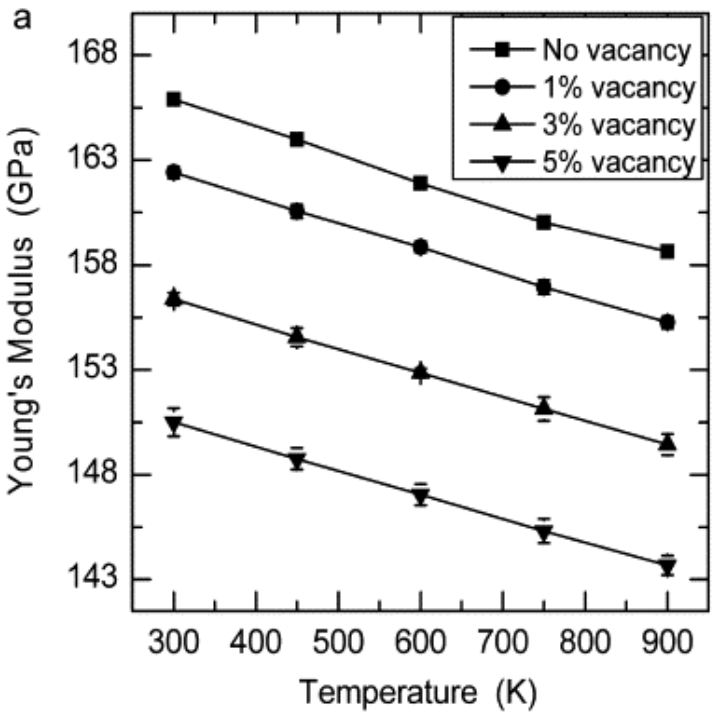
- 40% reduction in yield stress
- 30% reduction in fracture stress
- Doubling of creep rate from molecular dynamics (MD) simulations

Yuasa et al. Materials Transactions, Vol. 49, No. 10 (2008).

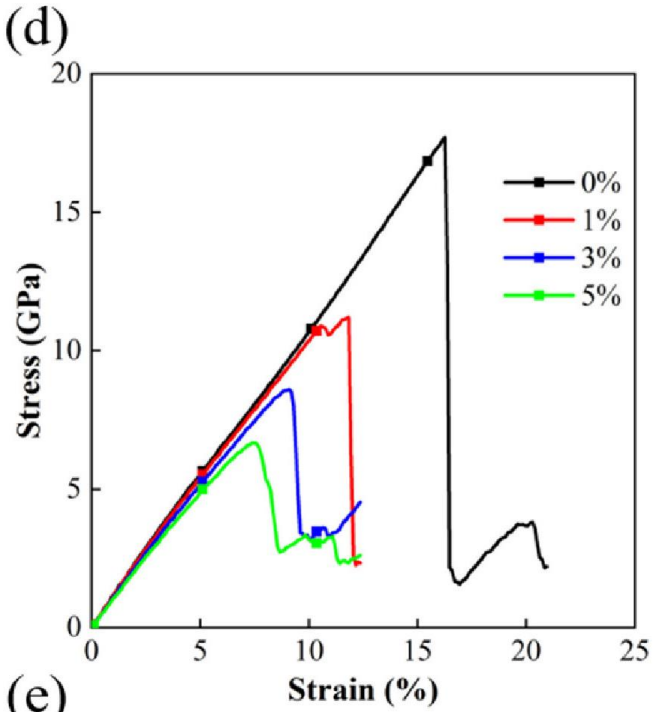
Higher creep displacements with vacancy concentration.

Cui et al. *Coatings* 2024, 14(1), 63.

Introduction: Impact of Vacancies on Strength



Yang et al. Physica B: Condensed Matter Volume 407, Issue 12, 15 June 2012, Pages 2234-2238.

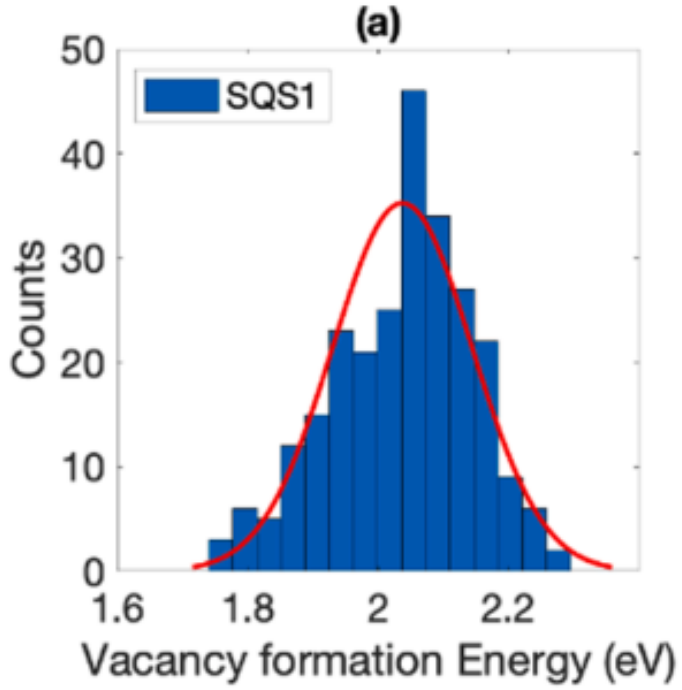


Peng et al., International Journal of Mechanical Sciences Volume 218, 15 March 2022, 107065.

Crucial to understand vacancy thermodynamics in alloys.

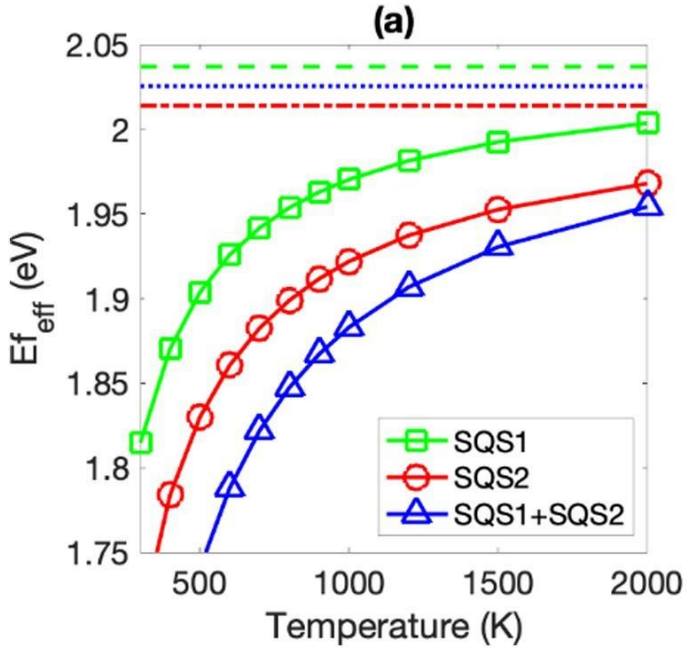
Introduction: Vacancies in Multi-Element Alloys

Distribution of Formation Energies



$$E_{\text{vac}}^{\text{eff}}(T) = -k_B T \times log\left[\int g(E_v^f) exp\left(-\frac{E_v^f}{k_B T}\right) dE_v^f\right]$$

