Machine Learning Vacancy Formation Energy in Nickel-Based Superalloys

Aditya Sundar

NETL Support Contractor

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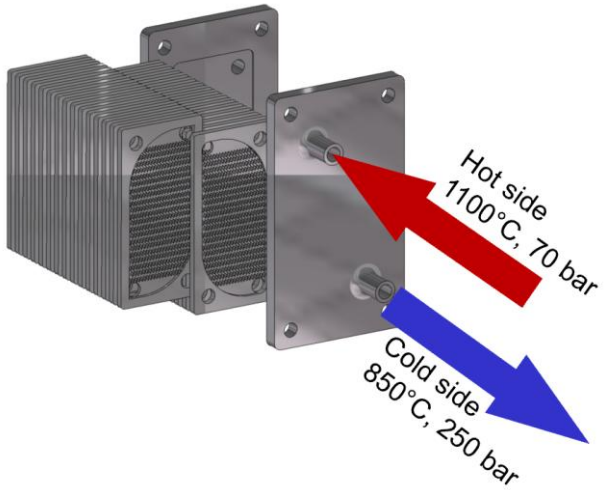
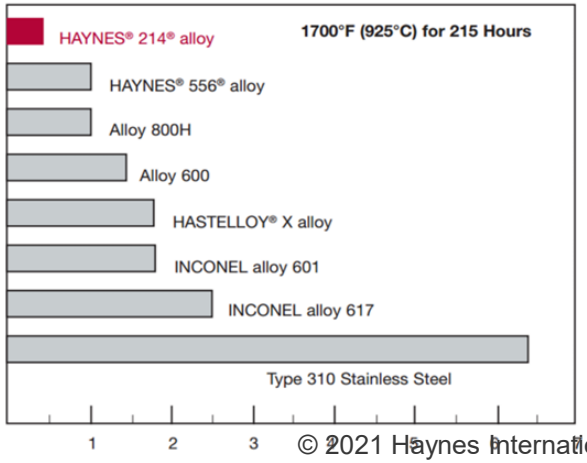
Introduction: Ni Superalloys and Applications

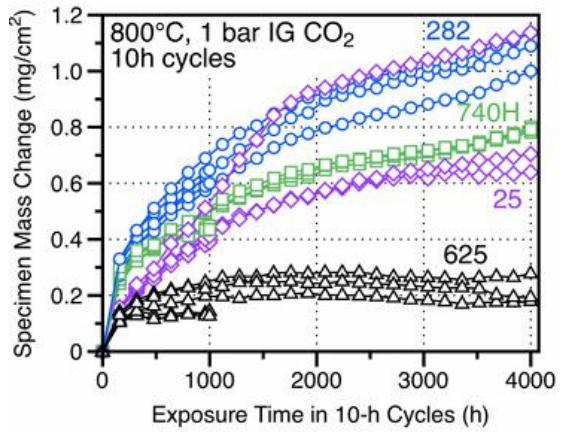
Turbine Blades

Heat Exchangers

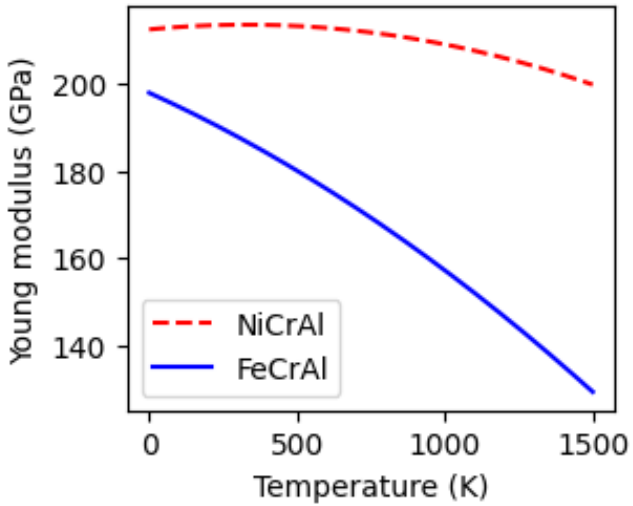


Corrosion Resistance





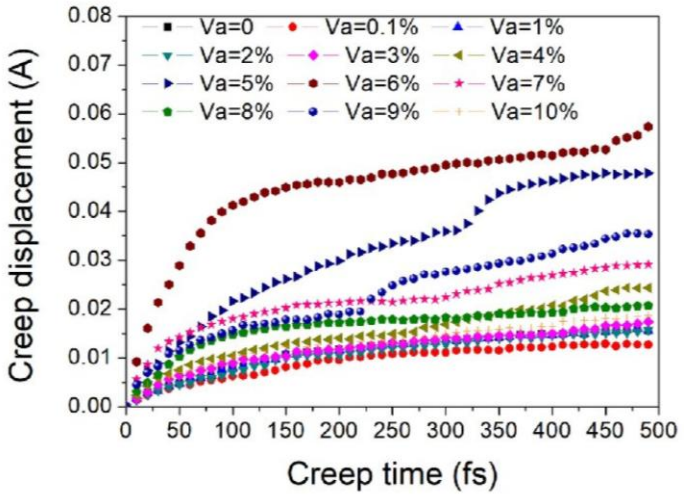
High Strength

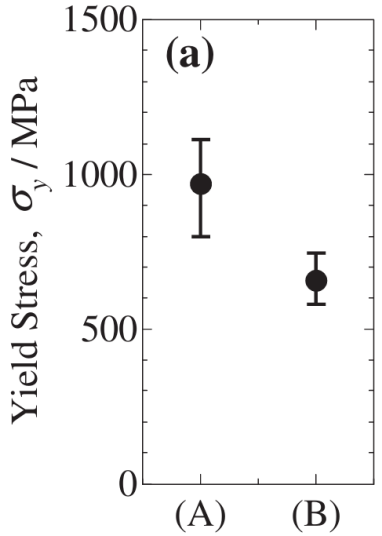


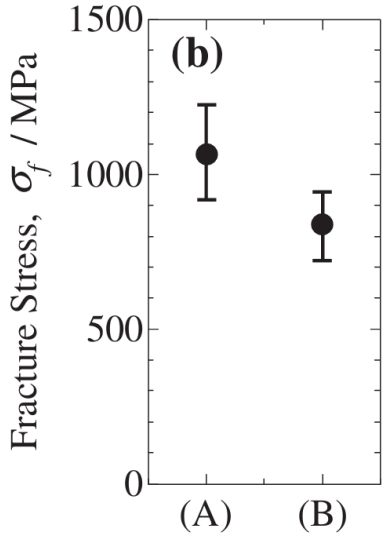
https://mooseframework.inl.gov
https://www.haynesintl.com/

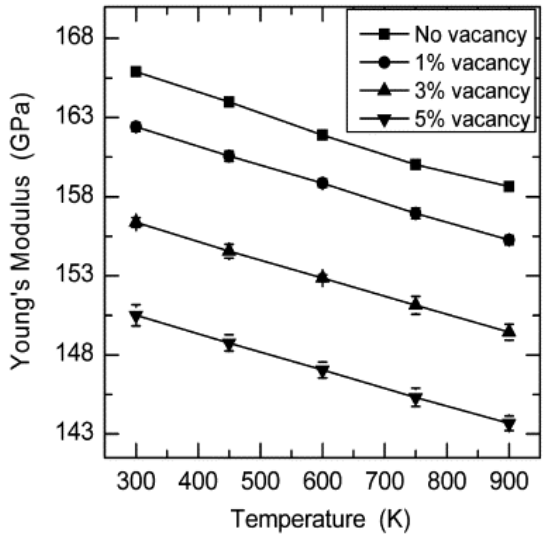
Pint and Pillai DE-EE0001556 Lifetime Model Development for sCO2 CSP (2019).

