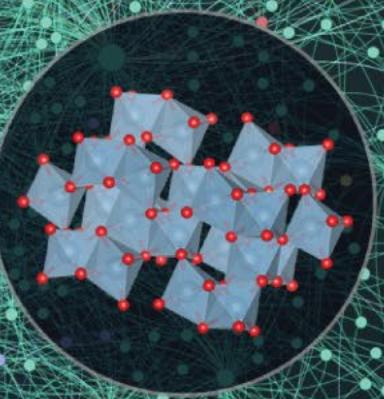
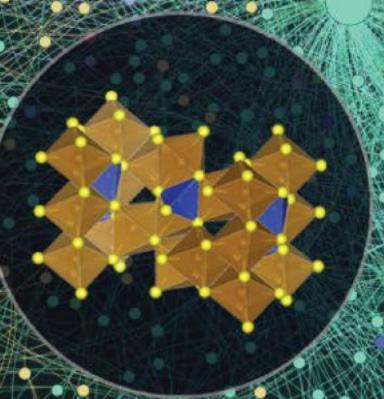


ML for HEAs



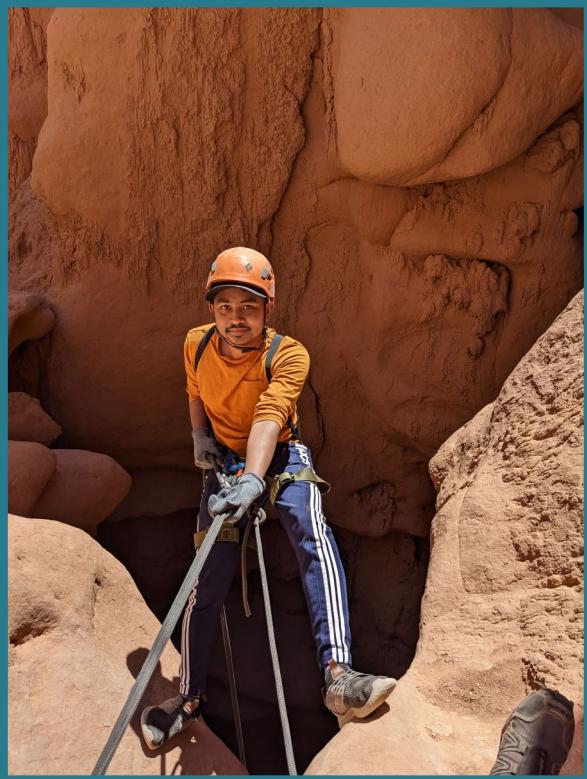
Special thanks to my awesome graduate students who helped prepare this material!



Sparks Group



Trupti Mohanty

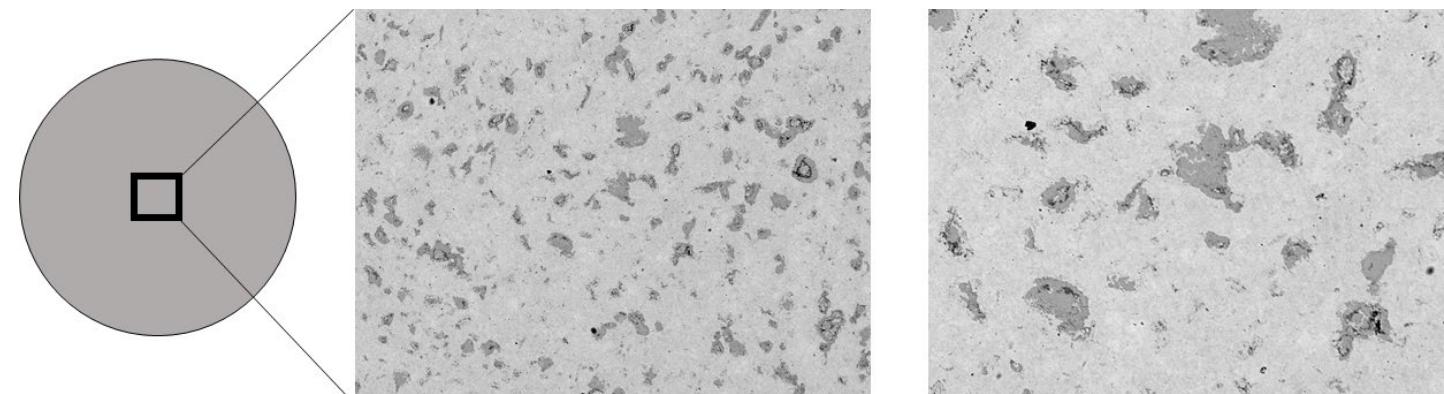
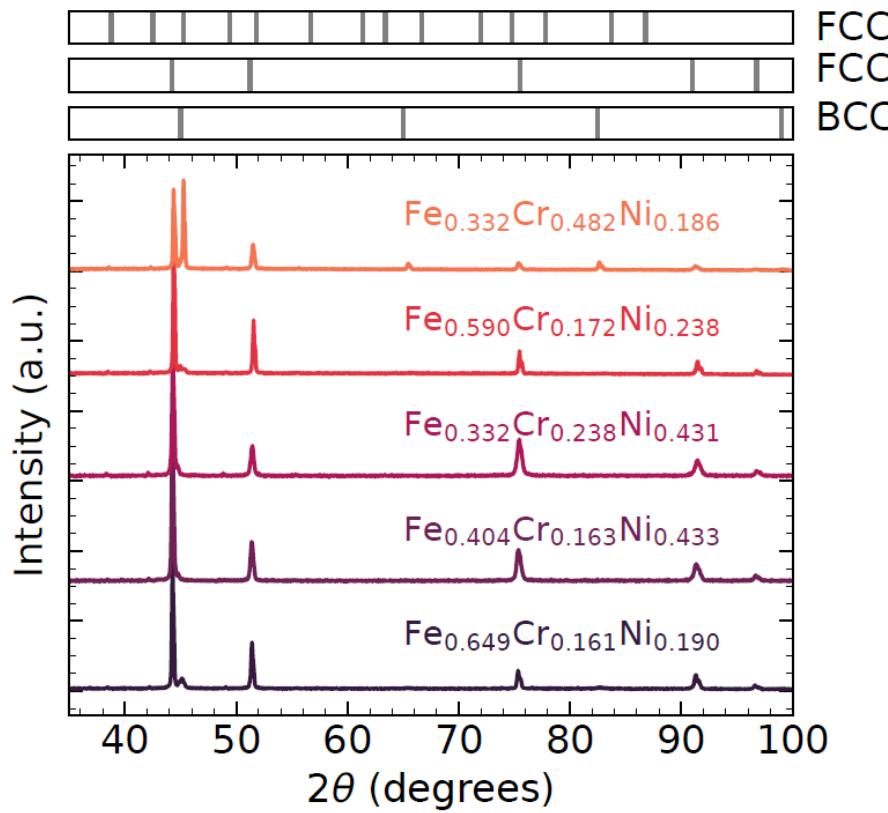


Hasan Sayeed

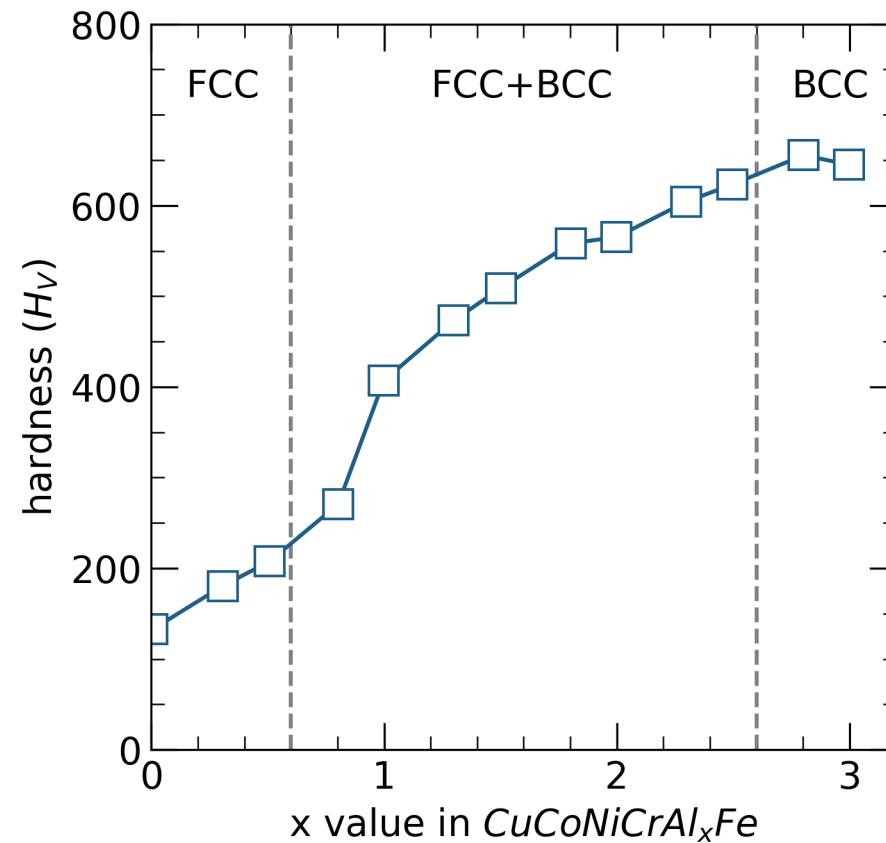


Ramsey Issa

HEAs often consist of multiple phases



Since phases impact properties, can we use ML to predict the phases ahead of time?



Yeh et al Nanostructured high-entropy alloys with multiple principal elements: novel alloy design concepts and outcomes, Adv Eng Mater, 6 (5) (2004), pp. 299-303

ML Models	Classification Target	Performance and Dataset	Ref
NN	AM, SS, IM	NN 80% accuracy 118 MPEA dataset	Islam et al Comp Mat Sci, 2018, DOI
SVM, KNN , ANN	SS , SS+IM, IM	ANN with 74.3% accuracy, 401 dataset : 174 SS, 54 IM, and 173 SS + IM phases.	Huang et al Acta Mat 2019, DOI
SVM	FCC, BCC, NSP (Not forming Solid Solution)	SVM 90% accuracy 322 data samples	Li & Guo Phys Rev Mat 2019, DOI
Model and Feature Selection with GA	Task 1 : SS and NSS Task 2 : FCC / BCC/ Dual Phase	SVM 88.7 % accuracy Task 1 NN 91.3% accuracy Task 2 550 samples	Zhang et al Acta Mat 2020, DOI
DNN with GAN	SS, IM, SS+IM, AM	DNN + GAN 93.17% 989 dataset + 600 samples generated using GAN	Lee et al Mat Design 2020, DOI
Gradient Boost	SS and Non SS	GB 96.4% accuracy 1807 data	Yan et al Comp Mat Sci 2021, DOI

Machine learning can do basic phase prediction

Task:	Classification of amorphous vs single phase solid solution vs intermetallic phases
Data:	Guo and Li's 2011 Prog Nat Sci: Mat Int. 118 MPEAs (33 AM, 64 SS, 21 IM)
Models and features:	Neural network: (5,10,10,3), ReLU VEC , $\Delta\chi$, δ , ΔH_{mix} , ΔS_{mix}
Findings:	"83% accuracy" overall
Code:	Not available 
Comment:	Unbalanced data, no augmentation, small sets for NN. No test set used, cross entropy used on validation dataset. Classes not balanced, just equally divided in 4 CV folds. No per class performance reported.



Computational Materials Science

Volume 150, July 2018, Pages 230-235



Machine learning for phase selection in multi-principal element alloys

Nusrat Islam ^a, Wenjiang Huang ^b, Houlong L. Zhuang ^a  

Domain knowledge for feature engineering is based on Hume-Rothery solubility rules

Hume-Rothery rules

From Wikipedia, the free encyclopedia

Hume-Rothery rules, named after [William Hume-Rothery](#), are a set of basic rules that describe the conditions under which an [element](#) could dissolve in a [metal](#), forming a [solid solution](#). There are two sets of rules; one refers to substitutional solid solutions, and the other refers to interstitial solid solutions.

Contents [hide]

- 1 Substitutional solid solution rules
- 2 Interstitial solid solution rules
- 3 Solid solution rules for multicomponent systems
- 4 See also
- 5 References
- 6 Further reading

Substitutional solid solution rules [edit]

For substitutional solid solutions, the Hume-Rothery rules are as follows:

1. The [atomic radius](#) of the solute and [solvent](#) atoms must differ by no more than 15%:^[1]

$$\% \text{ difference} = \left(\frac{r_{\text{solute}} - r_{\text{solvent}}}{r_{\text{solvent}}} \right) \times 100\% \leq 15\%.$$

2. The [crystal structures](#) of solute and solvent must be similar.
3. Complete [solubility](#) occurs when the solvent and solute have the same [valency](#).^[2] A metal is more likely to dissolve a metal of higher valency, than vice versa.^[3]^[4]^[5]
4. The solute and solvent should have similar [electronegativity](#). If the electronegativity difference is too great, the metals tend to form [intermetallic compounds](#) instead of solid solutions.

Interstitial solid solution rules [edit]

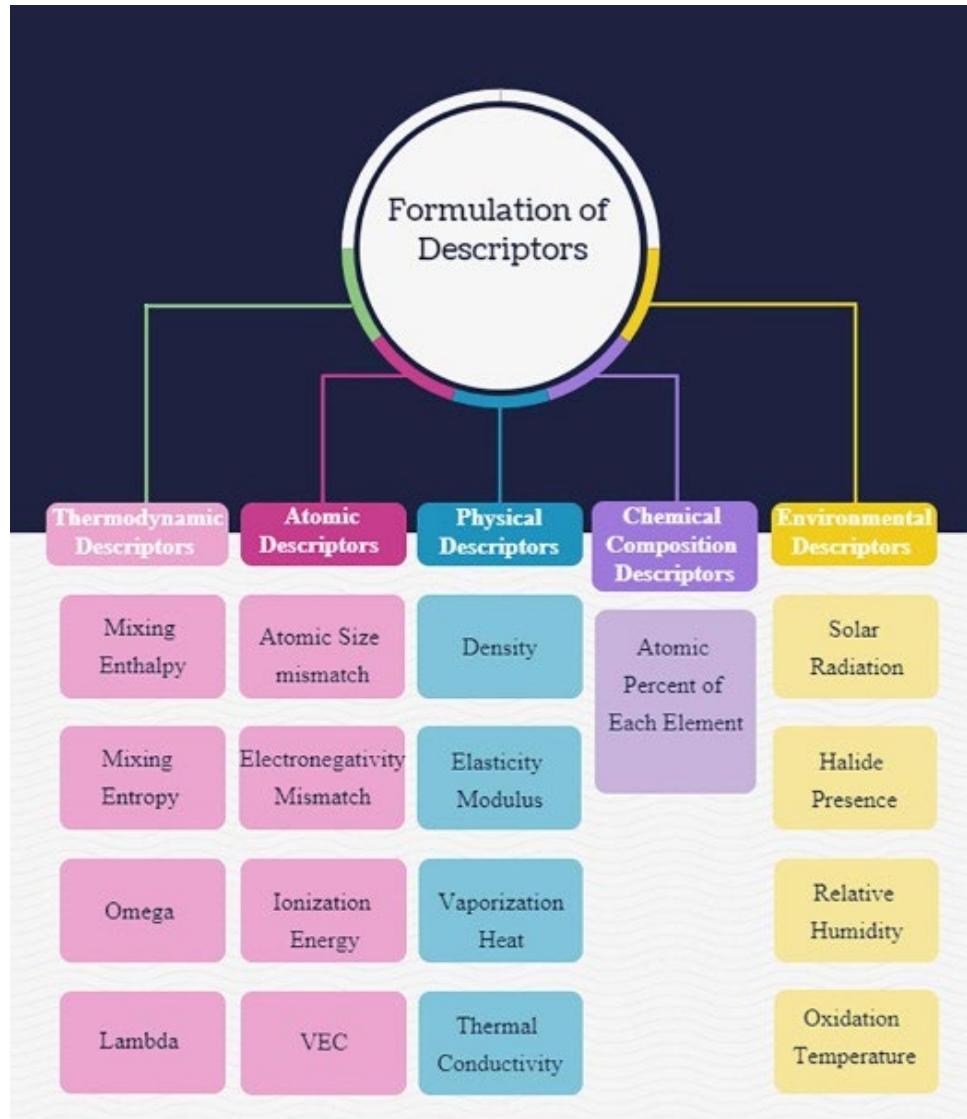
For [interstitial](#) solid solutions, the Hume-Rothery Rules are:

1. Solute atoms should have a smaller radius than 59% of the radius of solvent atoms.^[6]^[7]
2. The solute and solvent should have similar [electronegativity](#).^[8]
3. Valency factor: two elements should have the same valence. The greater the difference in valence between solute and solvent atoms, the lower the solubility.