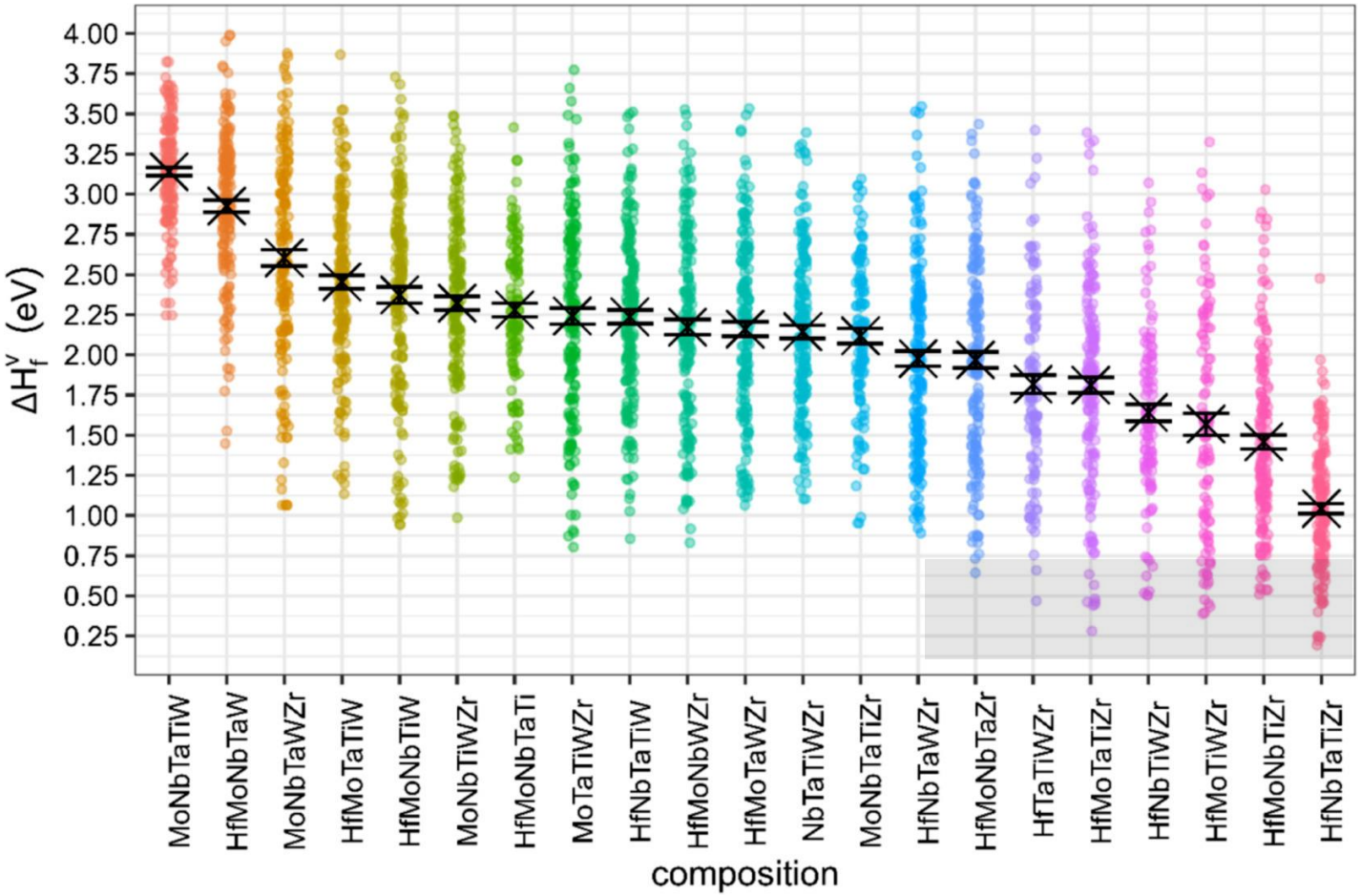
$$c_v(T) = exp\left(\frac{S_v^f}{k_B T}\right)\left[\int g(E_v^f) exp\left(-\frac{E_v^f}{k_B T}\right) dE_v^f\right]$$

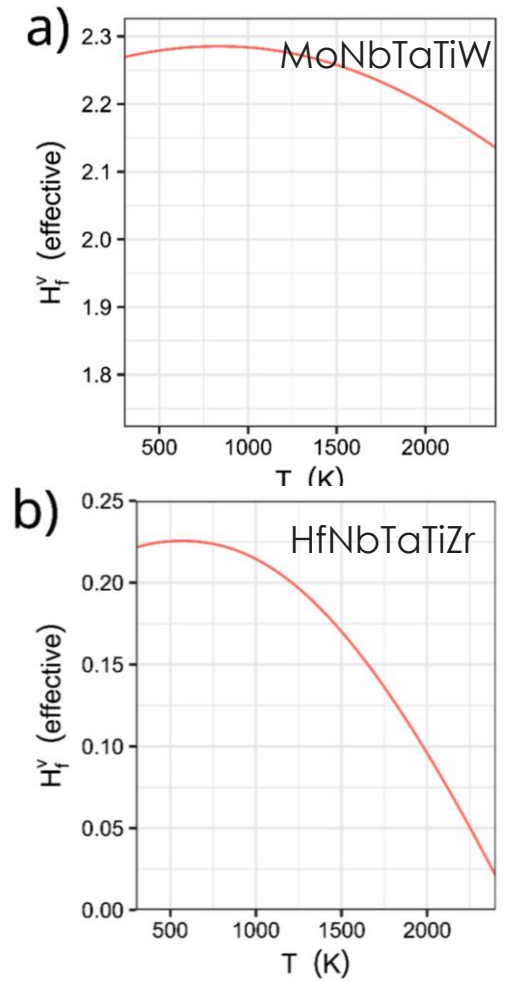


- Tail states can get activated at low temperature
- Reduced effective formation energy
- Increased vacancy concentration

Zhang et al. Computational Materials Science Volume 190, 1 April 2021, 110308.

Introduction: Vacancies in Multi-Element Alloys



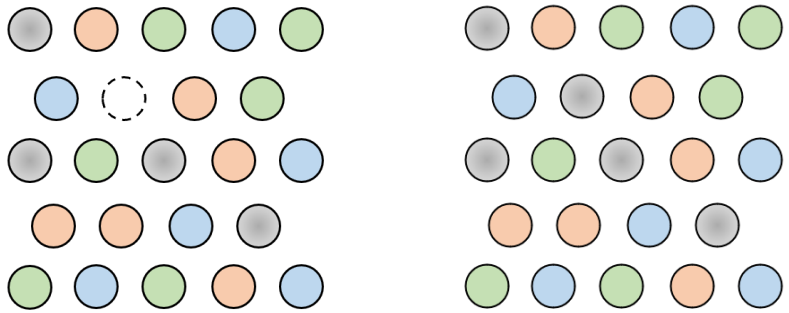


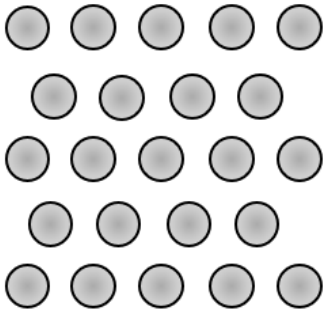
Can be active at low temperatures and result in large vacancy concentrations.