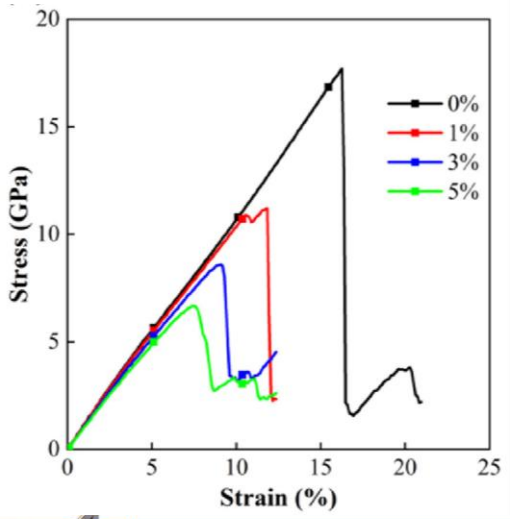
Impact of Vacancies on Strength & Creep Properties







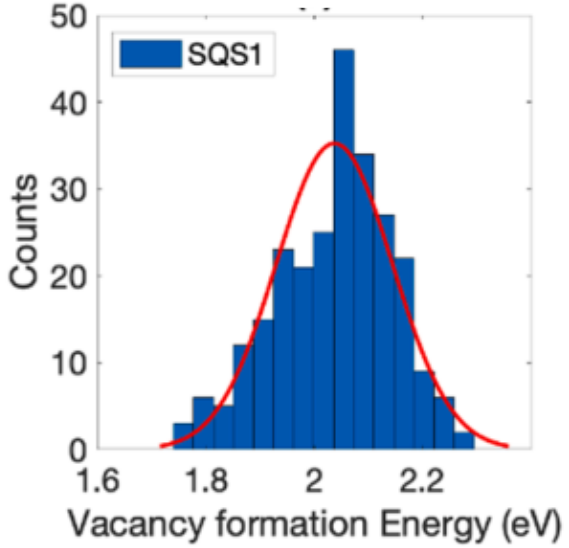


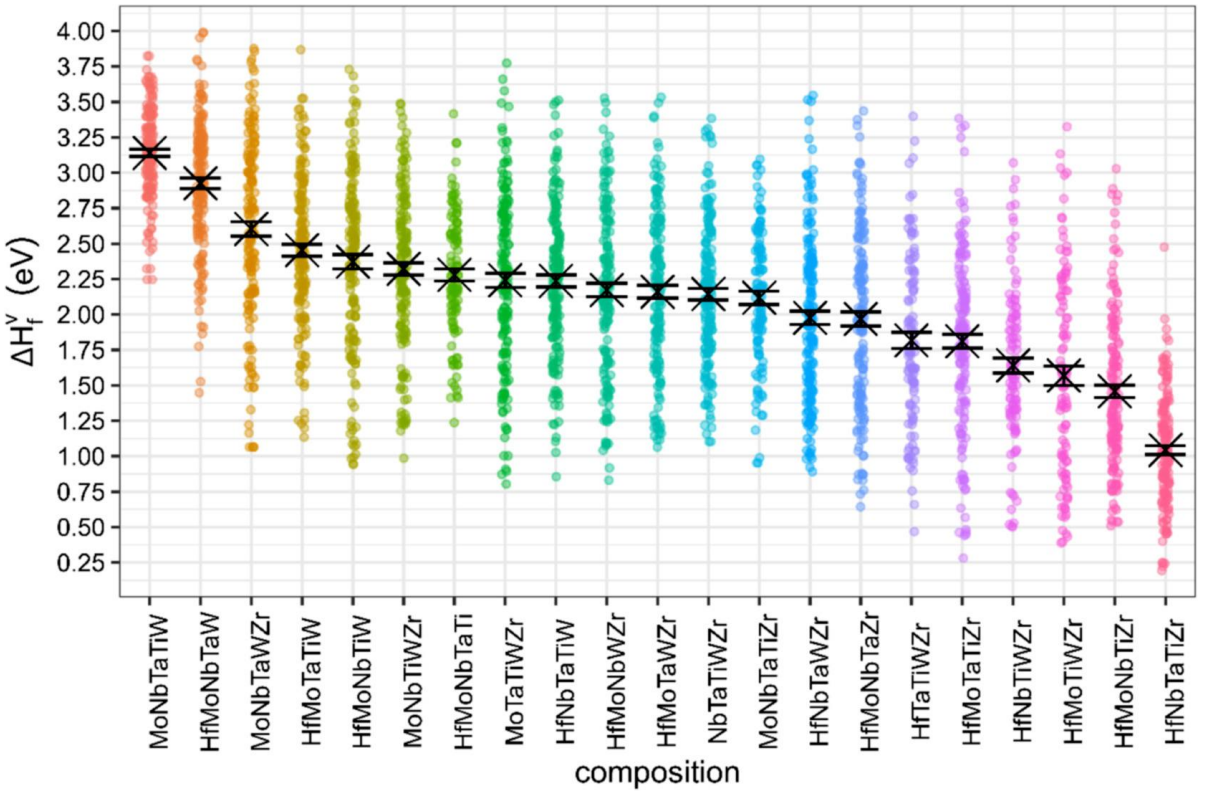


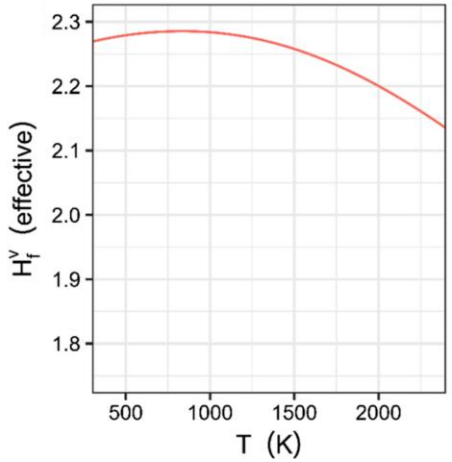
- Higher creep displacements
- Reduced yield and fracture stress, elastic moduli

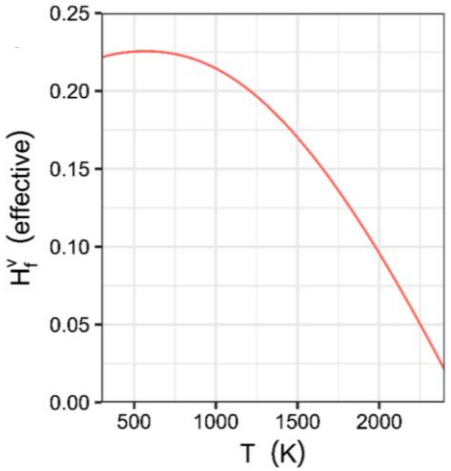
Yang et al. Yuasa et al. Materials Transactions, Vol. 49, No. 10 (2008).
Cui et al. *Coatings* 2024, 14(1), 63.
Physica B: Condensed Matter Volume 407, Issue 12, 15 June 2012, Pages 2234-2238.
Peng et al., International Journal of Mechanical Sciences Volume 218, 15 March 2022, 107065.