Solid solution rules for multicomponent systems [edit]

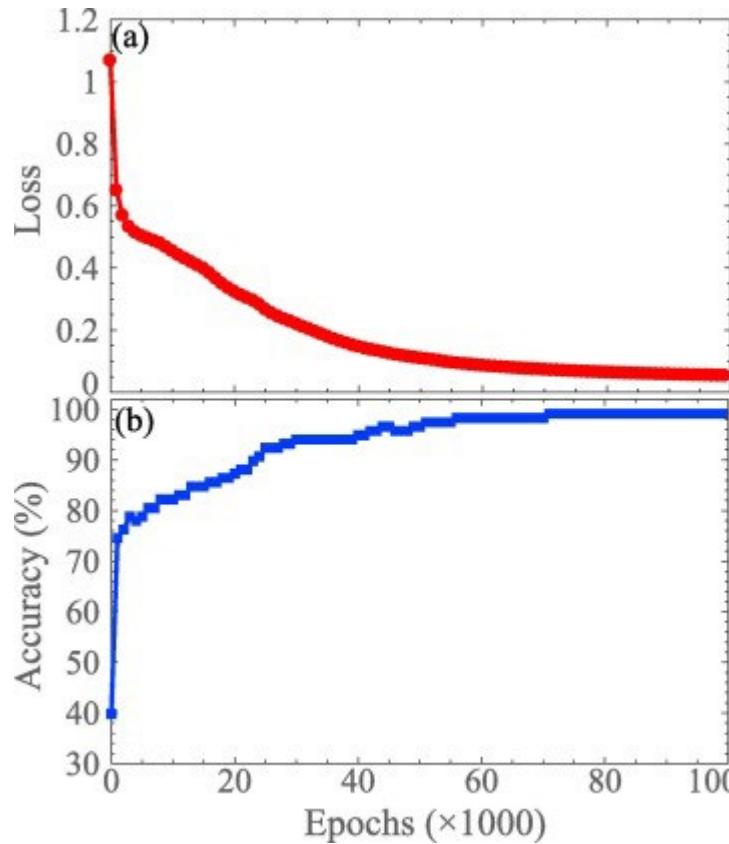
Fundamentally, the Hume-Rothery rules are restricted to binary systems that form either substitutional or interstitial solid solutions. However, this approach limits assessing advanced alloys which are commonly multicomponent systems. Free energy diagrams (or [phase diagrams](#)) offer in-depth knowledge of equilibrium restraints in complex systems. In essence the Hume-Rothery rules (and [Pauling's rules](#)) are based on geometrical restraints. Likewise are the advancements being done to the Hume-Rothery rules. Where they are being considered as critical contact criterion describable with [Voronoi diagrams](#).^[9] This could ease the theoretical phase diagram generation of multicomponent systems.

HEA features generally fall into the following categories



Roy and Balasubramanian, Predictive descriptors in machine learning and data-enabled explorations of high-entropy alloys. *Comp Mat Sci* (2021)

Remember, accuracy and loss are not the same thing!



$$\text{Cross-entropy} = - \sum_{i=1}^n \sum_{j=1}^m y_{i,j} \log(p_{i,j})$$

$$\text{Accuracy} = \frac{\text{No of correct predictions}}{\text{Total no of predictions}}$$

1	Ground Truth			Prediction			Cross-entropy	Accuracy
	A	B	C	P(A)	P(B)	P(C)		
Sample 1	1	0	0	0.7	0.15	0.15	0.357	1
Sample 2	1	0	0	0.7	0.15	0.15	0.357	1
Sample 3	1	0	0	0.33	0.33	0.34	1.109	0

$$\text{Loss: } -(\log(0.70) + \log(0.70) + \log(0.33)) = 1.82$$

$$\text{Accuracy: } (1+1+0)/3 = 66.67\%$$

2	Ground Truth			Prediction			Cross-entropy	Accuracy
	A	B	C	P(A)	P(B)	P(C)		
Sample 1	1	0	0	0.5	0.25	0.25	0.693	1
Sample 2	1	0	0	0.5	0.25	0.25	0.693	1
Sample 3	1	0	0	0.5	0.25	0.25	0.693	1

$$\text{Loss: } -(1*(\log(0.5) + \log(0.5) + \log(0.5)) + 0*(0.25 + 0.25 \dots \text{six times})) = 2.07$$

$$\text{Accuracy: } (1+1+1)/3 = 100\%$$

[https://kharshit.github.io/blog/2018/12/07/loss-vs-accuracy#:~:text=Cross%2Dentropy%20loss%20awards%20lower,0%20\(for%20false%20ones\).](https://kharshit.github.io/blog/2018/12/07/loss-vs-accuracy#:~:text=Cross%2Dentropy%20loss%20awards%20lower,0%20(for%20false%20ones).)

If your classes are highly imbalanced, the metrics can fool you!

	$\text{Ca}(\text{Ca}_{0.5}\text{Nd}_{0.5})_2\text{NbO}_6$	$\text{Ca}_2\text{Nb}_2\text{O}_7$	CaTiO_3	CeAl_2Ga_2	Cu	CuZrSiAs	FeAs	GdFeO_3	K_2NiF_4	LaAlO_3	MgAl_2O_4	MgCu_2	NaCl	NaFeO_2	TiNiSi	Other	Recall
$\text{Ca}(\text{Ca}_{0.5}\text{Nd}_{0.5})_2\text{NbO}_6$	94	0	0	0	0	0	0	0	0	0	0	0	0	0	0	43	0.686
$\text{Ca}_2\text{Nb}_2\text{O}_7$	0	95	0	0	0	0	0	0	0	0	0	0	0	0	0	50	0.655
CaTiO_3	0	0	133	0	0	0	0	12	0	5	0	0	0	0	1	105	0.522
CeAl_2Ga_2	0	0	0	161	0	0	0	0	0	0	0	0	0	0	0	28	0.847
Cu	0	0	0	0	56	0	0	0	0	0	0	0	0	0	0	70	0.444
CuZrSiAs	0	0	0	0	0	93	0	0	0	0	0	0	0	0	0	21	0.816
FeAs	0	0	0	0	0	0	88	0	0	0	0	0	0	0	0	15	0.854
GdFeO_3	0	0	9	0	0	0	0	454	0	19	0	0	1	0	0	120	0.753
K_2NiF_4	0	0	0	0	0	0	0	3	81	2	0	0	0	0	0	56	0.570
LaAlO_3	0	0	2	0	0	0	0	33	1	92	0	0	0	0	0	27	0.594
MgAl_2O_4	0	0	0	0	0	0	0	0	0	0	315	0	0	0	0	69	0.820
MgCu_2	0	0	0	0	0	0	0	0	0	0	0	58	0	0	0	53	0.523
NaCl	0	0	0	0	0	0	0	1	0	0	1	0	140	1	0	81	0.625
NaFeO_2	0	0	0	0	0	0	0	0	0	0	0	0	0	105	0	34	0.755
TiNiSi	0	0	0	1	0	0	3	0	0	0	0	0	0	0	45	65	0.395
Other	0	5	29	5	37	2	18	59	21	16	31	14	21	12	8	20984	0.986
Total	105	100	173	167	93	95	109	562	103	134	103	72	162	118	54	21821	0.950
Precision	0.895	0.950	0.769	0.964	0.602	0.979	0.807	0.808	0.786	0.687	0.908	0.806	0.864	0.890	0.833	0.962	

Graser et al Machine learning and energy minimization approaches for crystal structure predictions: a review and new horizons, Chem Mater, 30 (11), (2018) 3601-3612

Expanding approach to other models is useful

Task:	Classification of single phase solid solution vs intermetallic phase vs mixed
Data:	Miracle and Senkov's 2017 Acta Mater 648 curated (duplicates) to 401 (174 SS, 54 IM, 173 mixed)
Models and features:	KNN: k value 1-10, fine and weighted SVM: 1, 2, n-dimensional Gaussian kernel Neural network: (5,20,15,25,3), ReLU VEC , $\Delta\chi$, δ , ΔH_{mix} , ΔS_{mix}
Findings:	KNN 68% accuracy SVM 64% NN 74%
Code:	Not available  , used Matlab 
Comment:	Unbalanced data, no augmentation, small sets for NN. No test set used, cross entropy used on validation dataset. Classes not balanced, just equally divided in 4 CV folds. No per class performance reported. Break problem into binaries and slightly better Feature ablation ($\delta > VEC > \Delta S_{mix} > \Delta H_{mix} > \Delta\chi$)



Acta Materialia
Volume 169, 1 May 2019, Pages 225-236



Full length article

Machine-learning phase prediction of high-entropy alloys

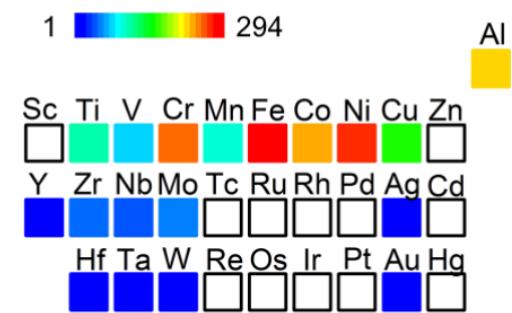
Wenjiang Huang, Pedro Martin, Houlong L. Zhuang  

Dealing with imbalance and learning restricted generalizability

Task:	Classification of BCC vs FCC vs NSP (no formation of solid solution)
Data:	Miracle and Senkov's 2017 Acta Mater 648 curated to 322 (18 BCC, 43 FCC, 281 mixed)
Models and features:	SVM: radial basis function kernel VEC, var χ , var δ , ΔH_{mix} , ΔS_{mix} , T_m
Findings:	SVM 90%, 31 new refractory HEA, validation (11 exp, 20 DFT)
Code:	Not available 
Comment:	Unbalanced data, unbalanced elements. 10% test set used, cross entropy used on validation dataset. SMOTE oversampling to balance classes (261 each) Per class performance reported. ΔH_{mix} from DFT Only consider as cast (ignore processing) Exclude REE and light elements (except Al), no HCP b/c REE Feature importance ($VEC > \text{var } \delta > \text{rest}$), $\text{var } \chi$ not helpful



Machine-learning model for predicting phase formations of high-entropy alloys
Yao Li and Wanlin Guo
Phys. Rev. Materials **3**, 095005 – Published 20 September 2019



Genetic algorithms can be used to select optimal models and features

Task:	Task 1: NSS and SS Task 2: FCC vs BCC vs mixed
Data:	???? 550 samples from literature
Models and features:	GA: natural selection of many simple models 70 features including: VEC, Z, r, period, group, χ , C_p , ΔH_{mix} , ΔS_{mix} , T_m , DFT data(Ω , E_{gap} , $E_{cohesive}$, dielectric, phonon, etc)
Findings:	Task 1: 88.7 % accuracy Task 2: NN 91.3% accuracy
Code:	Data not available / Data will be made available on request 
Comment:	10% test set No citation of where data even came from  Complicated and time consuming GA for modest improvement that may only be overfitting

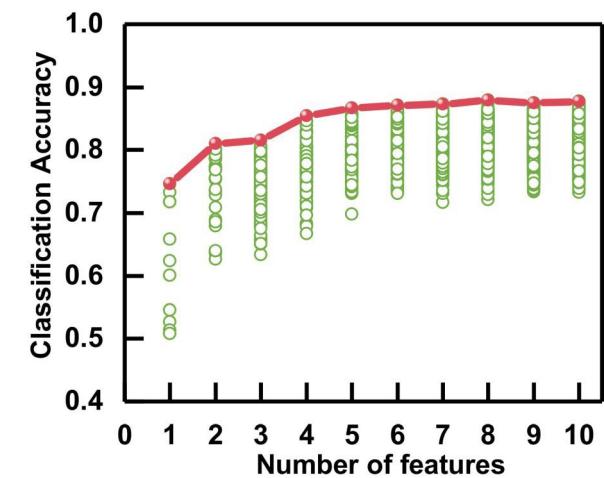


Acta Materialia
Volume 185, 15 February 2020, Pages 528-539



Phase prediction in high entropy alloys with a rational selection of materials descriptors and machine learning models

Yan Zhang ^{a, b}, Cheng Wen ^{a, b}, Changxin Wang ^{a, b}, Stoichko Antonov ^{a, c}, Dezhen Xue ^{a, d}✉, Yang Bai ^{a, b}, Yanjing Su ^{a, b}✉



Common features can be assessed and discussed in context of task

Mixing Entropy

$$\Delta S_{mix} = -R \sum_{i=1}^n C_i \ln C_i$$

Mixing Enthalpy

$$\Delta H_{mix} = \sum_{i=1, i < j}^n 4 H_{mix}^{ij} C_i C_j$$

Average VEC

$$VEC = \sum_{i=1}^n C_i (VEC)_i$$

VEC Difference

$$\delta_{VEC} = \sqrt{\sum_{i=1}^n C_i \left(1 - \frac{(VEC)_i}{VEC}\right)^2}$$

Average Electronegativity

$$\chi_{arg} = \sum_{i=1}^n C_i \chi_i$$

Electronegativity Difference

$$\delta_\chi = \sqrt{\sum_{i=1}^n C_i \left(1 - \frac{\chi_i}{\chi_{arg}}\right)^2}$$

Atomic Radius Difference

$$\delta_r = \sqrt{\sum_{i=1}^n C_i \left(1 - \frac{r_i}{\bar{r}}\right)^2}$$

Melting Point Difference

$$\delta_T = \sqrt{\sum_{i=1}^n C_i \left(1 - \frac{T_i}{\bar{T}_m}\right)^2}$$

Ω Parameter

$$\Omega = \frac{T_m \Delta S_{mix}}{|\Delta H_{mix}|}$$

Geometrical Parameter

$$\lambda = \frac{\Delta S_{mix}}{\delta_r^2}$$

Genetic algorithms rely on operators such as mutation, crossover and selection

Population of 100 “solutions”

Fitness determined by average of 100 CV accuracies

Tournaments run 100 times between 2 parents selected from population. Higher accuracy is selected

Crossover: randomly choose two materials descriptor subsets from a preselected 80% of the population as parents.