Wilson et al. Assessing the high concentration of vacancies in refractory high entropy alloys. Materialia Volume 28, May 2023, 101764.

Chemical Potentials

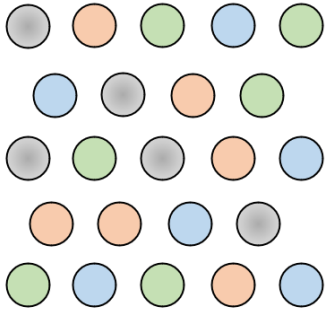
$$E_v^f = E_v - E_0 + \boldsymbol{\mu}$$





Single element in ground state

$$\mu = E_{atom}$$



Self-consistent chemical potential in alloy

$$\mu_{el} = E_0^{DFT} + E_{vac}^{mean} - \frac{1}{N_{el}} \sum_{i=1\ldots N_{el}} E_{v,i}^{DFT}$$

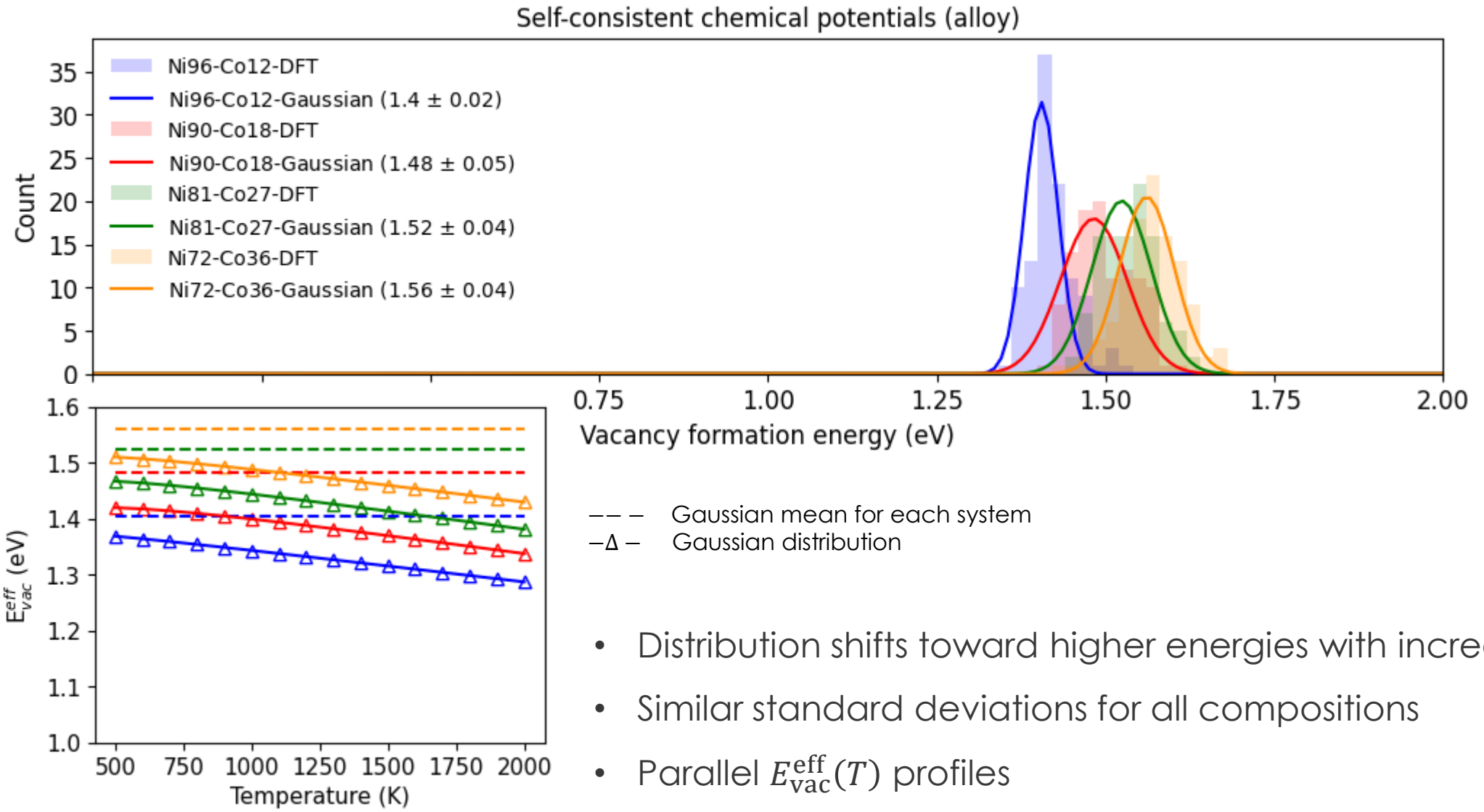
Zhang et al. Computational Materials Science Volume 190, 1 April 2021, 110308.

Methodology

1. **Define composition space**
   a) Base alloy: face-centered cubic (FCC) Ni (108 atoms)
   b) Alloying elements
   - Major transition metals (up to 33%)
   - Refractory metals (up to 16%)
   - Minor transition metals (up to 11%)

2. **Density functional theory (DFT) calculations in Vienna Ab initio Simulation Package (VASP)**
   - Various 2, 3, 4, 5-element alloys
3. **Machine learning (ML) models to predict vacancy formation energy**