Vacancies in Multi-Element Alloys









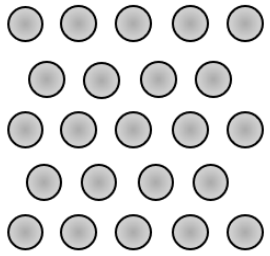
- Tail states can get activated at low temperatures
- Reduced effective formation energy
- Increased vacancy concentration

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

Zhang et al. Computational Materials Science Volume 190, 1 April 2021, 110308., 2) Wilson et al. Materialia Volume 28, May 2023, 101764.

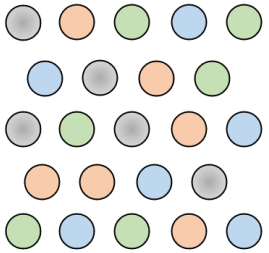
Methodology

1. **Define composition space**
   a) Base alloy: Face Centered Cubic Ni with 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 calculations for various 2-6 element alloys**
   (36 binary, 20 ternary, 11 quaternary, 9 quinary, and 3 senary)



Single element in ground state

$$\mu = E_{atom}$$

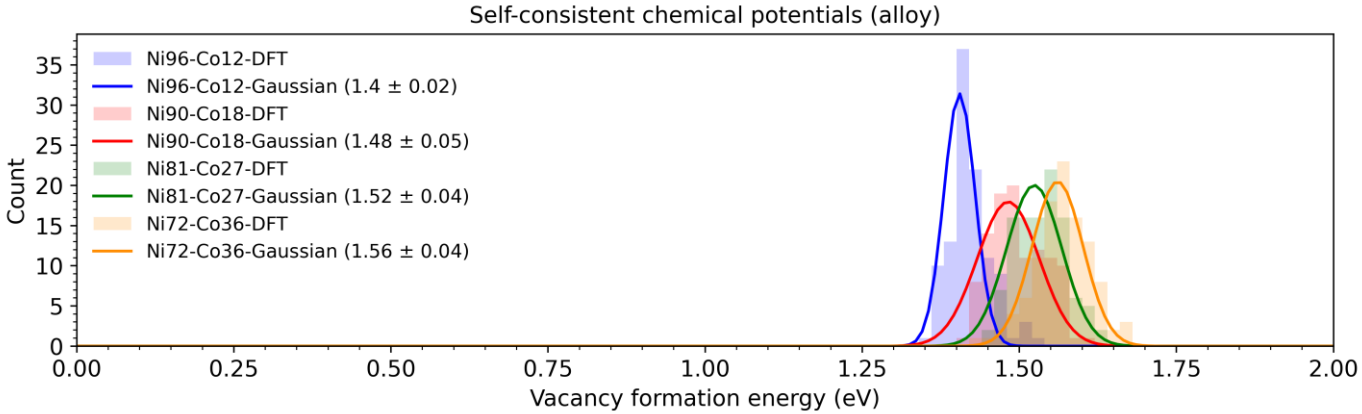


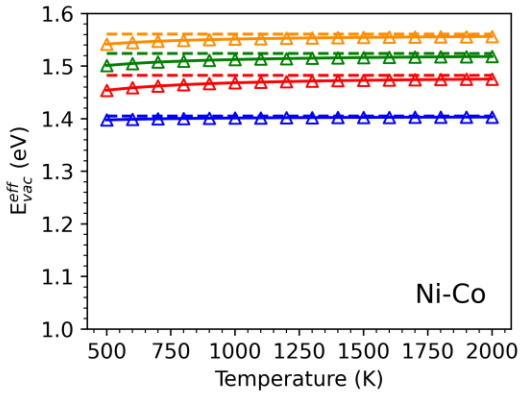
Self-consistent chemical potential in alloy

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

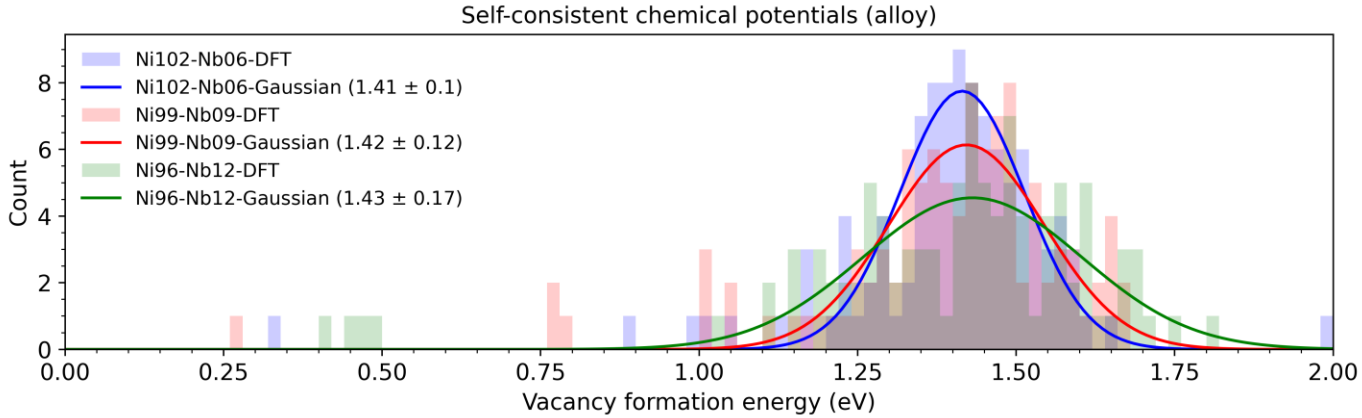
Zhang et al. Computational Materials Science Volume 190, 1 April 2021, 110308

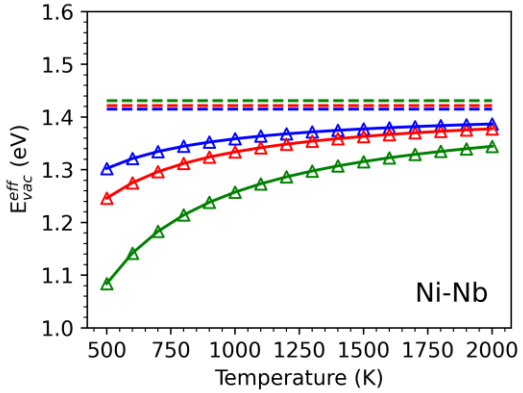
Ni-Transition Metal Binary Alloy Systems