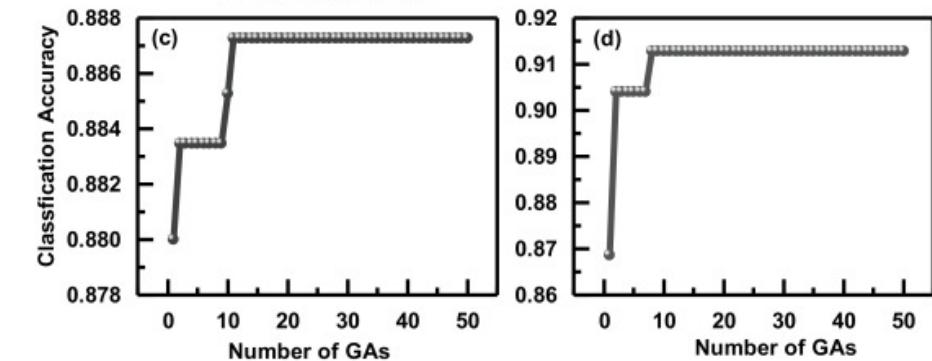
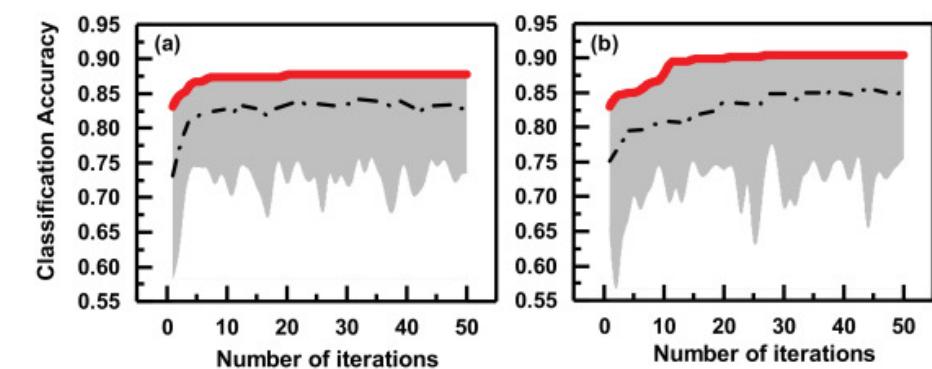
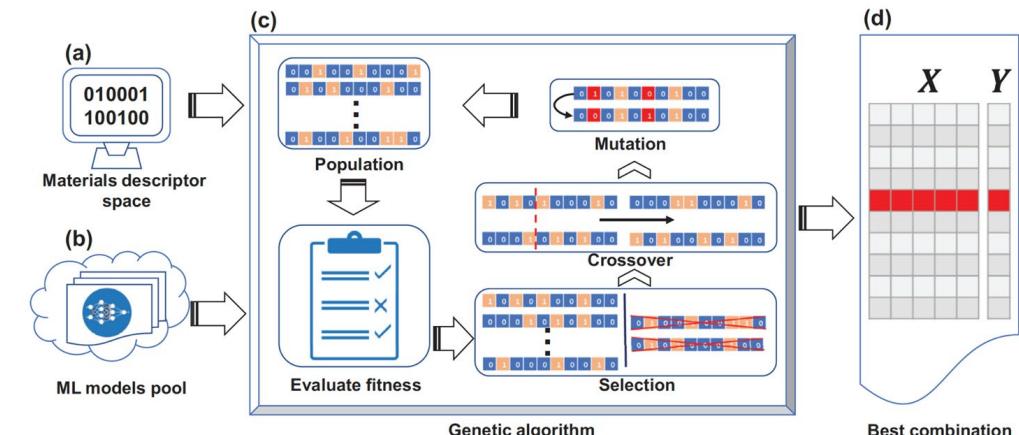
Each bit chosen equal probability from either parent.

Retain 100 new children from among those with 4 or fewer descriptors.

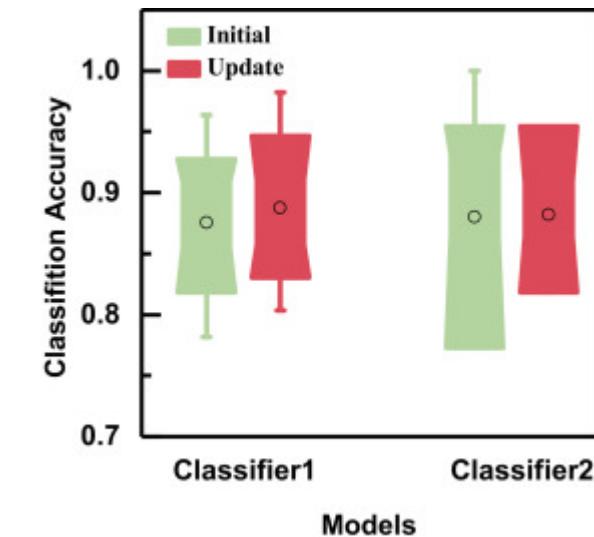
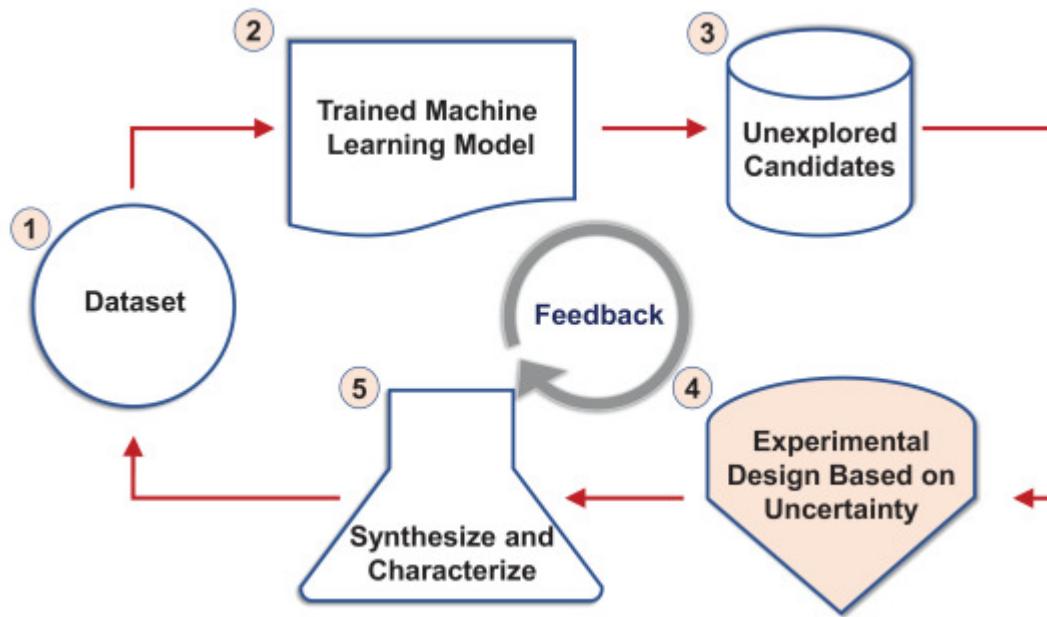
Mutation: use bit flip mutation on 1% of genes to switch them “on/off.” Filter to max 4 descriptors

Reevaluate accuracies and repeat selection, crossover, mutation.

Complete 50 GA runs on each ML model.



An active learning framework was used with bootstrapped uncertainty



Original training data
+ 10 new samples from active learning

Increasing the data size sufficiently allows for us to extract deep learning benefit

Task:	Classification of solid solution, intermetallic, mixed, or amorphous
Data:	Tsai 2019 Metals, Huang 2019 Acta Mat, Miracle and Senkov's 2017 Acta Mater, Zhou 2019 npj Comp Mat 989 after duplicate curation (250 SS, 267 SS+IM, 307 IM, 165 AM)
Models and features:	DNN: (13,150,128,80,30,16,3), dropout 0.3, batch 15 GAN: Gen (44,64,207,128,36,13), Disc (17,64,100,50,14,1) VEC, var χ , var δ , ΔH_{mix} , ΔS_{mix} , stdev($T_m, E, \Delta H_{mix}, \Delta \chi, K$)
Findings:	DNN 84% accuracy vs 64-72% from classic models
Code:	The data used in this manuscript are available from the corresponding author upon reasonable request. 😊
Comment:	Bayesian hyperparameter tuning L2 regularization, dropout used to mitigate overfitting Batches < 30 for stability Data augmentation w/con-GAN, accuracy (84%→93%) Feature weighting from layerwise relevance propagation



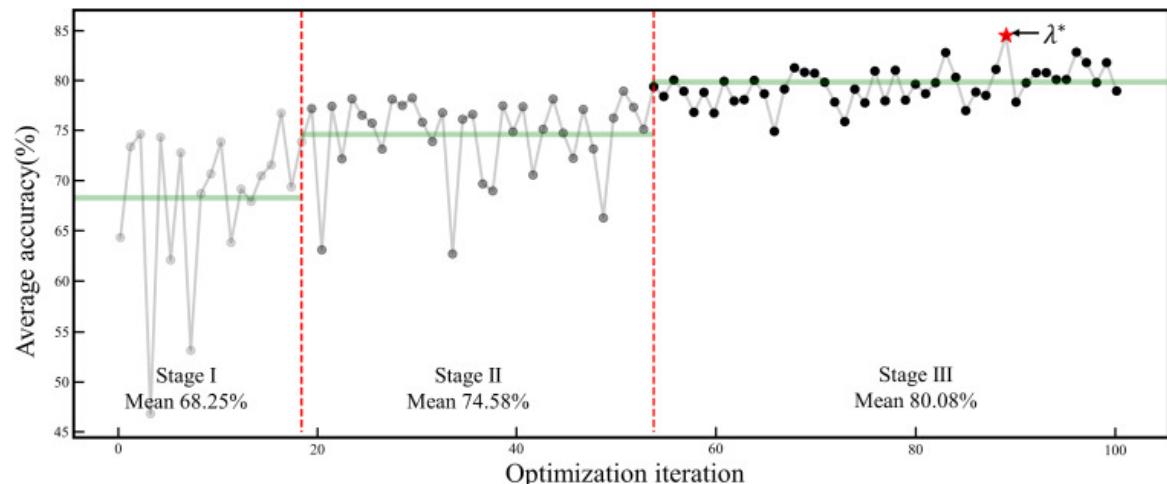
Materials & Design
Volume 197, 1 January 2021, 109260



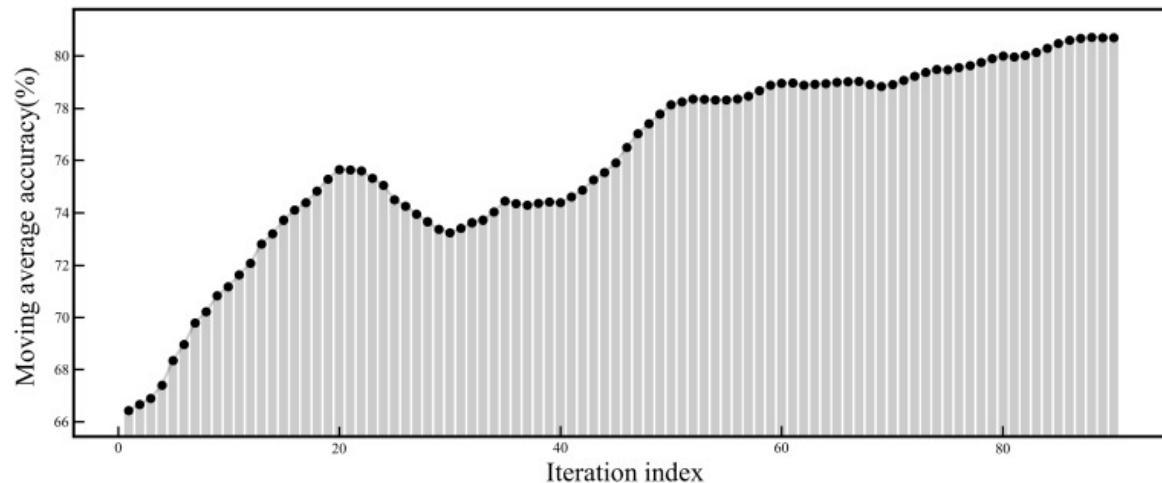
Deep learning-based phase prediction of high-entropy alloys: Optimization, generation, and explanation

Soo Young Lee ^{a, 1}, Seokyeong Byeon ^{a, 1}, Hyoung Seop Kim ^{b, c, d}, Hyungyu Jin ^a✉, Seungchul Lee ^{a, e}✉

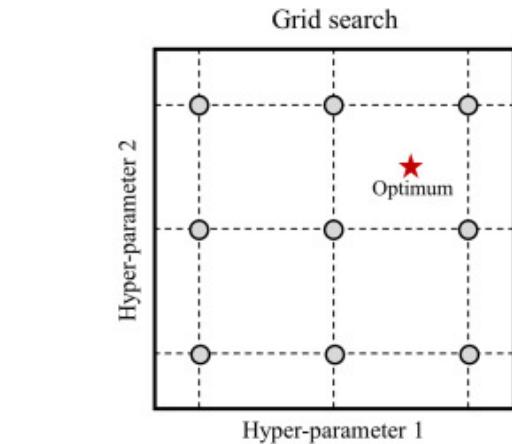
Tune the neural network using Bayesian optimization



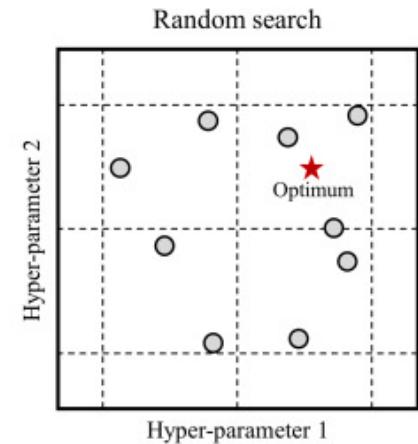
(a)



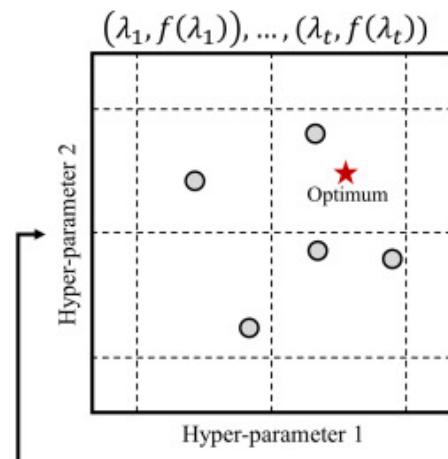
(b)