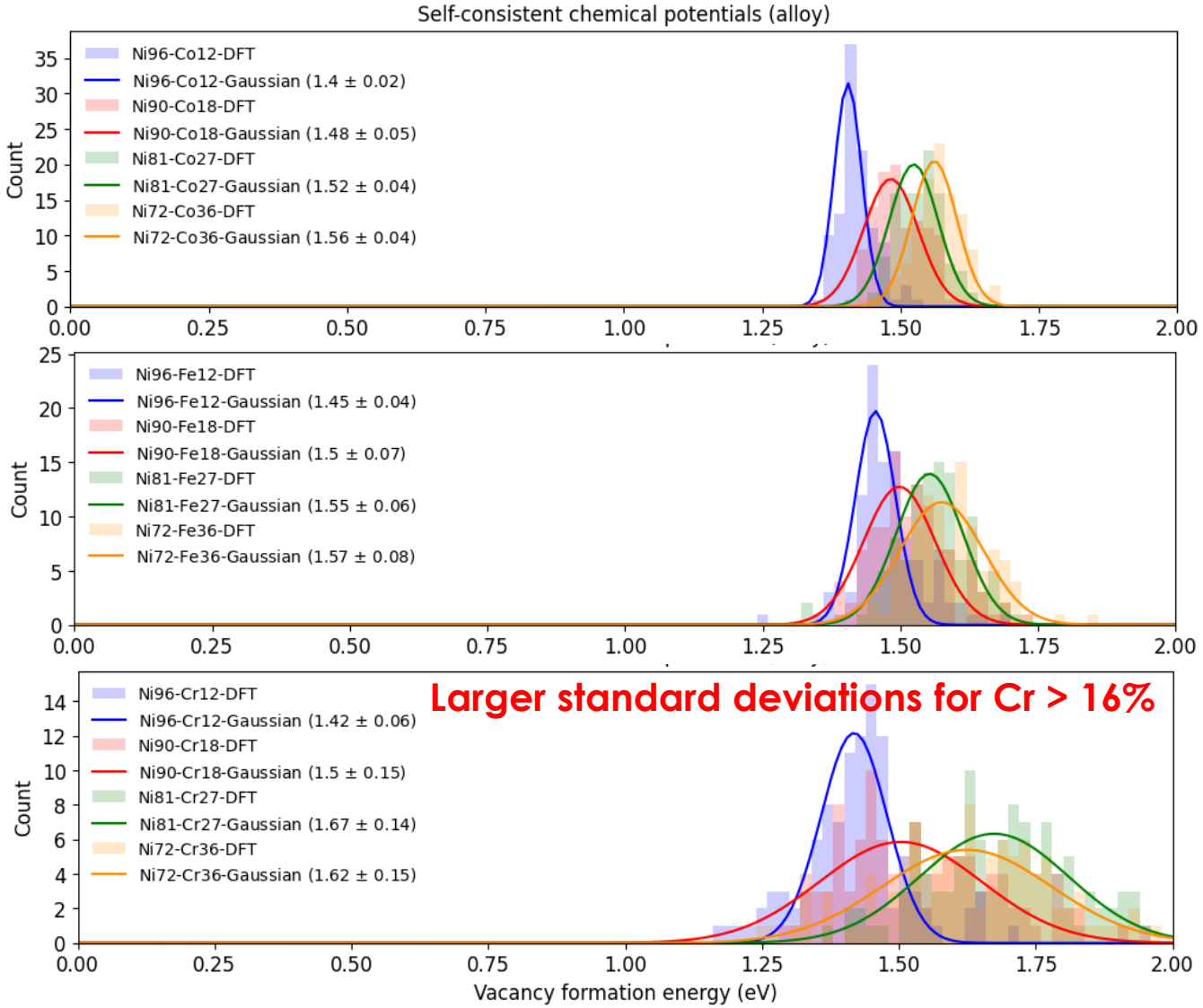
Ni-Transition Metal Binary Alloy Systems: Ni-Co

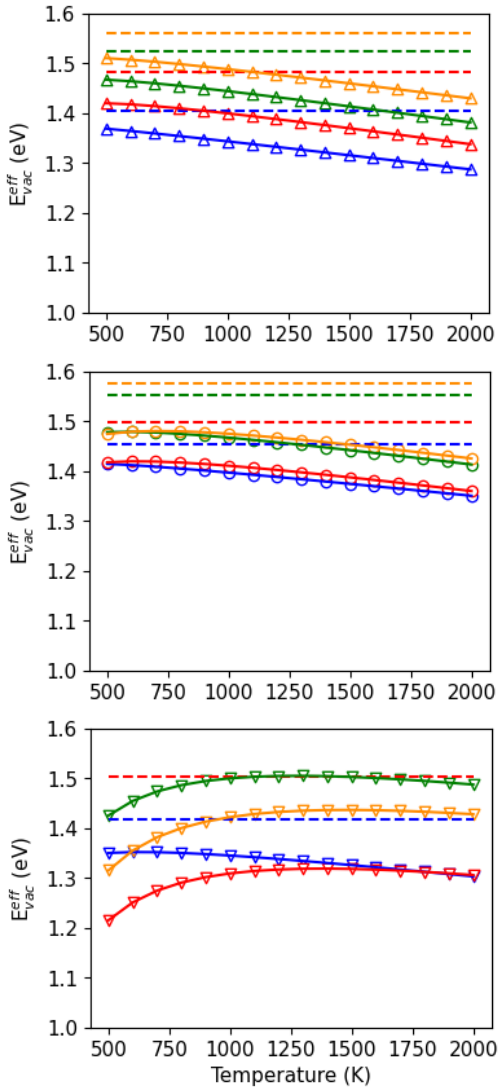


– – – Gaussian mean for each system
–Δ– Gaussian distribution

- Distribution shifts toward higher energies with increasing Co content
- Similar standard deviations for all compositions
- Parallel $E_{\mathrm{vac}}^{\mathrm{eff}}(T)$ profiles