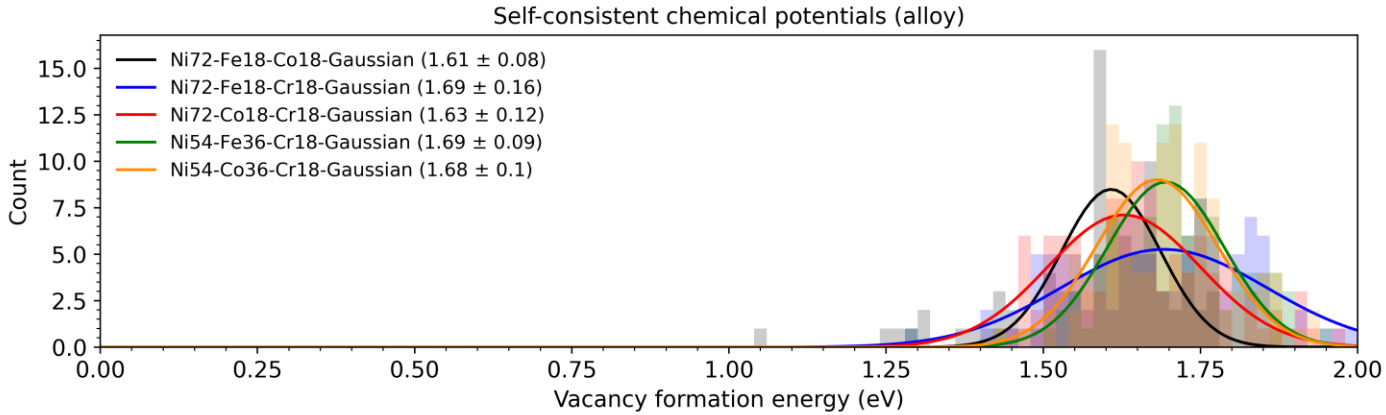
- Distribution shifts toward higher energies with increasing Co content
- Similar standard deviations for all compositions
- Parallel $E_{\mathrm{vac}}^{\mathrm{eff}}(T)$ profiles

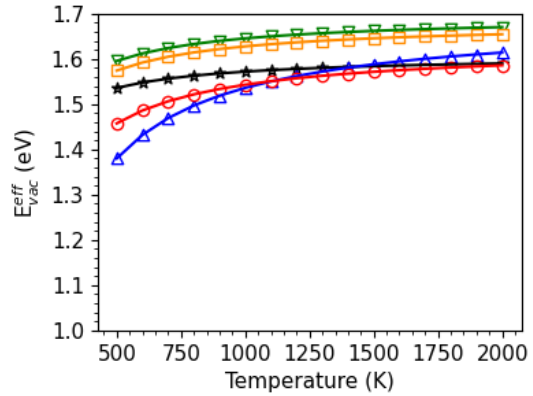




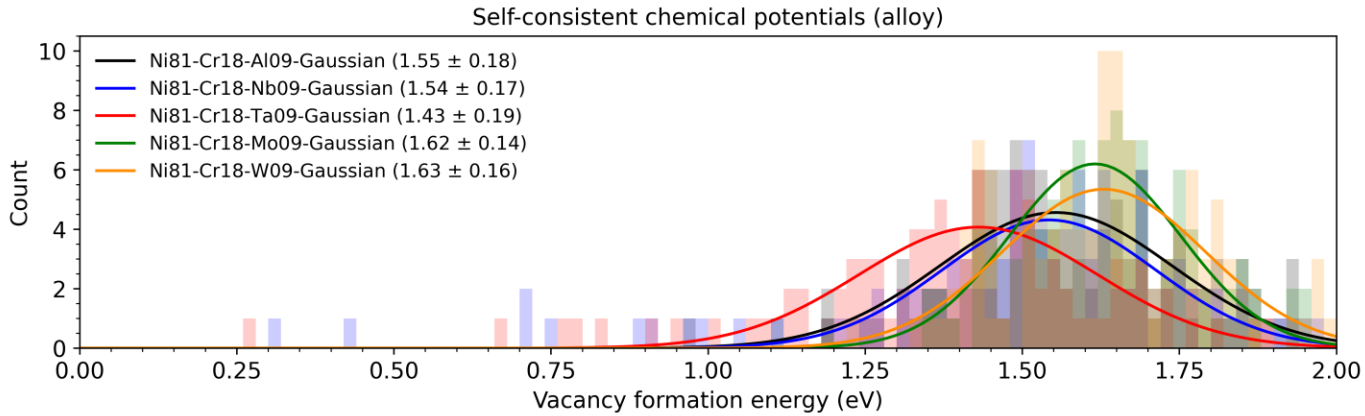
- Larger standard deviations for Ni-Nb
- Reduced $E_{\mathrm{vac}}^{\mathrm{eff}}(T)$ values at low temperatures

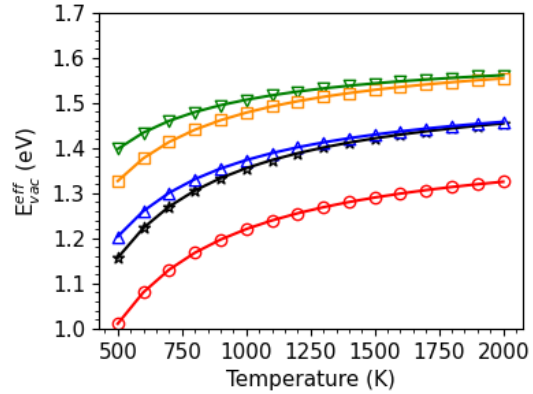
Effects in Multi-Element Alloys





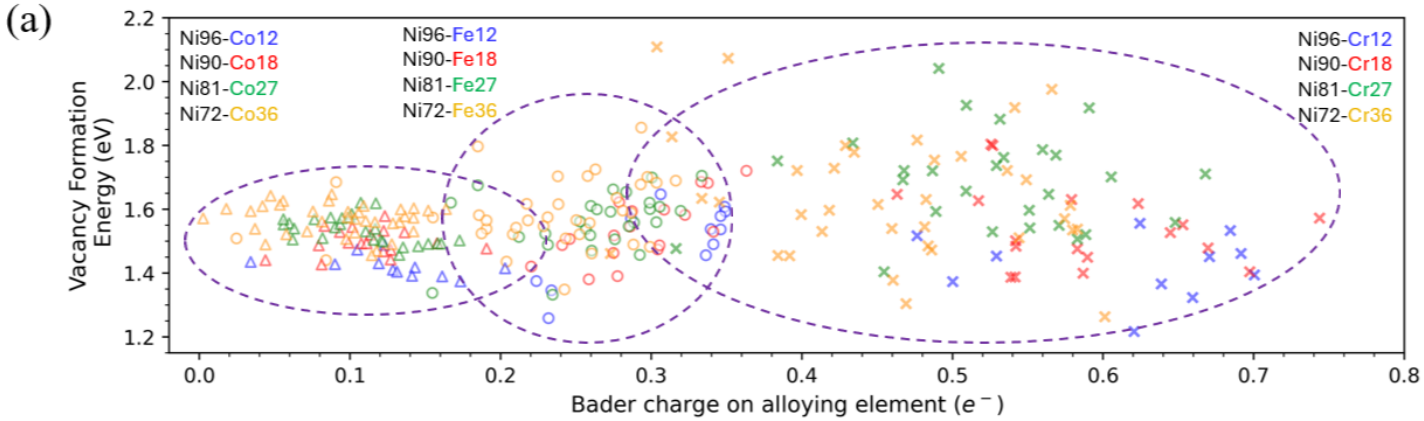
- Adding Fe/Co/Cr
- $\sigma(E_{vac})$ ~0.1–0.15 eV
- E_{vac}^{eff} ~1.4–1.6 eV at 500 K