Grid search

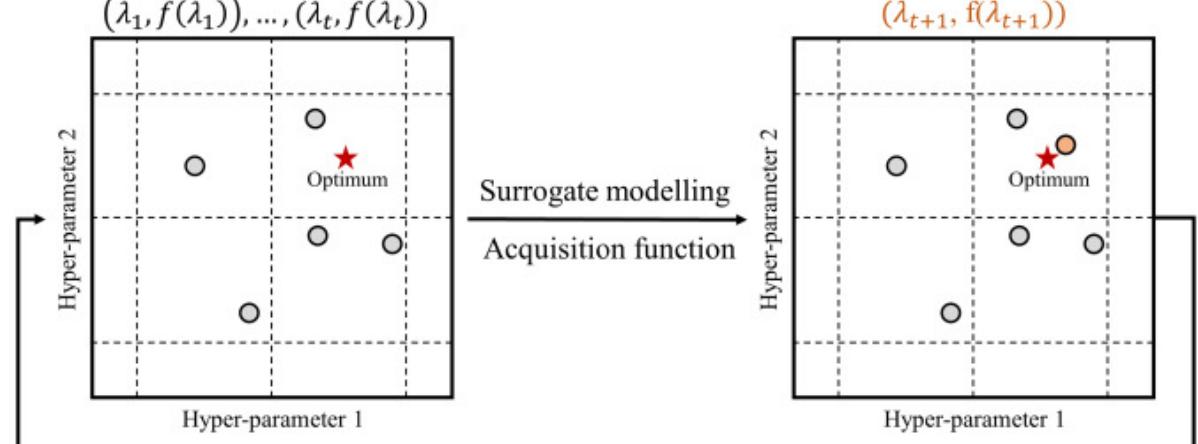


Random search



$(\lambda_1, f(\lambda_1)), \dots, (\lambda_t, f(\lambda_t))$

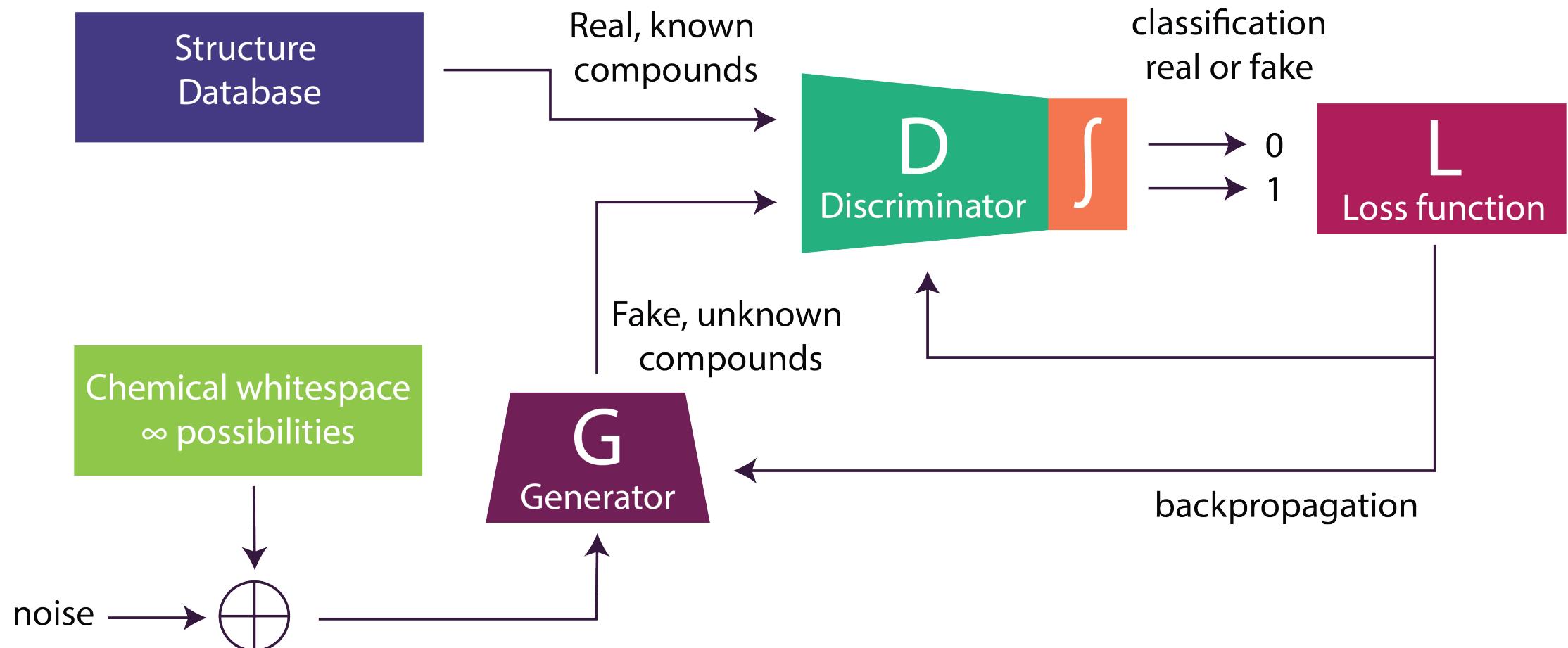
(a)



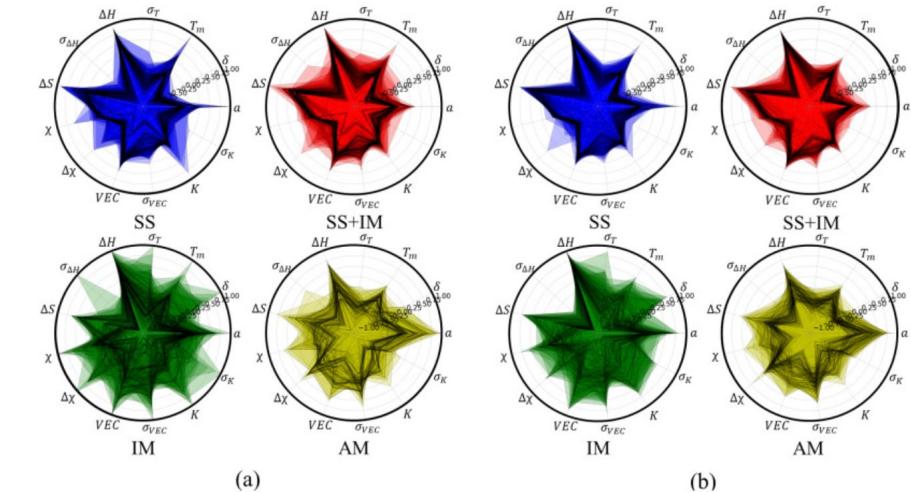
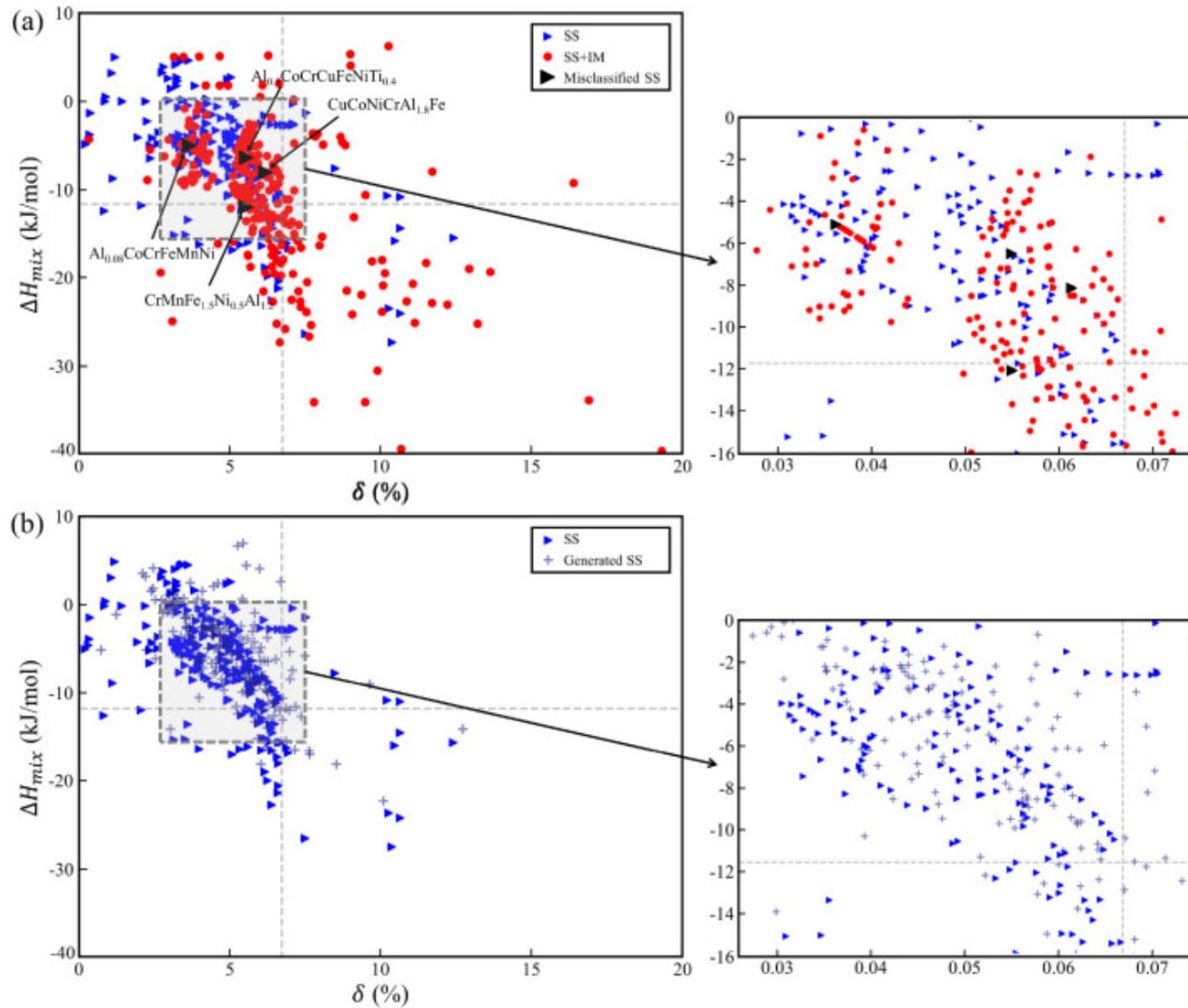
$(\lambda_{t+1}, f(\lambda_{t+1}))$

(b)

Generative adversarial networks are a type of generative machine learning model



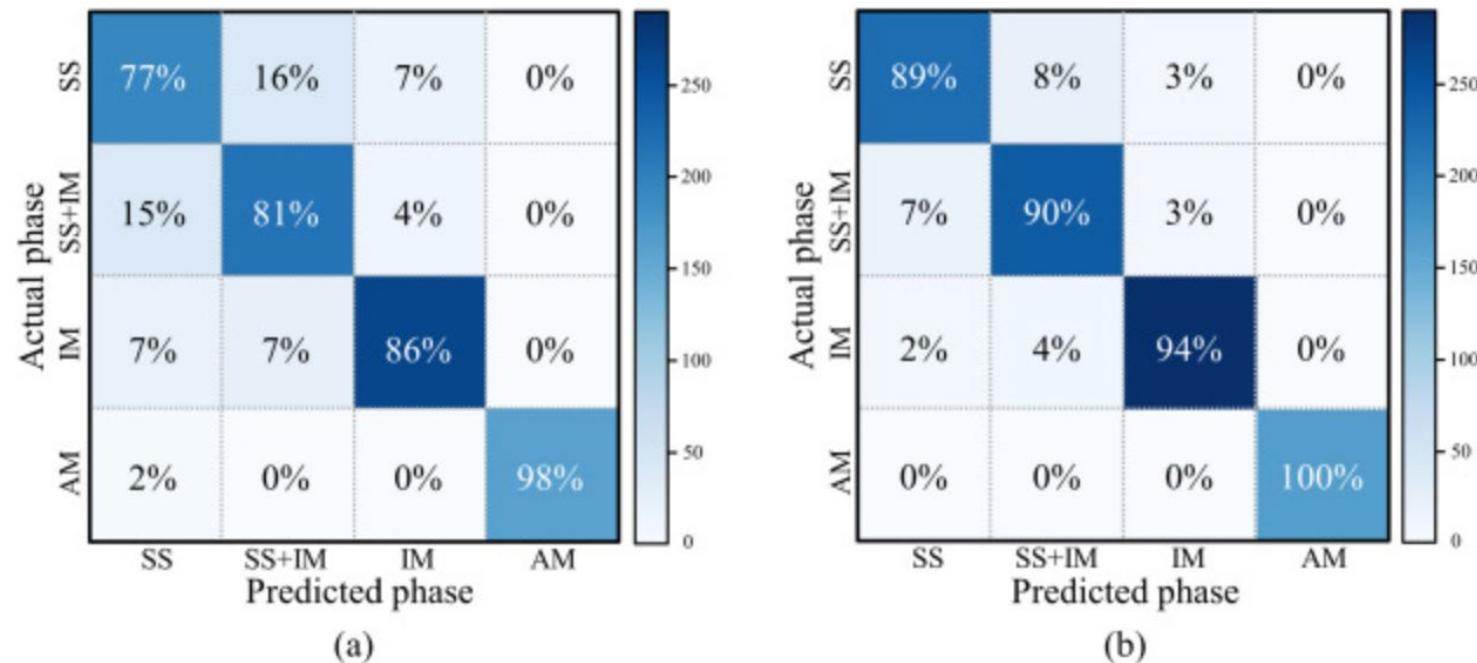
Conditional generative models can be used to augment data



Prediction model	Augmented samples per phase	Average test accuracy
DNN-BO	—	84.75%
DNN-Augment25	25	86.41%
DNN-Augment50	50	90.29%
DNN-Augment100	100	91.78%
DNN-Augment150	150	93.17%
DNN-Augment200	200	91.34%

Conditional generative models can be used to augment data

250SS
267 SS+IM
307 IM
165 AM



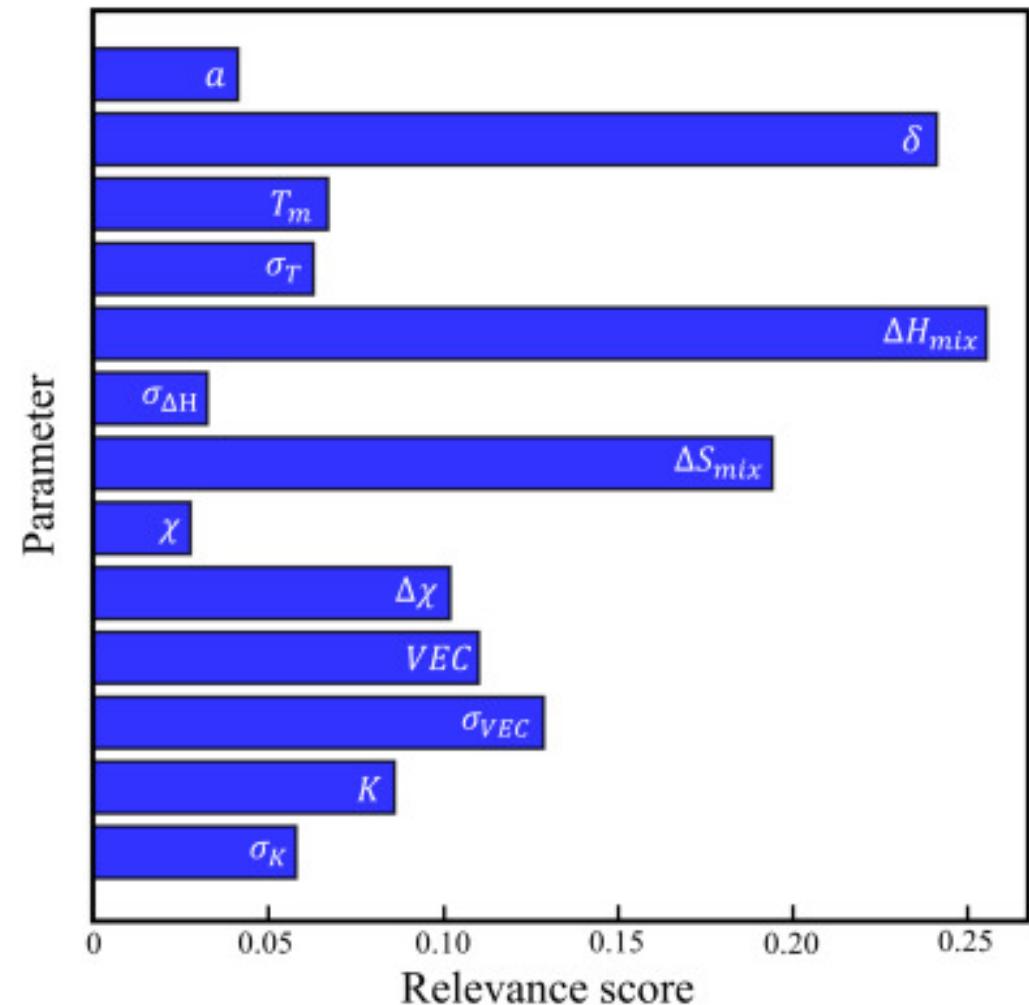
Prediction model	Augmented samples per phase	Average test accuracy
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DNN-Augment100	100	91.78%
DNN-Augment150	150	93.17%
DNN-Augment200	200	91.34%