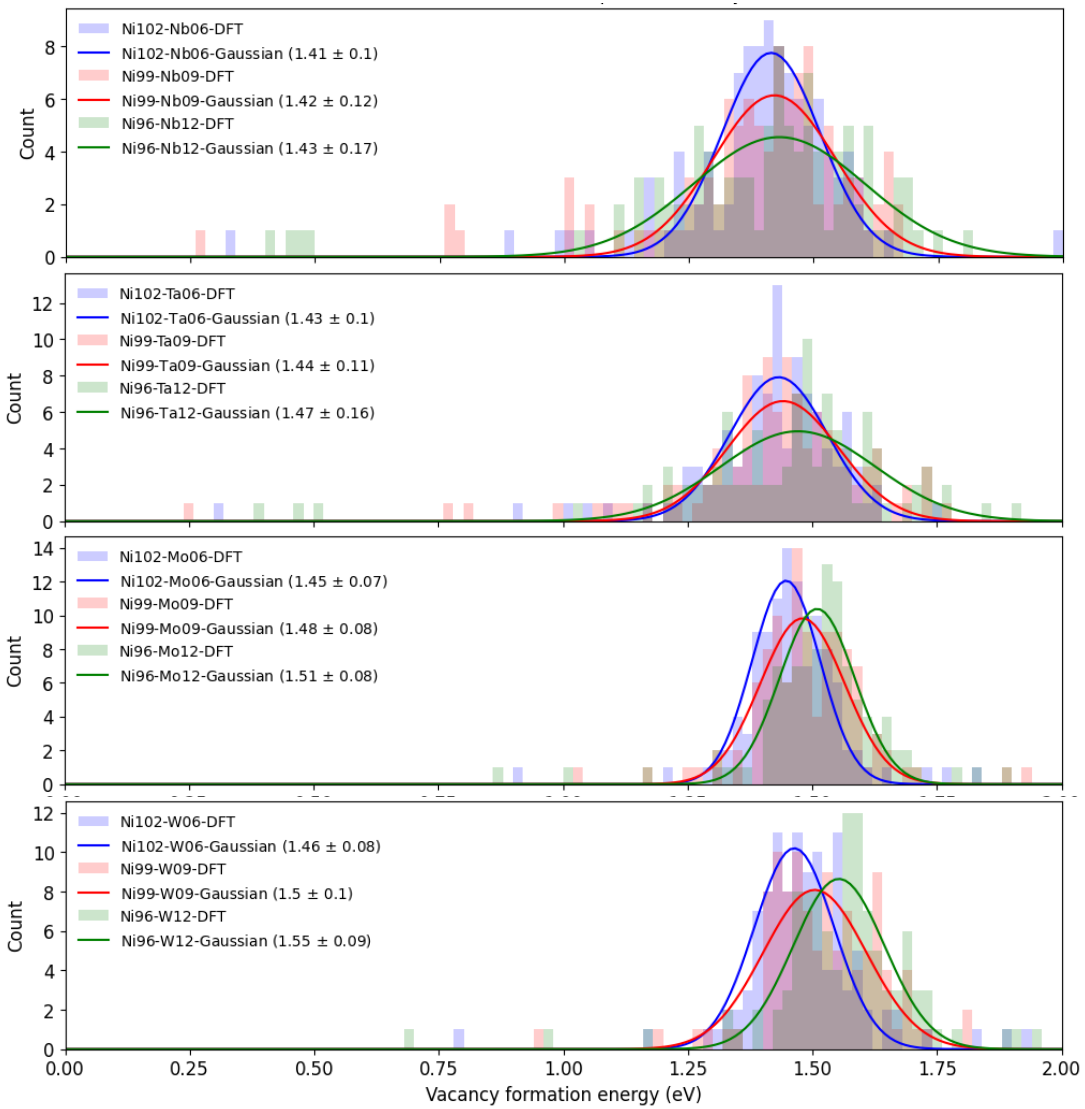
Ni-Transition Metal Binaries

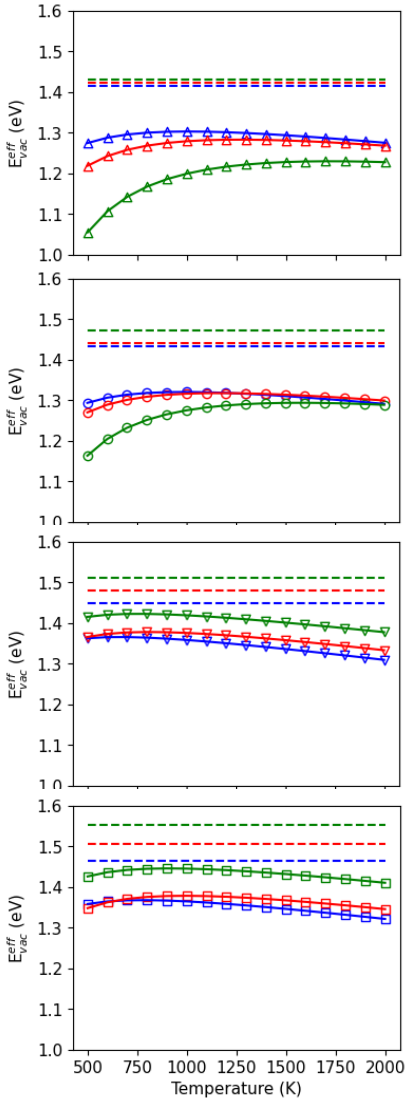




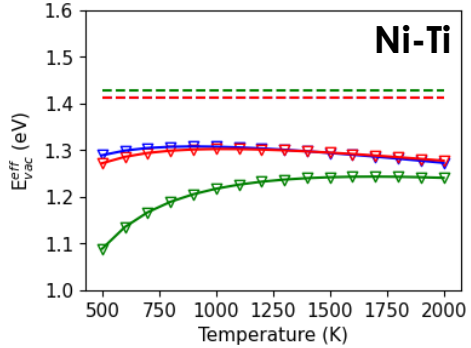
Reduced $E_{\mathrm{vac}}^{\mathrm{eff}}(T)$ at low temperature

Ni-Refractory Metal Binaries



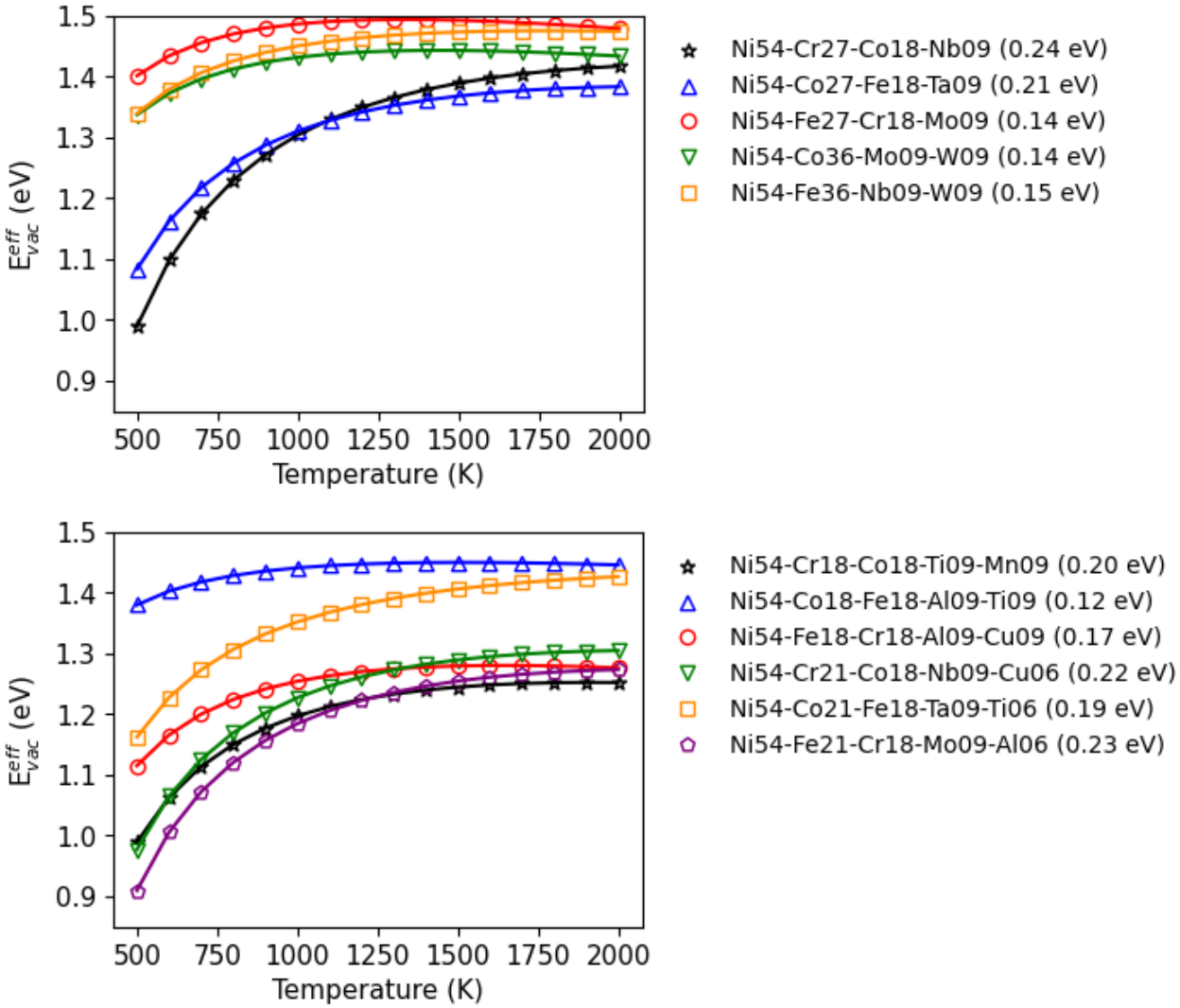


- Larger standard deviations for Nb, Ta
- Reduced $E^{\mathrm{eff}}_{\mathrm{vac}}(T)$ values at low temperature