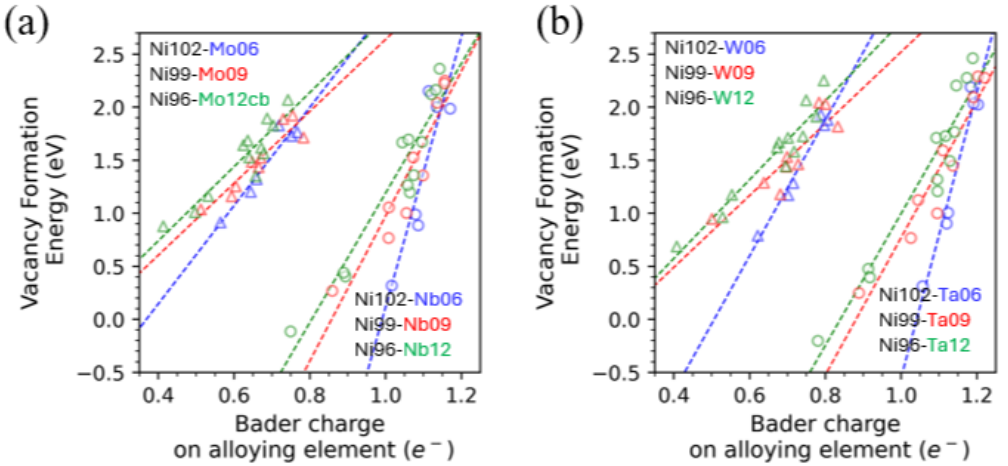


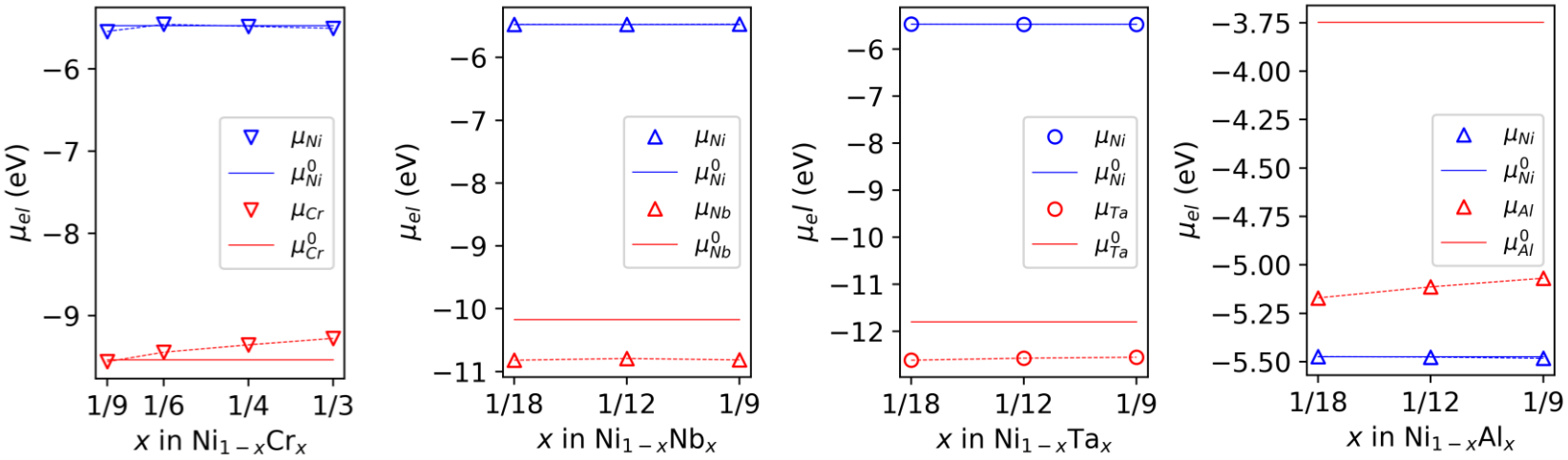


- Combining Cr and Ta/Nb
- $\sigma(E_{vac})$ ~0.14–0.2 eV
- E_{vac}^{eff} ~1 eV at 500 K

Correlations with Charge Localization and Impact on Chemical Potential



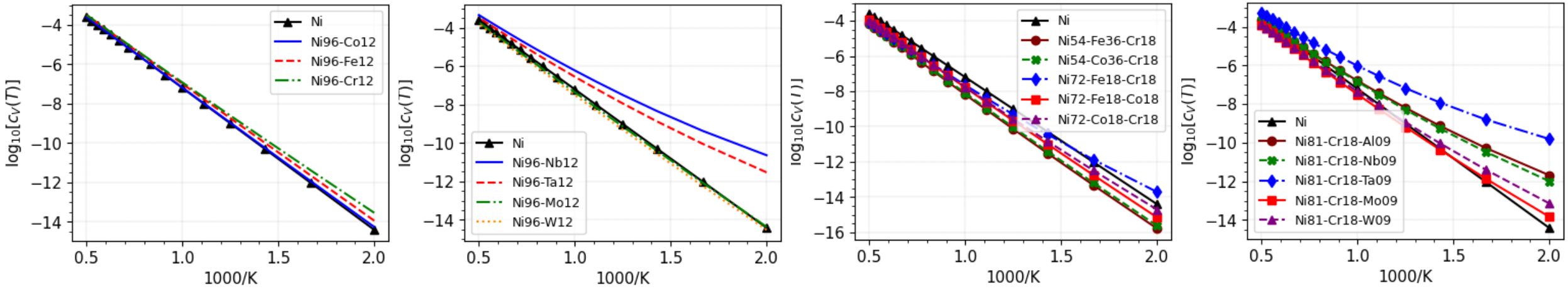




- Large charge localization at Cr, Nb, Ta, and Al sites results in decreased chemical potentials
- Impacts $\mu(E_{vac})$ and $\sigma(E_{vac})$ values

Effects systematically observed in multi-element alloys

Impact on Vacancy Concentration

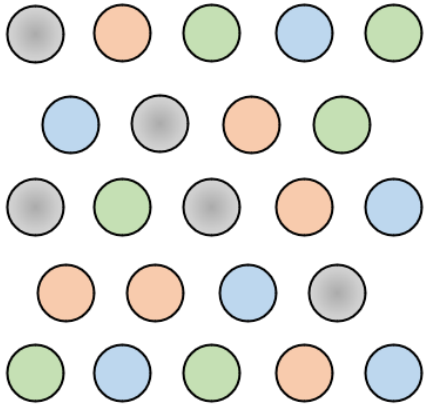


- **Minimal impact of Co and Fe in binary alloys, 1 order of magnitude rise upon adding Cr**
- **At 500 K, vacancy concentration increases 3-4 orders of magnitude upon Nb and Ta addition**
- **In ternary alloys:**
  - Adding Cr and Ta increases vacancy concentration by ~5 orders at 500 K
  - Adding Cr and Nb/Al results in ~3 orders increase