Conditional generative models can be used to generate future compositions

Validation HEAs	Actual phase	Predicted phase	Softmax output			
			SS	SS + IM	IM	AM
Fe _{4.5} Co ₃ CrVMn _{0.5} [54]	SS	SS	9.04e-1	9.50e-2	1.04e-5	4.14e-5
CoCu ₂ Mn ₃ Ni ₄ [55]	SS	SS	9.99e-1	5.70e-4	8.43e-5	2.21e-4
Al _{0.75} Cu _{3.0833} Fe _{3.0833} Mn _{3.0833} [56]	SS	SS	8.76e-1	1.23e-1	2.03e-4	5.04e-5
Cu _{3.33} Fe _{3.33} Mn _{3.33} [56]	SS	SS	9.99e-1	5.15e-5	8.98e-6	2.95e-6
Co _{1.75} Cr _{1.25} Fe _{5.5} NiMo _{0.3} C _{0.2} [57]	SS + IM	SS + IM	2.45e-2	9.74e-1	6.87e-4	5.19e-5

Feature weighting also possible in deep learning via layerwise relevance propagation

Features	Equations
Mean atomic radius	$a = \sum_i c_i \cdot r_i$
Atomic size difference	$\delta = \sqrt{\sum_i \left(c_i \cdot \left(1 - \frac{r_i}{\sum_i c_i \cdot r_i} \right)^2 \right)}$
Average melting temperature	$T_m = \sum_i c_i \cdot T_{mi}$
Standard deviation of melting temperature	$\sigma_T = \sqrt{\sum_i c_i \cdot \left(1 - \frac{T_{mi}}{T_m} \right)^2}$
Mixing enthalpy	$\Delta H_{mix} = 4 \sum_{i \neq j} c_i \cdot c_j \cdot H_{ij}$
Standard deviation of mixing enthalpy	$\sigma_{\Delta H} = \sqrt{\sum_{i \neq j} c_i \cdot c_j \cdot (H_{ij} - \Delta H_{mix})^2}$
Mixing entropy	$\Delta S_{mix} = -R \sum_i c_i \cdot \log c_i$
Electronegativity	$\chi = \sum_i c_i \cdot \chi_i$
Standard deviation of electronegativity	$\Delta \chi = \sqrt{\sum_i \left(c_i (\chi_i - \bar{\chi})^2 \right)}$
Valence electron concentration	$VEC = \sum_i c_i \cdot VEC_i$
Standard deviation of VEC	$\sigma_{VEC} = \sqrt{\sum_i \left(c_i \cdot (VEC_i - \bar{VEC})^2 \right)}$
Mean bulk modulus	$K = \sum_i c_i \cdot K_i$
Standard deviation of bulk modulus	$\sigma_K = \sqrt{\sum_i c_i \cdot (K_i - \bar{K})^2}$



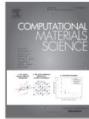
c_i is the stoichiometric ratio of i -th component of alloy, H_{ij} is binary mixing enthalpy in liquid phase, and R is [ideal gas constant](#), respectively.

Emphasis can be tuned to subsets of HEAs such as RHEAs

Task:	Classification of single phase solid solution vs non-single phase alloys
Data:	"historical literature" (not cited) 1807 (800 SS, 1000 NSS)
Models and features:	Tested 9 classical models, settled on GB $VEC, T_m, \Delta H_{mix}, \Delta\chi, \delta, \Delta S_{mix}, V_m$
Findings:	Gradient boosting 97% CV train, 96% test
Code:	The data and methods reported in this paper are available from the corresponding author upon reasonable request. 
Comment:	20% test set used Emphasis on oxidation resistant HEAs (RHEAs) Ti, Zr, Hf (Group IV), V, Nb, Ta (Group V), Cr, Mo, W (Group VI) No differentiation of phases (BCC vs FCC vs HCP etc) Predicted 100 new SS samples, experimentally validated 10 Feature score from permutation importance method



Computational Materials Science
Volume 199, November 2021, 110723



Accelerated discovery of single-phase refractory high entropy alloys assisted by machine learning

Yonggang Yan, Dan Lu, Kun Wang  

ML Models	Classification Target	Performance and Dataset	Ref
NN	AM, SS, IM	NN 80% accuracy 118 MPEA dataset	Islam et al Comp Mat Sci, 2018, DOI
SVM, KNN , ANN	SS , SS+IM, IM		Huang et al Acta Mat 2019, DOI
SVM		Code and Data! Balance of elemental constituents Beyond classification – how much of each phase? True validation – where does it fail? Extrapolation	Li & Guo Phys Rev Mat 2019, DOI
Model and Feature Selection with GA			Zhang et al Acta Mat 2020, DOI
DNN with GAN		DNN + GAN 93.17% 989 dataset + 600 samples generated using GAN	Lee et al Mat Design 2020, DOI
Gradient Boost	SS and Non SS	GB 96.4% accuracy 1807 data	Yan et al Comp Mat Sci 2021, DOI

What's missing in the literature so far?

Code and Data!
Balance of elemental constituents
Beyond classification – how much of each phase?
True validation – where does it fail?
Extrapolation

Both thermodynamic and empirical models are unsatisfying predictors

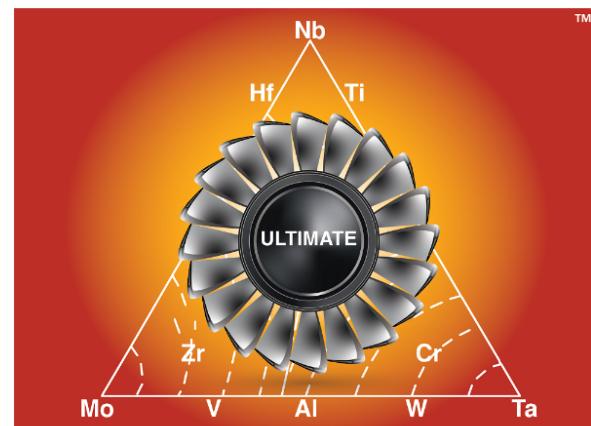
Numerous other studies, summarized elsewhere^{4,46,47,48}, have attempted to relate phase stability to parameters such as size misfits, valence electron concentrations, electronegativities and mixing enthalpies. However, many of these treatments ignore the formation enthalpies of competing compounds, typically intermetallic phases, which, in our view, limits their usefulness. The typical procedure is to construct empirical ‘phase stability maps’ as functions of the above parameters. ‘Critical’ values for the parameters are then estimated, above or below which certain types of microstructures (such as solid solutions) are stable. These values appear to be arbitrary and based on back-tested correlations, with little evidence of predictive capability. Other thermodynamically more consistent approaches^{49,50,51} have identified suitable compositional subspaces where single-phase HEAs might exist by additionally considering the enthalpies of competing intermetallic compounds. However, they also suffer from the drawback that ‘arbitrary’ values of formation enthalpies of competing compounds have to be used to ‘predict’ regions where single or multiple phases are stable.

Predicting phases isn't enough
We really want properties

Ultrahigh Temperature Impervious Materials Advancing Turbine Efficiency

- Manufacturing Efficiency
- Resource Efficiency
- Transportation Energy Conversion

[PRINT](#)



* Status:
Active

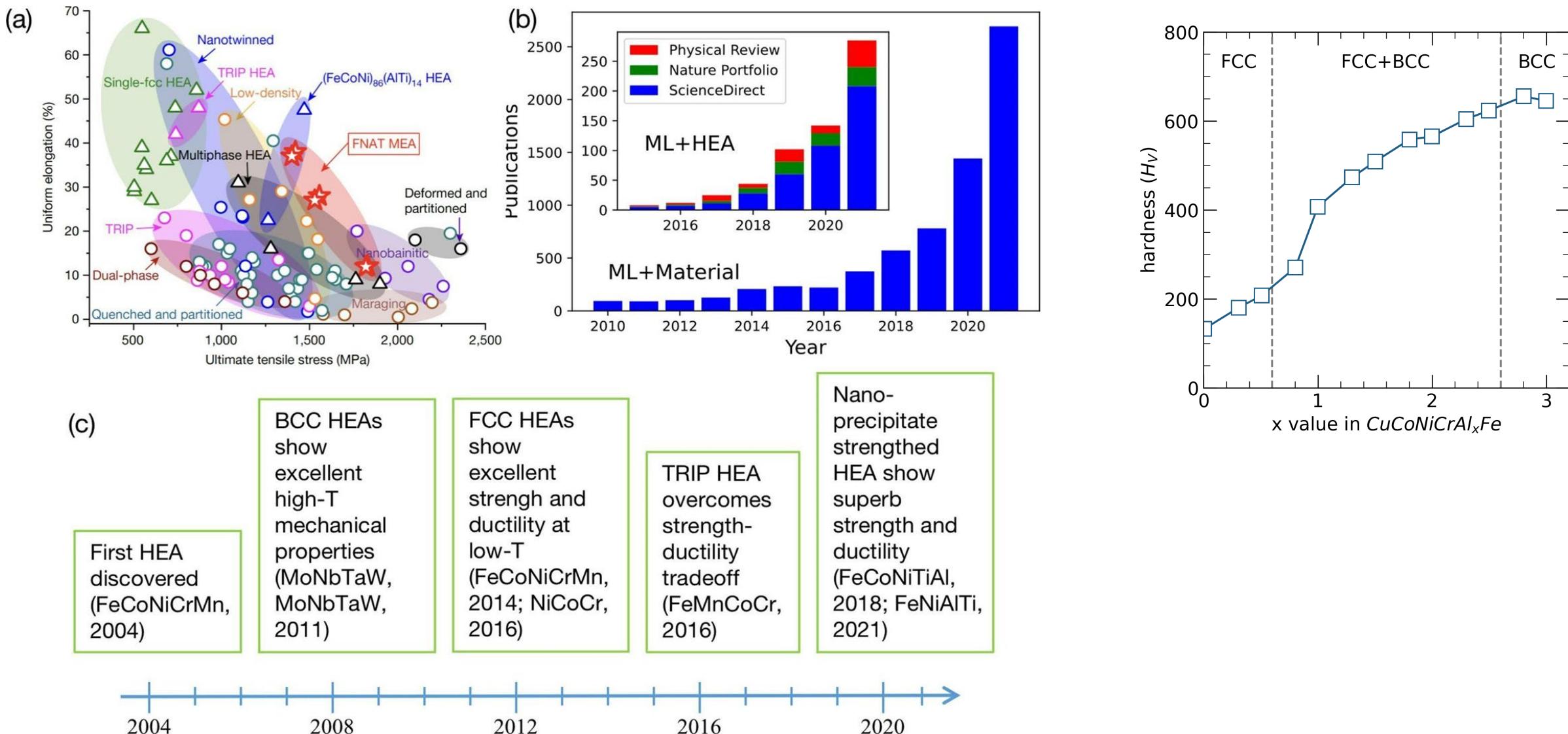
Release Date:
11/18/2020

Project Count:
16

Program Description:

The ULTIMATE program targets gas turbine applications in the power generation and aviation industries. ULTIMATE aims to develop ultrahigh temperature materials for gas turbines, enabling them to operate continuously at 1300 °C (2372 °F) in a stand-alone material test environment—or with coatings, enabling gas turbine inlet temperatures of 1800 °C (3272 °F) or higher. The successful materials must be able to withstand not only the highest temperature in a turbine but also the extreme stresses imposed on turbine blades. This program will concurrently develop manufacturing processes for turbine components using these materials, enabling complex geometries that can be seamlessly integrated in the system design. Environmental barrier coatings and thermal barrier coatings are within the scope of this program.

Properties vary widely in HEAs!



Dataset size	ML task	Model Accuracy	Ref
114	Rate constant of oxidation prediction	$R^2 = 0.92$	Bhattacharya et al. Oxid. Of Met. (2020) DOI
6,826	B and G prediction	< 5% error for B < 10% error for G predictions	Kim et al. Acta Mater. (2019) DOI
91	Hardness prediction	$R^2 = 0.94$ $MAE = 36$	Chang et al. JOM (2019) DOI
500*126*13	Predict stress-strain, texture, twinning volume fraction	$MSE < (10^{-3}, 10^{-2}, 10^{-4}$ respectively)	Gao et al. J Alloy Comp (2020) DOI
11,400	Stacking fault energy prediction	$RMSE = 0.57\text{--}2.76 \text{ mJ/m}^2$	Arora et al. Metals (2020) DOI
186	Experimental UTS prediction	$R^2 = 98.75$	Li et al. J. Mat. Sci. Tech. (2021) DOI

Some literature deserves skepticism

Task:	Predict rate constant of oxidation
Data:	"we build a dataset from 30 refs" not cited 114
Models and features:	GB, RF, KNN Composition, phases, time, temp, O ₂ & H ₂ O conc, atmosphere, mode
Findings:	GB R ² of 0.92 (no test set, no group by CV)
Code:	None 🛡
Comment:	Predictions as a function of temperature Composition, origin, balance of data set unassessed Data purely experimental Unclear if they had data leakage (interpolation within chemical families) due to lack of grouped CV No test set

 Springer Link

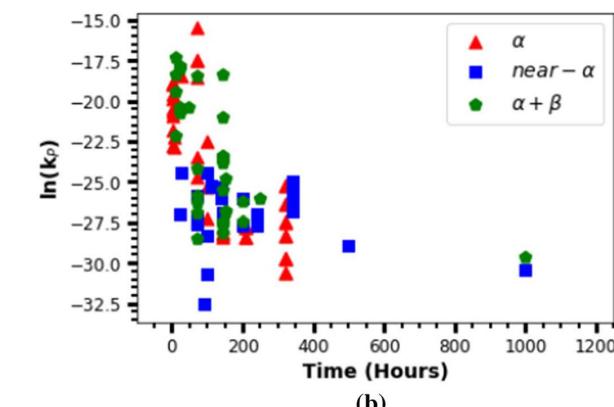
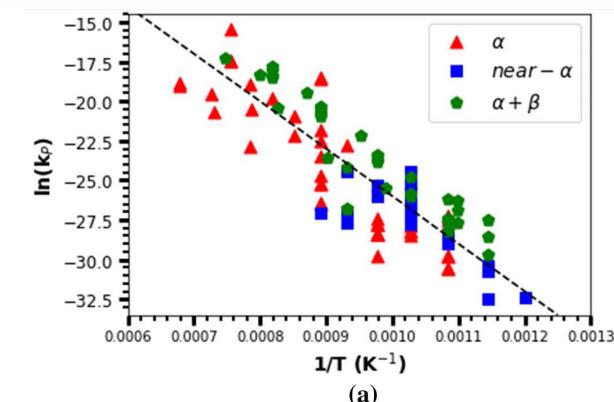
Original Paper | Published: 26 June 2020

Predicting the Parabolic Rate Constants of High-Temperature Oxidation of Ti Alloys Using Machine Learning

Somesh Kr. Bhattacharya , Ryoji Sahara & Takayuki Narushima

Oxidation of Metals 94, 205–218 (2020) | Cite this article

1135 Accesses | 8 Citations | Metrics



Can we train on non HEAs to then predict HEAs?