- **Ni binaries with Cr, Nb, Ta, and Ti have large σ values ~0.15 eV**
- **Vacancies are active at low temperature**

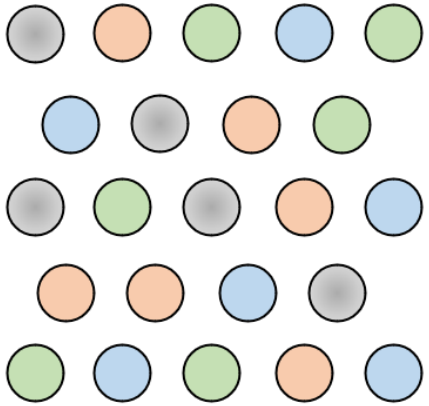
Quaternary and Quinary Alloys



- $E_{\mathrm{vac}}^{\mathrm{eff}}(T)$ depends non-trivially on alloy composition
- Need to develop ML models to rapidly predict values

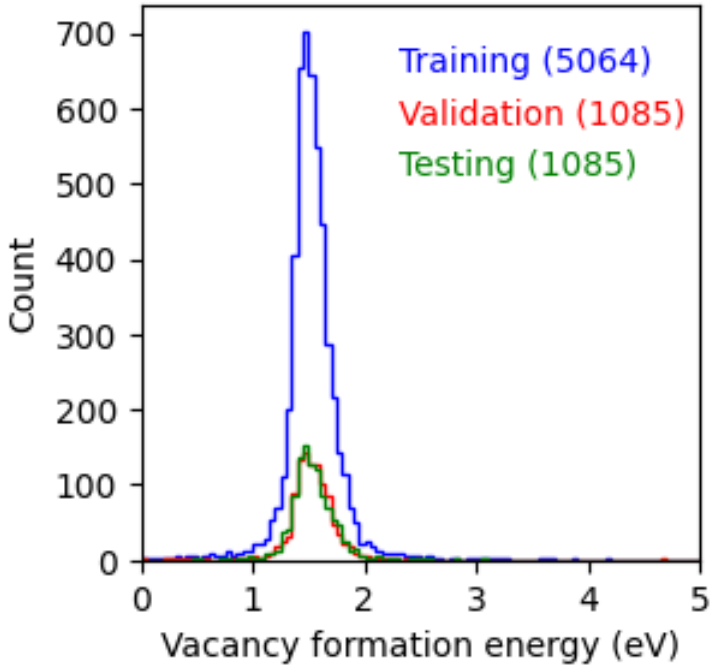
Graph Neural Network

- FCC lattice constant using rule-of-mixtures
- Create vacancy
- Construct graph using ALIGNN model



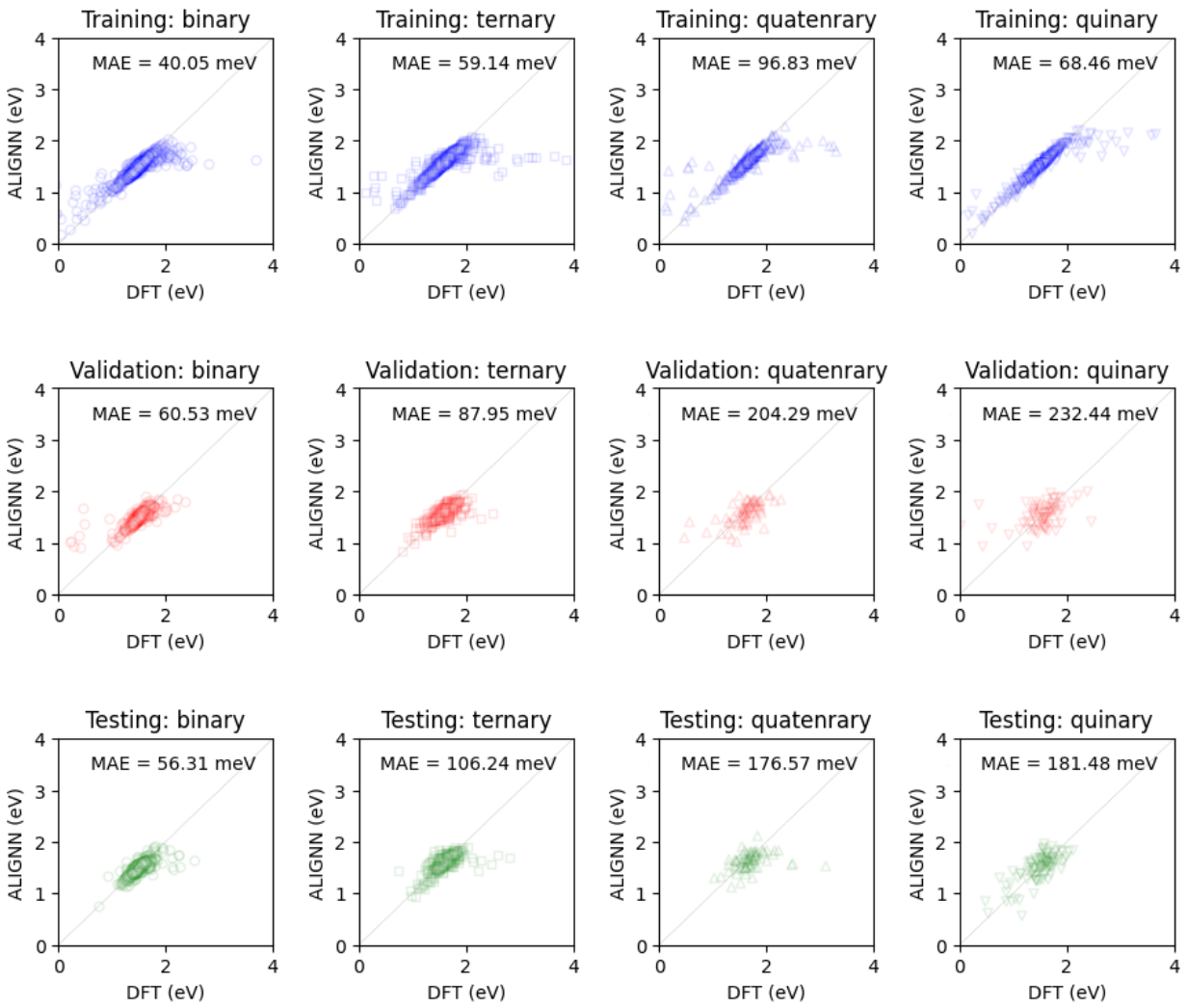
ALIGNN: Choudhary and DeCost, *npj Computational Materials* volume 7, Article number: 185 (2021).