Goal: Screen Ni-alloys with high $\mu(E_{vac})$ and low $\sigma(E_{vac})$

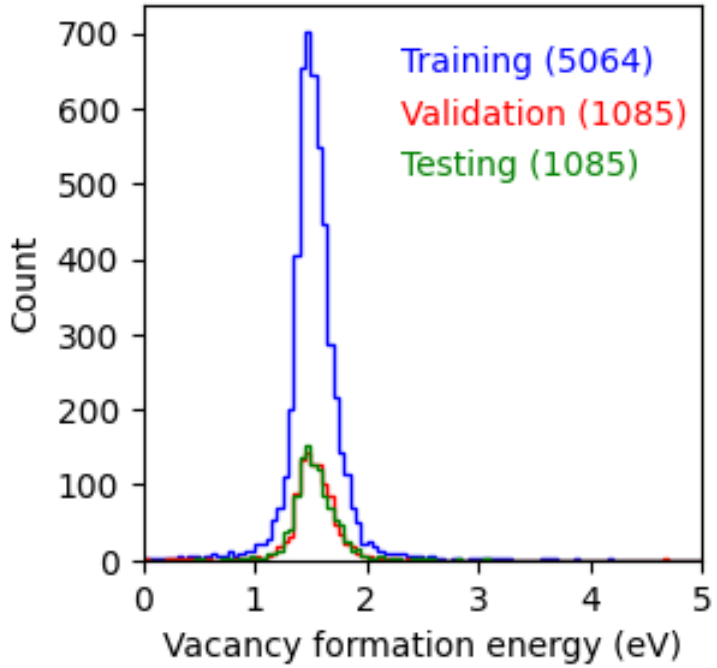
Graph Neural Networks

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



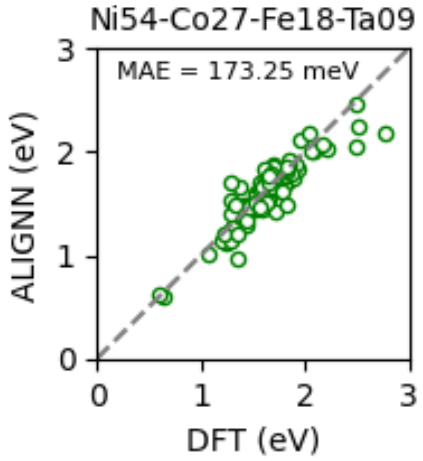
ALIGNN: Choudhary and DeCost, *npj Computational Materials* volume 7, Article number: 185 (2021).

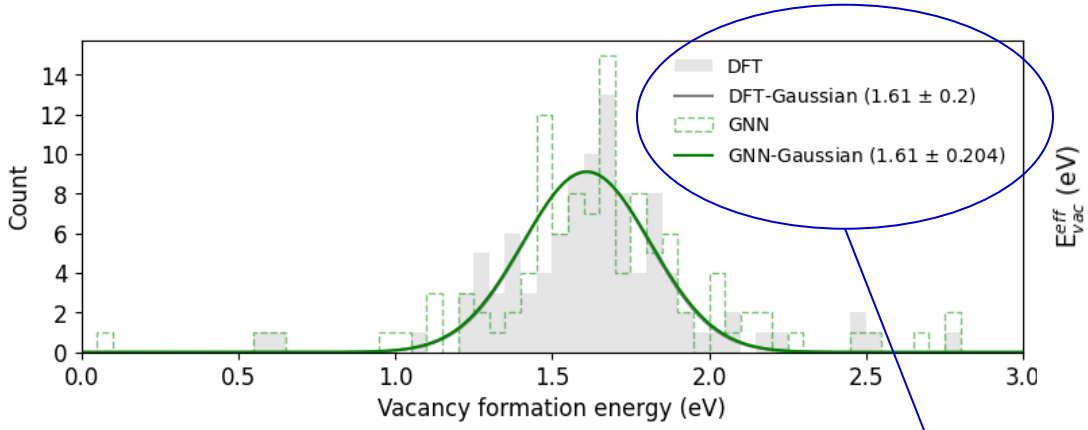
70-15-15 Training-Validation-Testing Split