Task:	Predict B, G for HEA and use to calculate H_V
Data:	B, G for 6826 ordered inorganics from Materials Project
Models and features:	Gradient Boosted Trees Combinations of (Density, group #, δ, χ)
Findings:	< 5% error for B < 10% error for G
Code:	None 😊
Comment:	Neutron diffraction as input alongside DFT 20% test set (DFT data on inorganics from MP) Experimental validation (neutron diffraction) from one sample SHAP values for feature importance Use ML to learn feature – elastic constant relationships?



Acta Materialia

Volume 181, December 2019, Pages 124-138



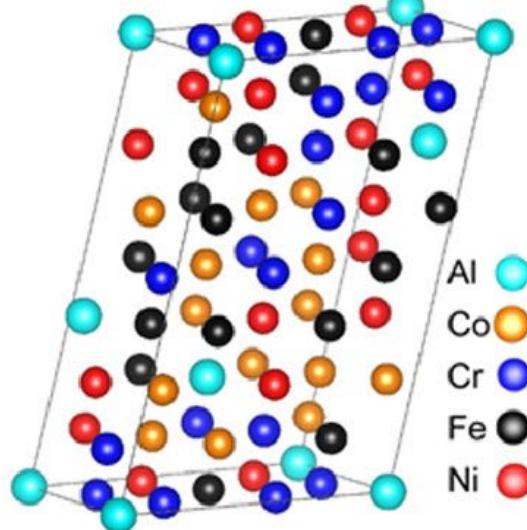
Full length article

First-principles and machine learning predictions of elasticity in severely lattice-distorted high-entropy alloys with experimental validation

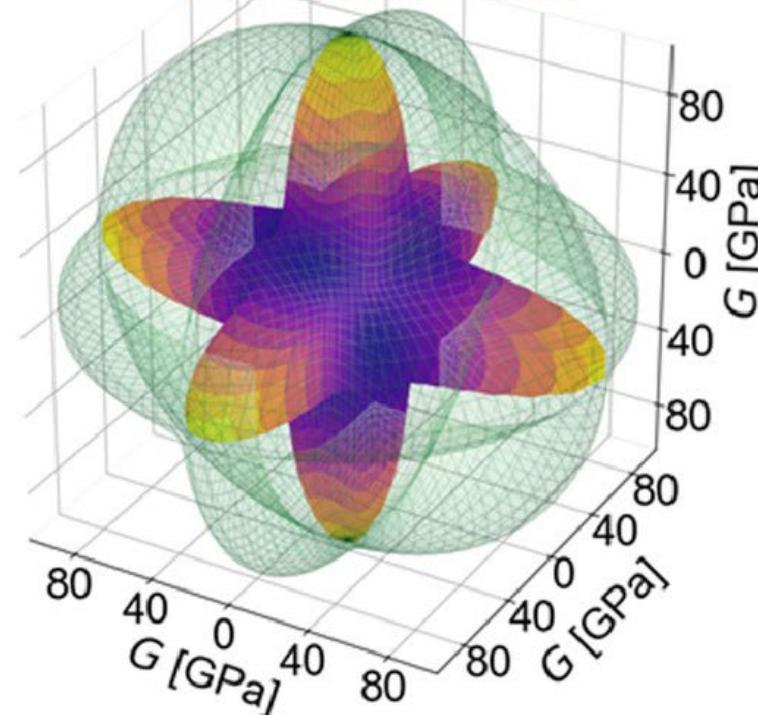
Kim, George ^a, Diao, Haoyan ^b, Lee, Chanho ^b, A.T. Samaei ^a, Phan, Tu ^a, de Jong, Maarten ^c, An, Ke ^d, Ma, Dong ^d, Liaw, Peter K. ^b, Chen, Wei ^a ☰

Simulations of disordered HEA was done with a large supercell (SQS)

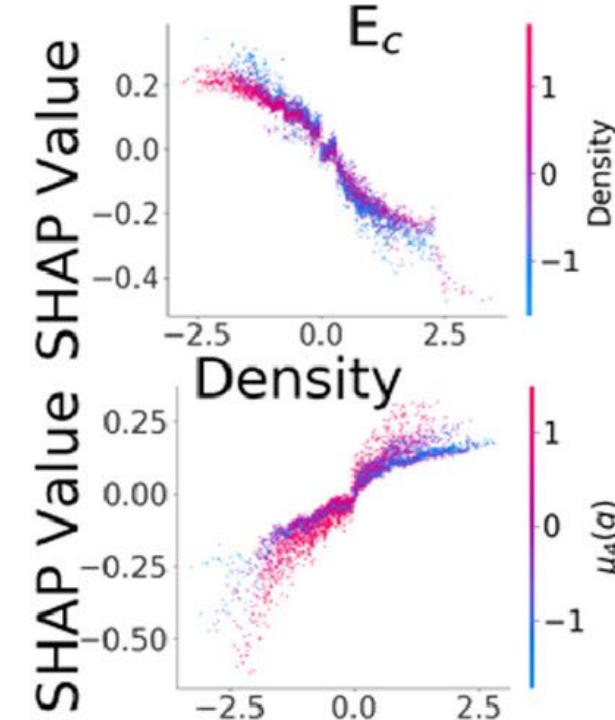
Special
Quasi-random
Structure



Elastic Moduli



Machine Learning



Can we really train on ordered compounds and then predict on disordered HEAs?

Effect of Al on experimental and calculated hardness values for the FCC $\text{Al}_x\text{CrCoFeNi}$ HEA. The experimentally-measured Vicker's hardness values are compared with calculated hardness values for increasing amounts of Al. Hardness increases with increasing the Al content in both calculated and measured values.

Composition	GB-trees predicted B (GPa)	GB-trees predicted G (GPa)	Calculated hardness (Hv)	Experimentally measured hardness (Hv)
$\text{Al}_{0.1}\text{CoCrFeNi}$	141.1	59.6	5.0	1.83 [108]
$\text{Al}_{0.3}\text{CoCrFeNi}$	161.5	73.2	6.8	3.4 [109]
$\text{Al}_{0.6}\text{CoCrFeNi}$	163.8	74.6	6.9	3.5 [109]

Comparison of the predicted elastic moduli of the $\text{Al}_{0.3}\text{CoCrFeNi}$ between the GBM-Locfit model and the GB Trees model.

	G (GPa)	B (GPa)
ND	81	172
GBM-Locfit (Benchmark)	85	208
GB-Trees (This work)	73	161

Some publications highlight poor application use cases

Task:	Predict hardness
Data:	Seeking mixtures of (Al, Co, Cr, Fe, Ni, and Mn) 632, but only 25 via VAM, so added Cu, Mo to get 91
Models and features:	Neural network: (24,3,1) Features ???
Findings:	$R^2 = 0.94$, MAE = 36
Code:	None and almost no description 😞
Comment:	Element and complexity imbalance Where is data taken from? Bootstrapped 512 NN for uncertainty “mild underfitting” so used 91 datapoints and NN??? No test set Features and ML model never even described

 SpringerLink

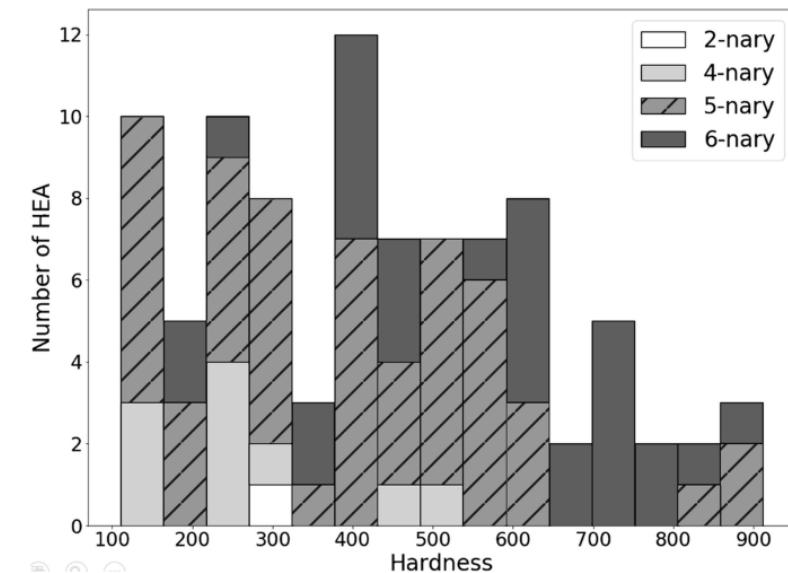
Progress in High-Entropy Alloys | Published: 30 July 2019

Prediction of the Composition and Hardness of High-Entropy Alloys by Machine Learning

[Yao-Jen Chang](#), [Chia-Yung Jui](#), [Wen-Jay Lee](#) & [An-Chou Yeh](#) 

[JOM](#) 71, 3433–3442 (2019) | [Cite this article](#)

3502 Accesses | 50 Citations | [Metrics](#)



Recognizing the need for generality is great, but using NN's to achieve it is not!

atomic mass, electronic negativity, melting points, cohesive energy, etc. For a material design problem with limited data such as that considered herein (with a dataset of 91), the main consideration is to capture the general trend for unexplored alloy compositions, i.e., achieve generality, rather than to minimize the difference between the predicted and experimental values. Pursuing accuracy with a small amount of data often risks reducing the generality, so this work intentionally proceeds with mild underfitting to trade accuracy for generality.

As a result, a small number of features was preferred for our ML model. After the feature engineering process, the combination of hardness, density of the solid, and atomic mass showed the best performance in terms of both R^2 score and Pearson's correlation. As a result, these three physical quantities were introduced for the eight elements. The features were further weighted by the concentration of each element in the compositions. As a result, the eight elements with these three physical quantities could then be transferred into 24 input feature vectors F_{in} . The output layer was defined as the hardness of the HEA. The NN



Rate-dependent constitutive models in CPFEM framework based on ML

Task:	Predict uniaxial twinning, stress-strain, and texture
Data:	819000 inputs (500 grains, 126 σ - ϵ points, 13 $\dot{\epsilon}$) including 6 experimental stress strain datasets
Models and features:	GA-ANN (use GA optimizer to set weights, 5X35X5 nodes) $VEC, T_m, \Delta H_{mix}, \Delta\chi, \delta, \Delta S_{mix}, V_m$
Findings:	Time: <2s for ML vs 15-890s for CPFEM (mesh dependent) stress-strain response ($MSE < 10^{-3}$) overall texture upon large strain ($MSE < 10^{-2}$) twin volume fraction ($MSE < 10^{-4}$)
Code:	No 🤖
Comment:	Expand to dynamic props ($\dot{\epsilon}$) 0.0001/s to 6000/s at RT Dynamic tension → twinning, texturing GA part is slow NN architecture lacking details Very limited niche application (not generalizable) Test dataset is $\dot{\epsilon}=10/s$, which means model interpolating



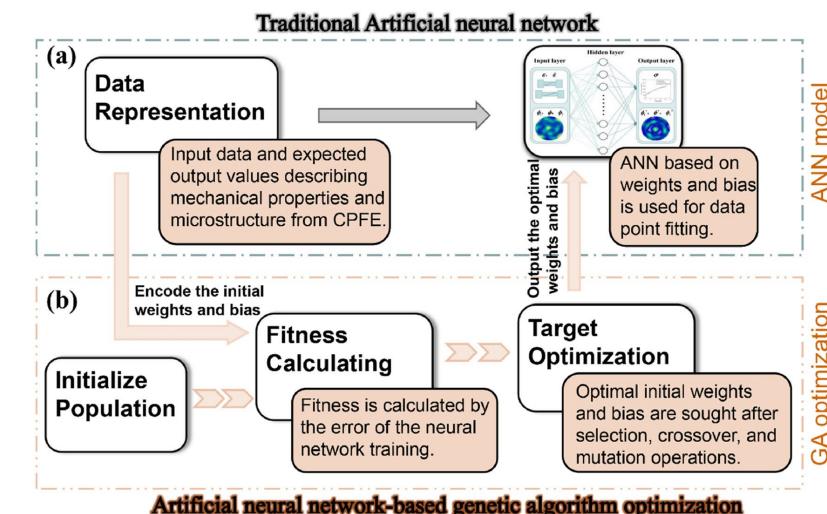
Journal of Alloys and Compounds

Volume 845, 10 December 2020, 155911

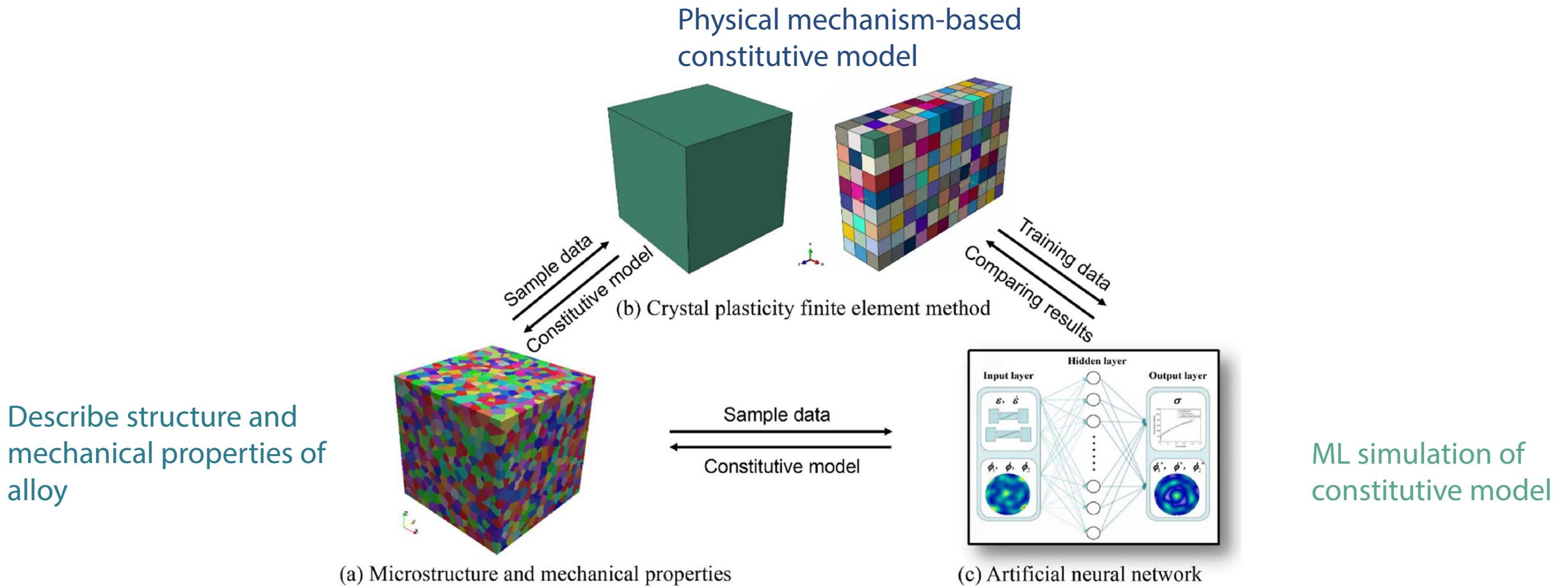


Strain-rate-sensitive mechanical response, twinning, and texture features of NiCoCrFe high-entropy alloy: Experiments, multi-level crystal plasticity and artificial neural networks modeling