- 70-15-15 training-validation-testing split



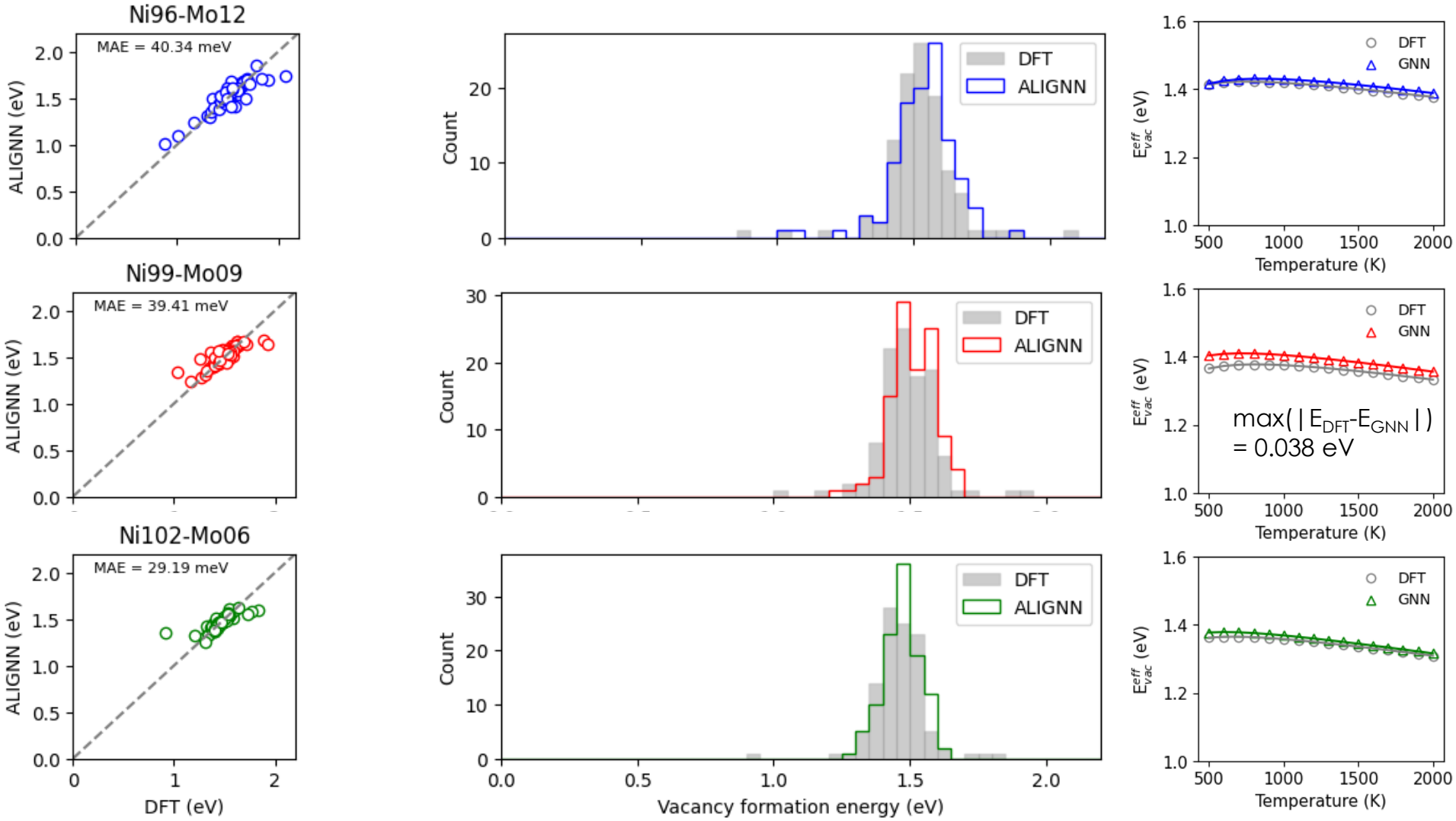
- Other regression models are not very efficient

Performance by Alloy Type

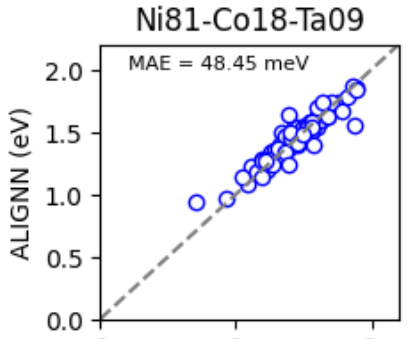


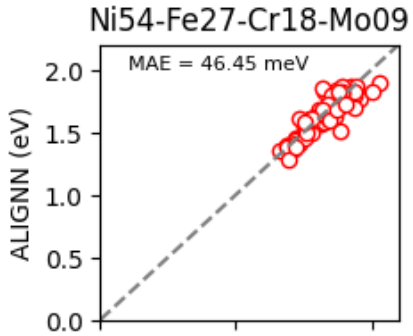
- MAE for 2, 3-element alloys < 0.1 eV
- MAE for 4, 5-element alloys ~ 0.2 eV