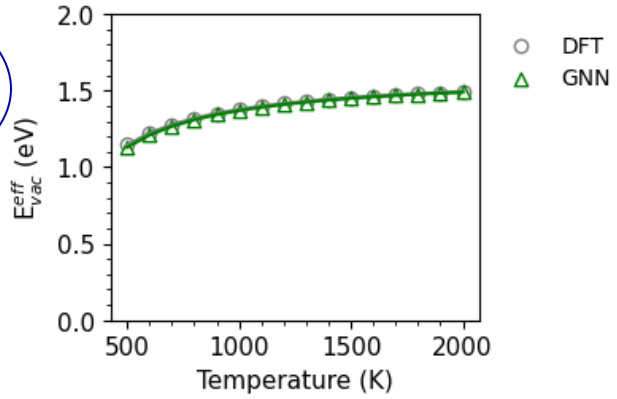


Other regression models are not very efficient

ML-Predicted Energy Distributions

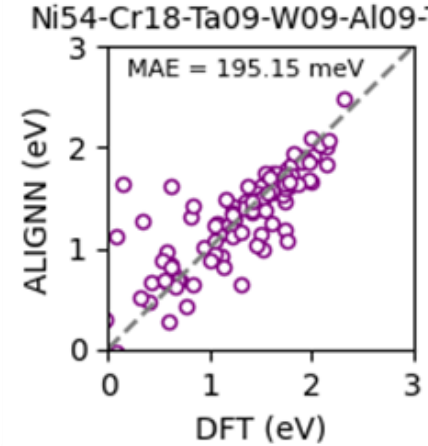


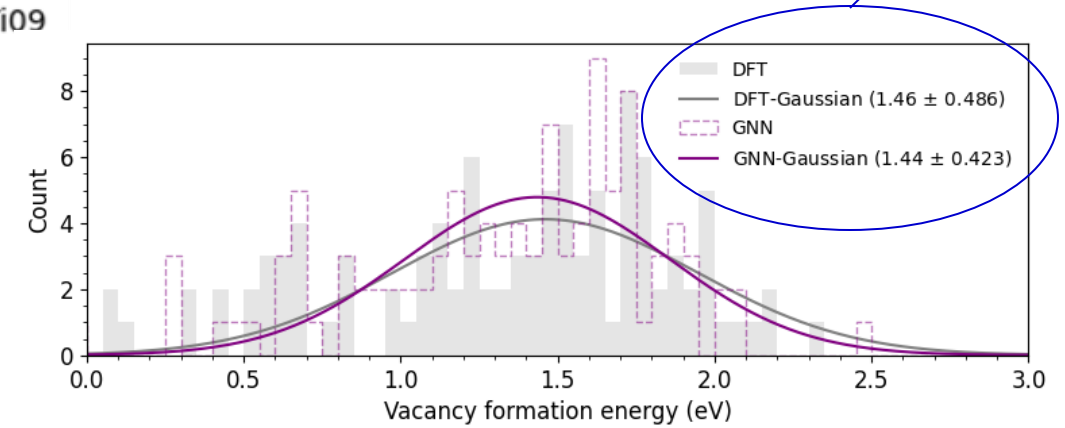


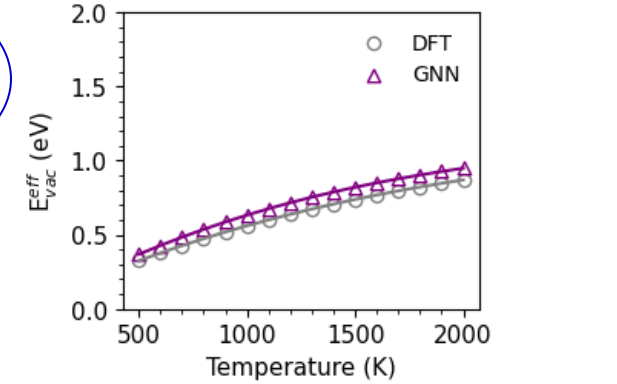


Error over each atomic site for this composition

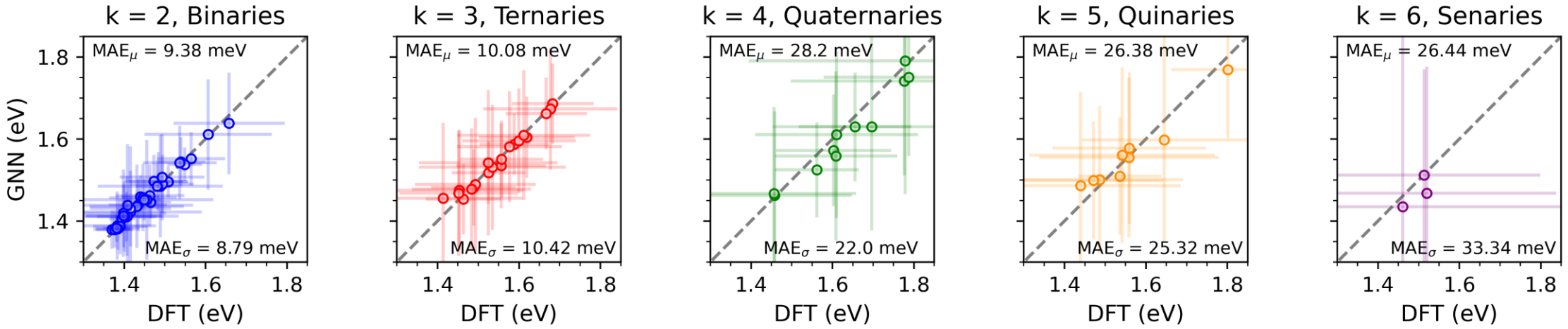
Much lower errors for $\mu(E_{vac})$ and $\sigma(E_{vac})$

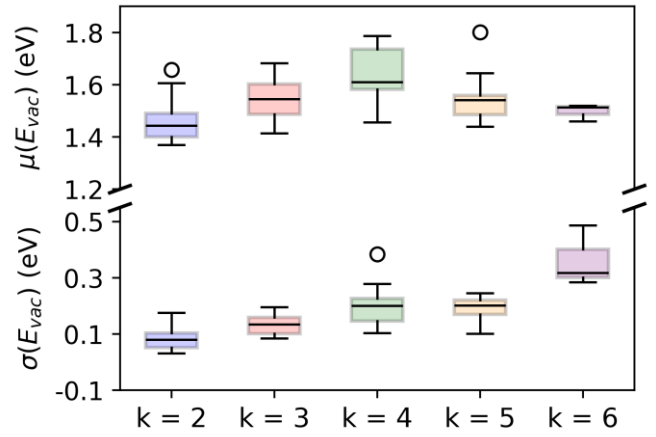






ML-Predicted Distributions for $\mu(E_{vac})$ and $\sigma(E_{vac})$

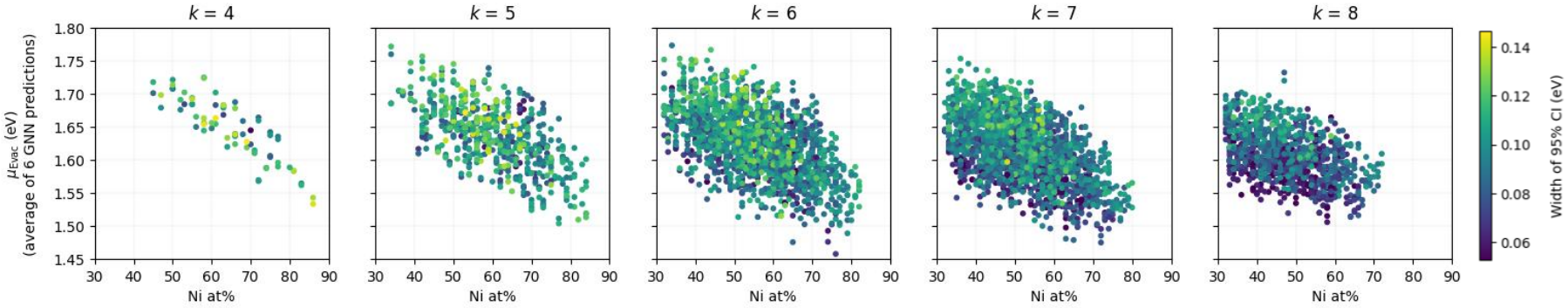


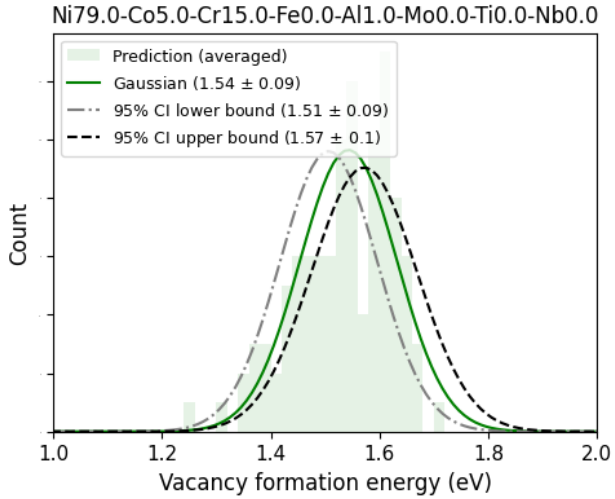


- $\mu(E_{vac})$ in 4, 5, 6-element alloys remains high 1.34-1.81 eV (value in pure Ni = 1.4 eV)
- However, $\sigma(E_{vac})$ is as low as 0.49 eV → vacancy formation energy as low as 0.5 eV at 1000 K → large vacancy concentrations