T.J. Gao ^{a, b}, D. Zhao ^{a, b} T.W. Zhang ^{a, b}, T. Jin ^{a, b}, S.G. Ma ^{a, b}, Z.H. Wang ^{a, b}



ML outperforms physical mechanism-based CPFEM in highly niche task



Task: predict the quasi-static and dynamic uniaxial tensile mechanical response, twinning and texture characteristic in NiCoCrFe high-entropy alloy

ML used to predict stacking fault energies for HEAs

Task:	Predict stacking fault energies
Data:	Varying data 20-200 structures of each supercell type atom supercells range (48, 480, 6000, 108,000)
Models and features:	Linear models (ridge & lasso) Features: type and number of local bonds (1-7 Angstroms up to 4 th nearest neighbor) binned by distance (0.1 Angstrom increments)
Findings:	480 atoms → 4320 atoms, $R^2=0.99$ ➔
Code:	None 😞
Comment:	Specifically use ML to generalize (train binary, predict ternary) and train small, predict large Simulations fail, so skip them directly with ML 20% test dataset Ambiguous data generation/splitting

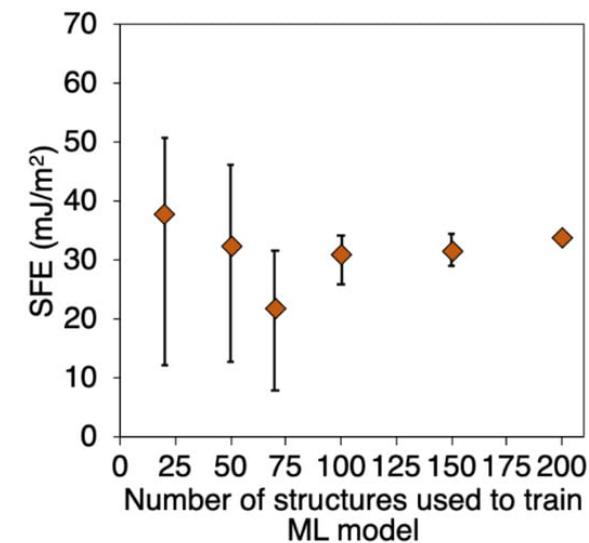
Machine Learning Enabled Prediction of Stacking Fault Energies in Concentrated Alloys

by  Gaurav Arora  and  Dipuneet S. Ailidhy * 

Department of Mechanical Engineering, University of Wyoming, Laramie, WY 82071, USA

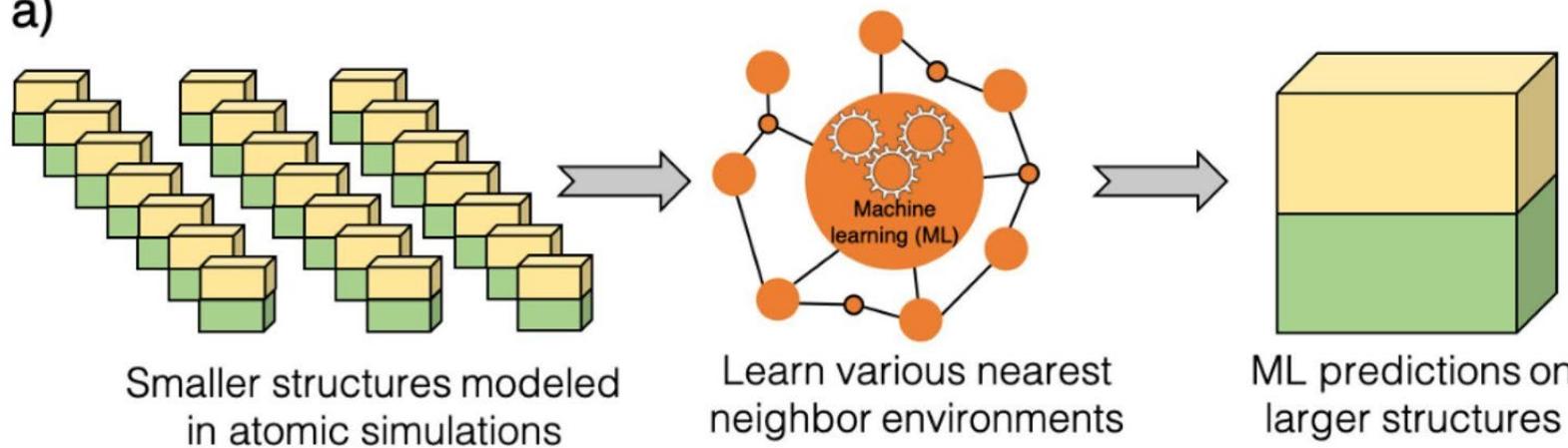
* Author to whom correspondence should be addressed.

Metals 2020, 10(8), 1072; <https://doi.org/10.3390/met10081072>



ML was specifically used to generalize to larger and more compositionally complex alloys

a)

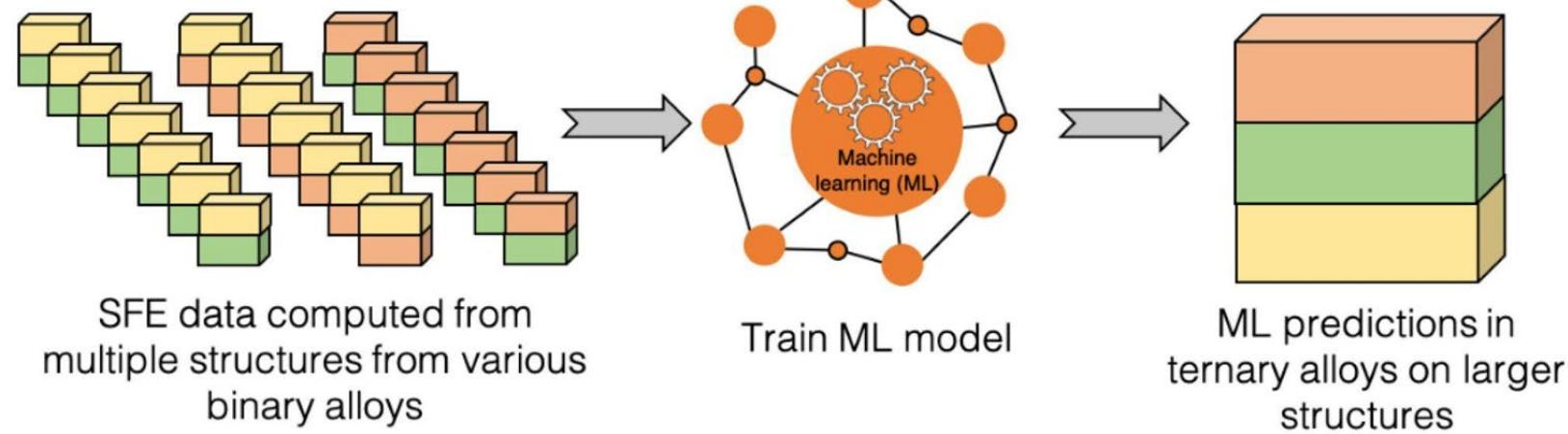


Smaller structures modeled
in atomic simulations

Learn various nearest
neighbor environments

ML predictions on
larger structures

b)



SFE data computed from
multiple structures from various
binary alloys

Train ML model

ML predictions in
ternary alloys on larger
structures

Prediction of MD simulated UTS

Task:	Predict ultimate tensile strength using MD simulation data
Data:	186 MD simulated in $\text{Cr}_x\text{Co}_y\text{Ni}_{100-x-y}$ system
Models and features:	NN: (3,10,10,1) Features: none (one hot encode elemental amount)
Findings:	$R^2 = 0.9875$
Code:	No code. No data 😬
Comment:	$\text{Cr}_x\text{Co}_y\text{Ni}_{100-x-y}$ MEA ($20 \leq x \leq 60$ at.%, and $20 \leq y \leq 60$ at.%) Test data 45 compositions in same family (not generalizable) Likely highly overfit or interpolated information



Journal of Materials Science & Technology

Volume 68, 30 March 2021, Pages 70-75

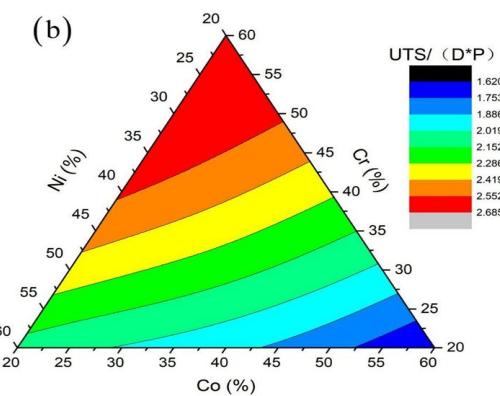
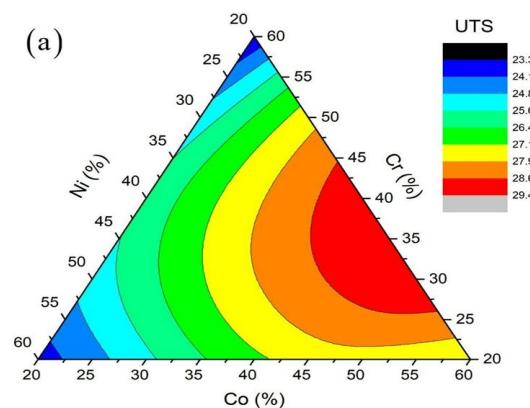
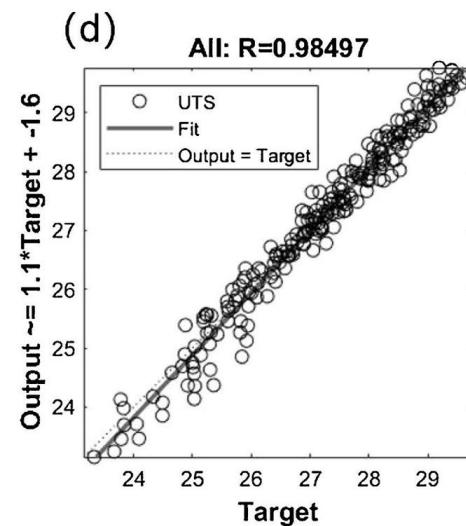
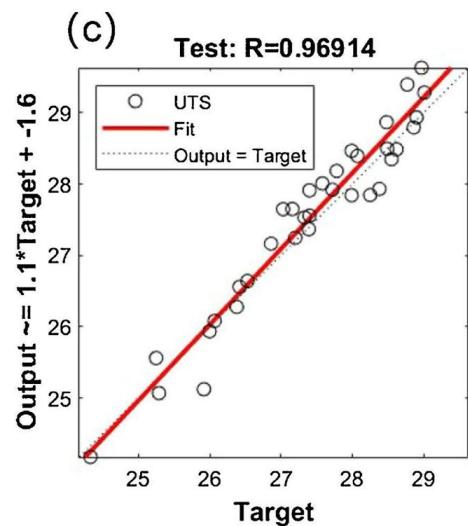
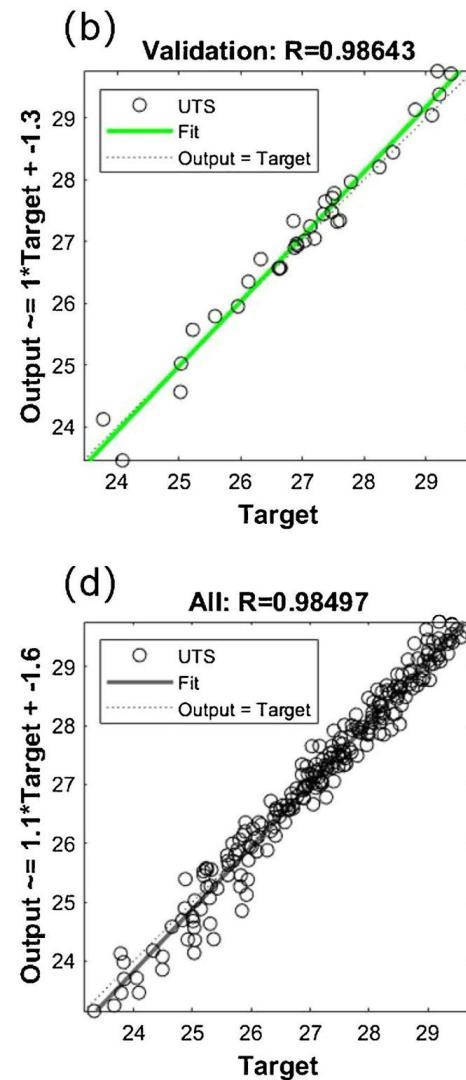
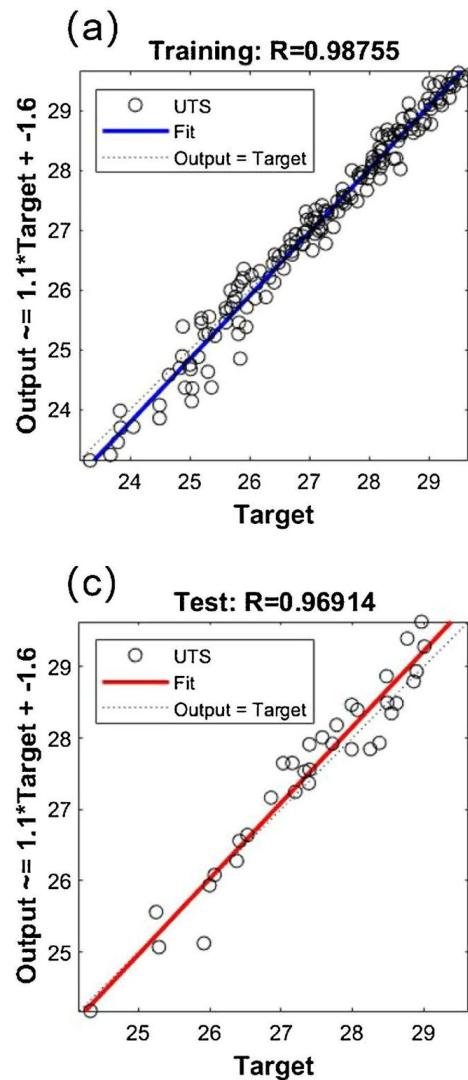


Research Article

High-throughput simulation combined machine learning search for optimum elemental composition in medium entropy alloy

Jia Li ^a, Baobin Xie ^a, Qihong Fang ^a, Bin Liu ^b, Yong Liu ^b, Peter K. Liaw ^c

Properties are mapped to ternary space and can be normalized by density, cost etc



The strength and error in different compositional regions.