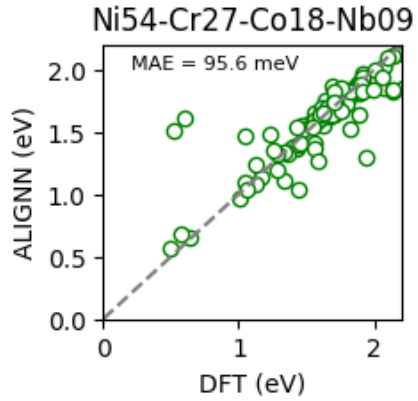
Energy Distribution for Binary Alloys

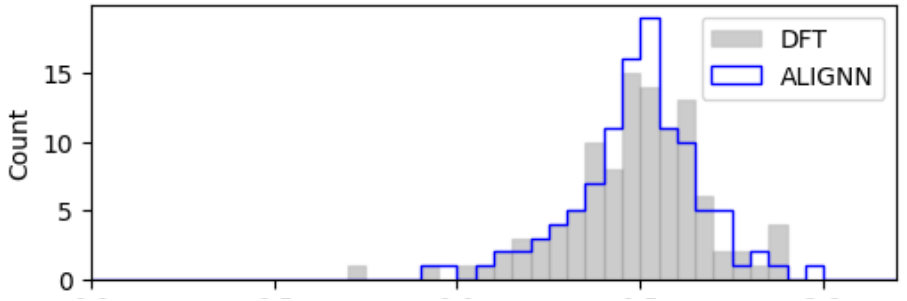


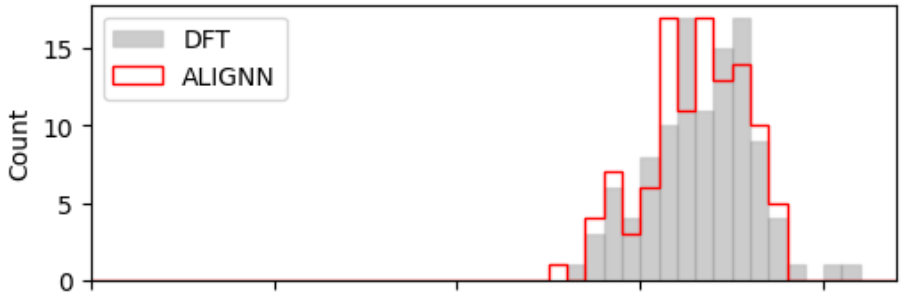
Energy Distribution for Complex Alloys

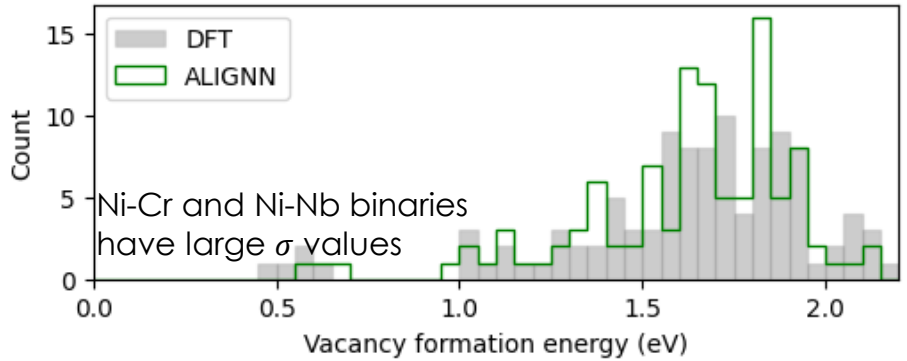


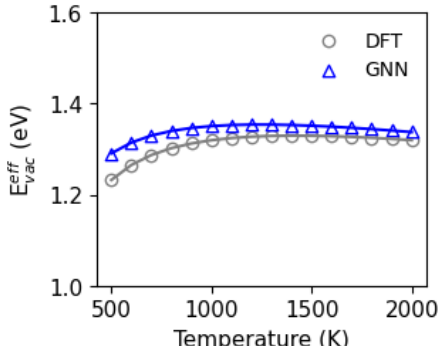


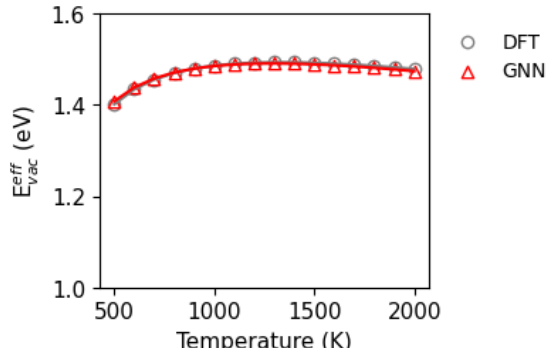


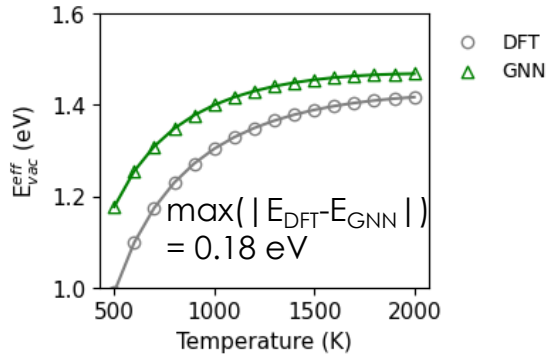




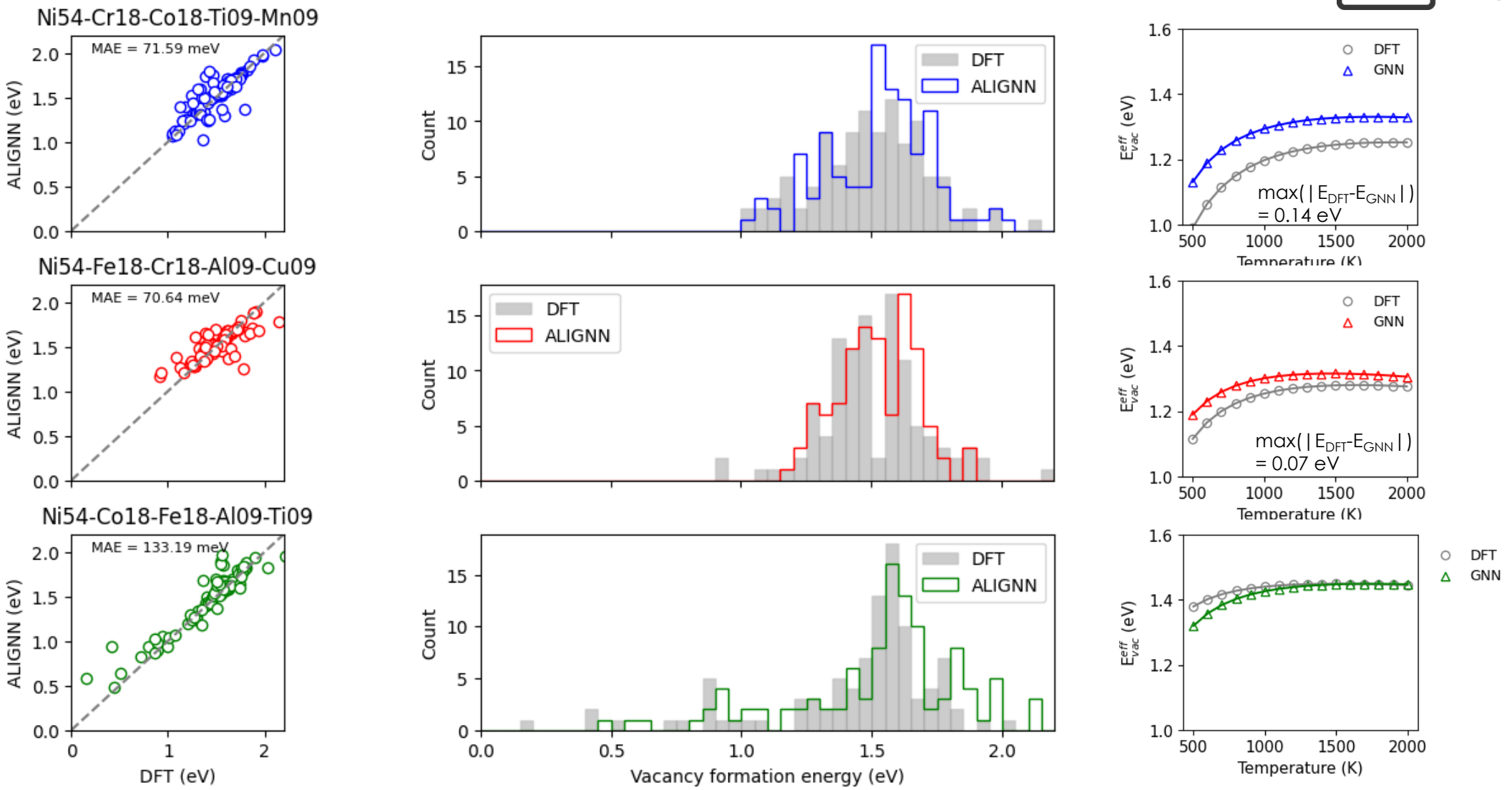








Quinary Alloys



Conclusion and Future Work

- Studied vacancy formation energy in Ni alloys systematically, which is important to creep performance and oxidation behavior.
- Created database of vacancy formation energies for a number of Ni alloys: Al, Ti, Cr, Mn, Fe, Co, Cu, Nb, Mo, Ta, and W.
- Cr, Nb, Ta, and Ti induced low formation energy states.
- Activation at low temperatures can result in reduced effective formation energy values, and high vacancy concentration.
- Model the migration energy barrier in Ni alloys.

Acknowledgements

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NETL colleagues: Shiqiang Hao, Richard Oleksak, William Trehern, Madison Wenzlick

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