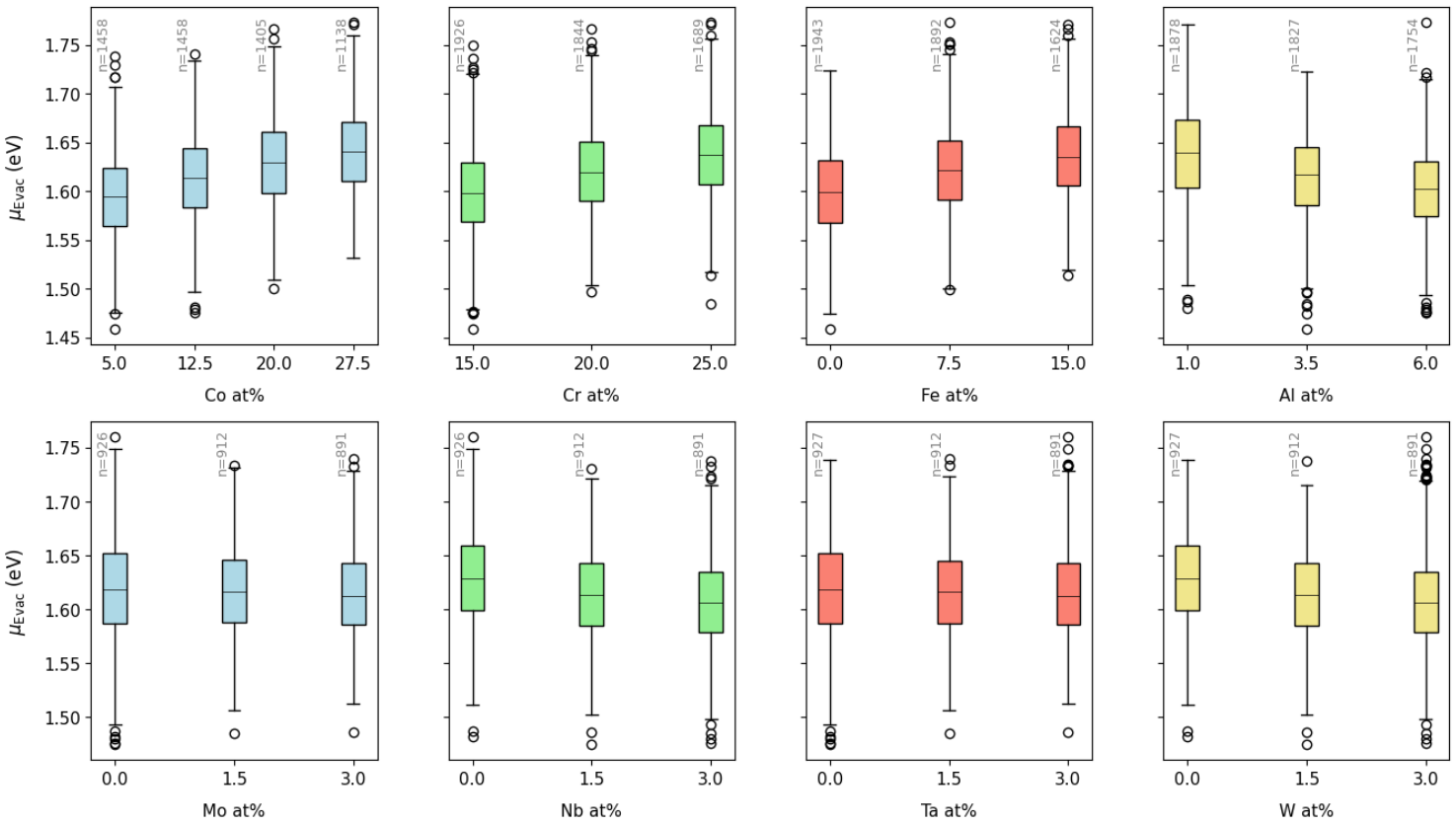
Screening Alloys with Low Vacancy Concentrations

Composition (at%)			
Elem	Lower bound	Upper bound	δ
Ni	Balance ≥ 30		
Co	5	27.5	7.5
Cr	15	25	5
Fe	0	15	7.5
Mo/W	0	3	1.5
Nb/Ta	0	3	1.5
Al	1	6	2.5
Ti	0	4	2
Total	2730 x 2 = 5460		

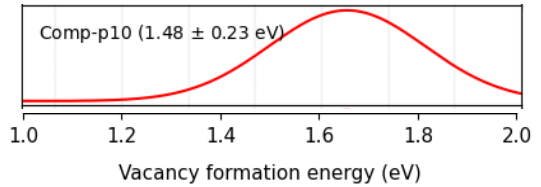


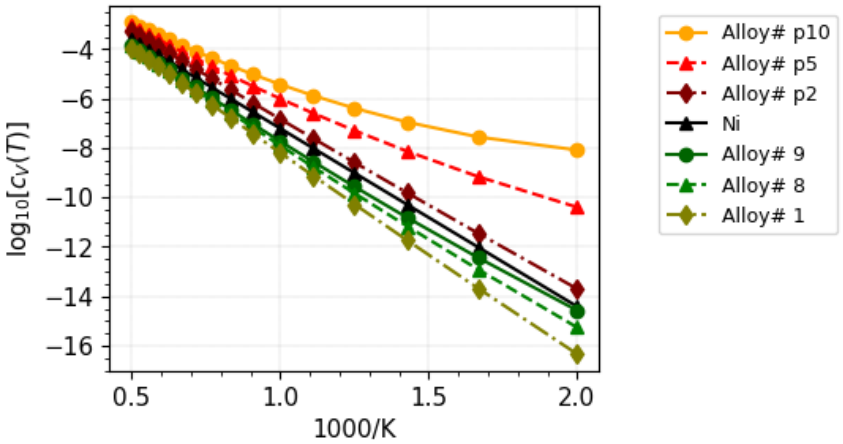


Screening Alloys with Low Vacancy Concentrations

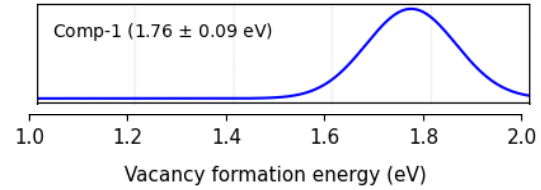


Ni67.0-Co5.00-Cr15.0-Al6.0-W0.0-Ti4.0-Ta3.0





Ni31.5-Co27.5-Cr25.0-Fe15.0-Al1.0

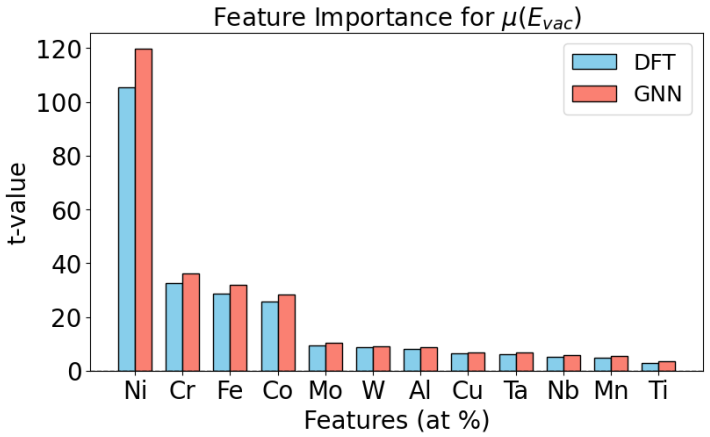


- ML models rapidly identify alloys with $\mu(E_{vac})$ > 1.75 eV and $\sigma(E_{vac})$ < 0.1 eV
- Vacancy concentration ~10^2 lower than in pure Ni

Conclusion and On-Going Work

- Cr/Nb/Ta/Al/Ti increased $\sigma(E_{vac})$ and introduced low-energy vacancy states
- ML models rapidly identify alloys with $\mu(E_{vac})$ > 1.75 eV and $\sigma(E_{vac})$ < 0.1 eV
- Vacancy concentration 2 orders of magnitude lower than pure Ni at 1000 K
- These alloys are rich in Fe-Co-Cr and contain Al, W

Multivariate Linear Regression



AI/ML/Data Informatics for Materials Discovery: Bridging Experiment, Theory, and Modeling: Property and Performance Predictions

- Predicting the High-Temperature Oxidation Response of Nickel Superalloys Using CALPHAD-Enhanced Machine Learning – *Aditya Sundar*
- A Design-Focused Machine Learning Framework for Creep Behavior in Structural Alloys - *Madison Wenzlick*