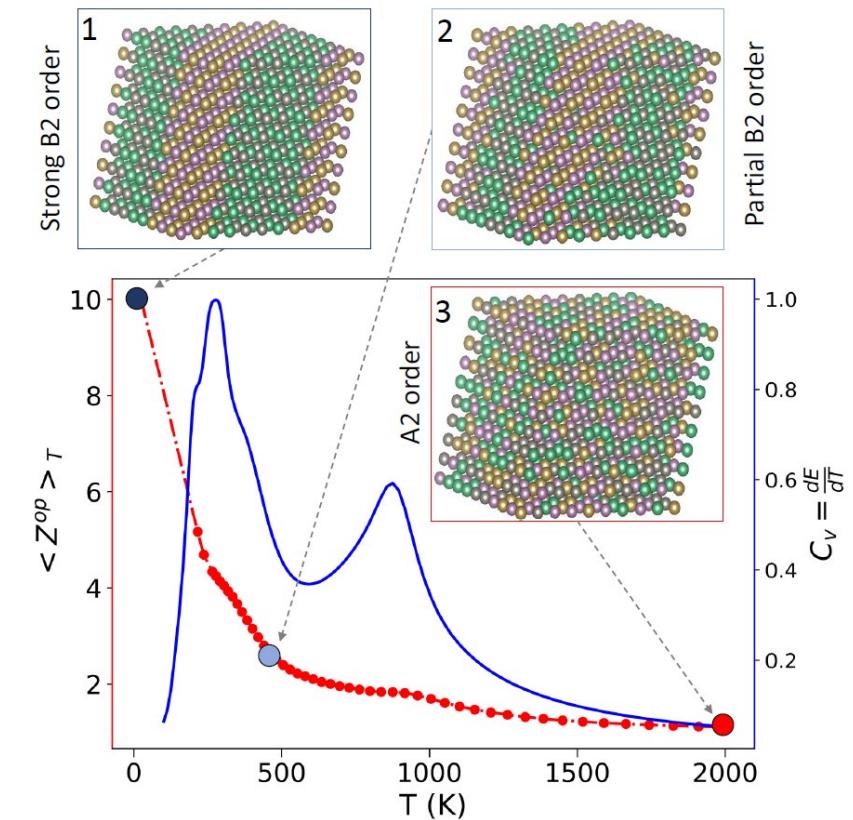
System	MD result (GPa)	Machine learning result (GPa)	Error (%)
Co ₂₁ Cr ₂₀ Ni ₅₉	23.5	23.9	1.7
Co ₂₉ Cr ₃₀ Ni ₄₁	26.7	26.9	0.7
Co ₄₉ Cr ₃₀ Ni ₂₁	29.4	29.2	0.7

ML could be used as a fast, accurate proxy for predicting order parameters in HEAs

Current state of the art proxy for order parameter is Warren-Cowley short range order parameter

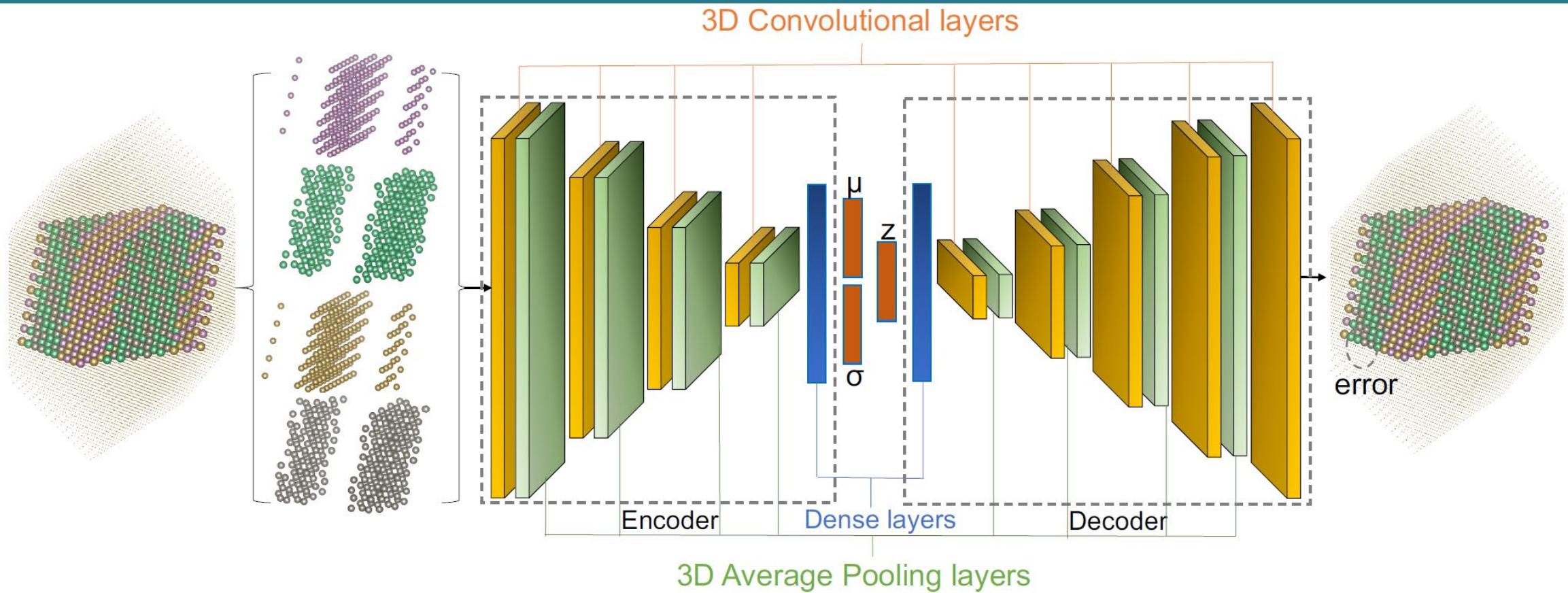
$$a_l^{ij} = 1 - \frac{P_l^{i|j}}{c_i}$$

where $P_l^{i|j}$ is probability of finding atom i at l -th neighbor for a given atom j and c_i is the concentration of atom i



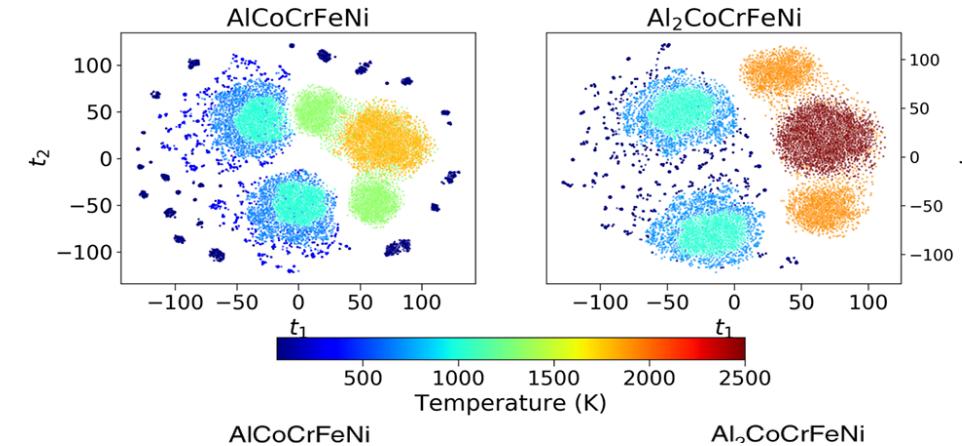
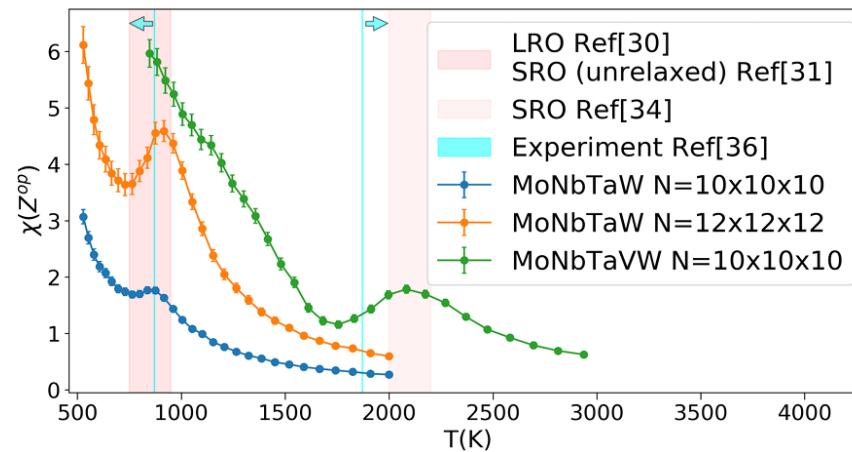
Problem: yields matrix, not scalar, unclear how long range calculation should be

ML has been used to predict order parameters in HEAs



"if we hypothesize the symmetry of input X is preserved by the latent variable z, the DKL loss is minimized when the more symmetric phases are encoded into data points near the origin of the latent space"

ML has been used to predict order parameters in HEAs



"Although defined through neural networks, its physical meaning is transparent and simple, where the crystallographic symmetry during phase transitions is quantitatively preserved. It is surprising to see the physical information can be so precisely preserved after going through the highly nonlinear neural-network operations. This is an excellent example to demonstrate the power of machine learning to understand the physics in materials science. Its power is far beyond its common applications as a mathematical black box. More future studies are merited to fully explore its power."

Data availability



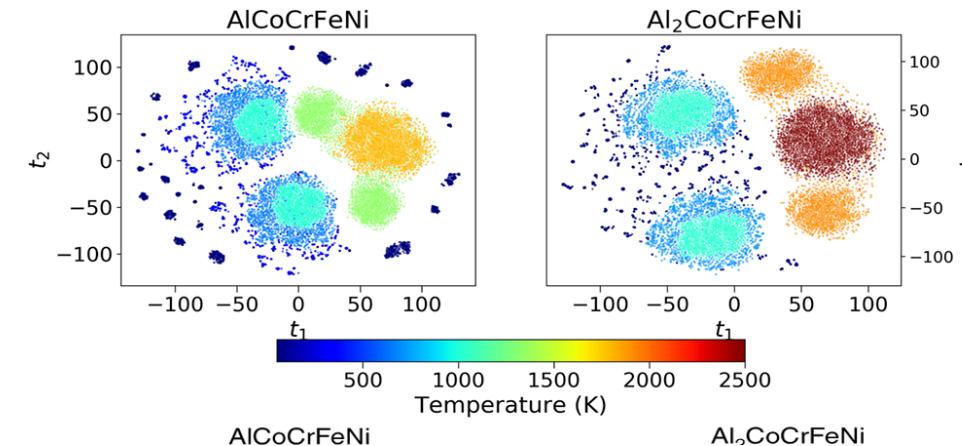
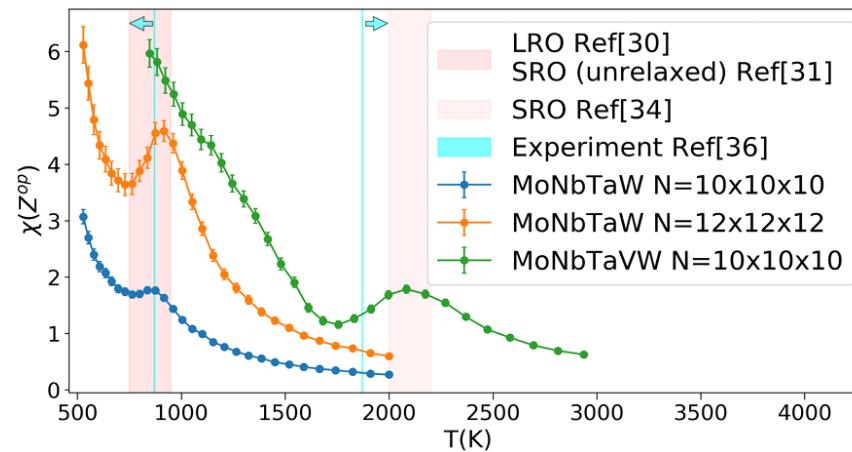
The sample training and validation data, along with a pre-trained model for Ti₃₈V₁₆Nb₂₃Hf₂₄ are available at <https://doi.org/10.6084/m9.figshare.14417225.v3>. The data for all figures are available at <https://code.ornl.gov/jqyin/deepthermo/-/tree/master/data>.

Code availability



The VAE model training and order parameter inferencing codes are available at <https://code.ornl.gov/jqyin/deepthermo>. The figures are plotted with the notebook at https://code.ornl.gov/jqyin/deepthermo/-/blob/master/utils/heavae_analysis.ipynb.

They also include code and data!



Data availability



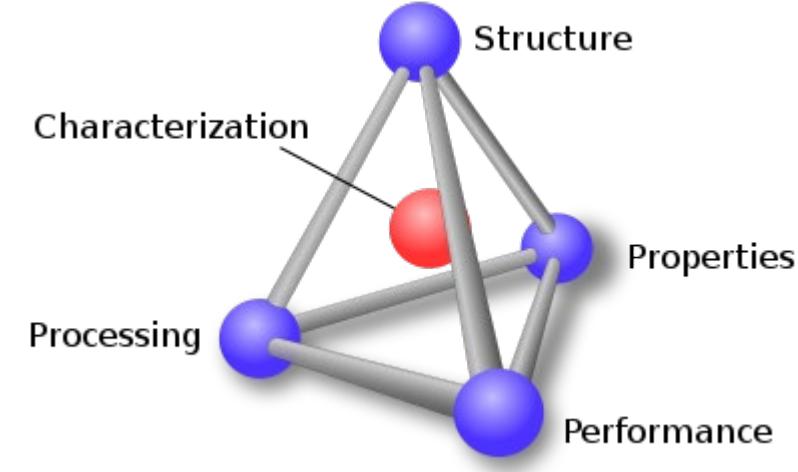
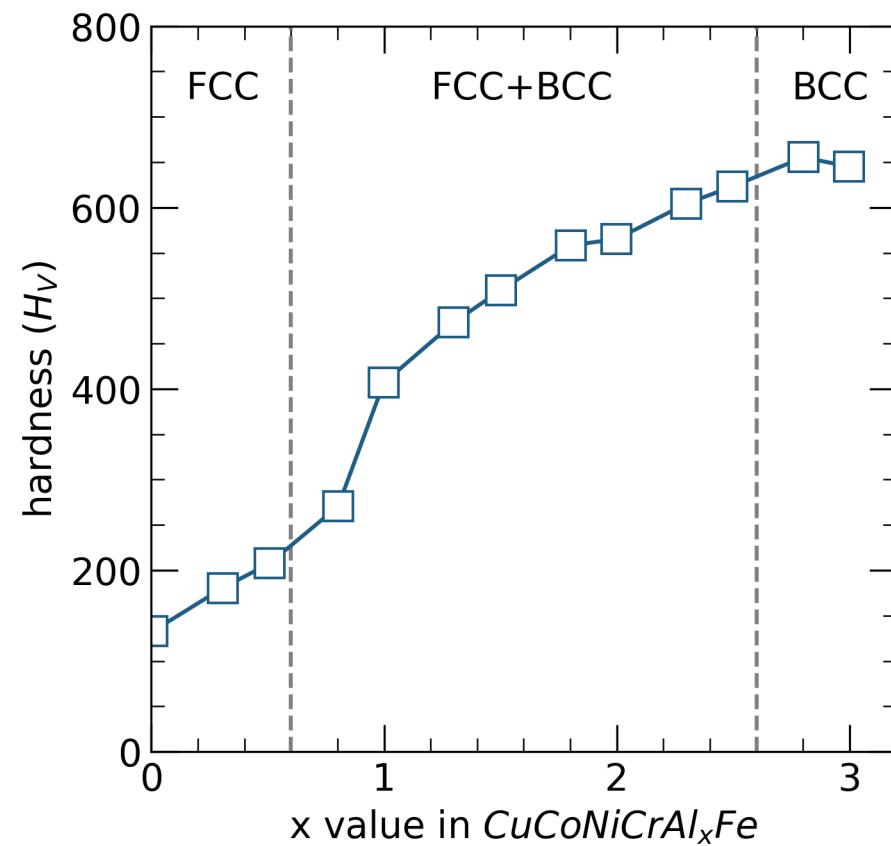
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Code availability



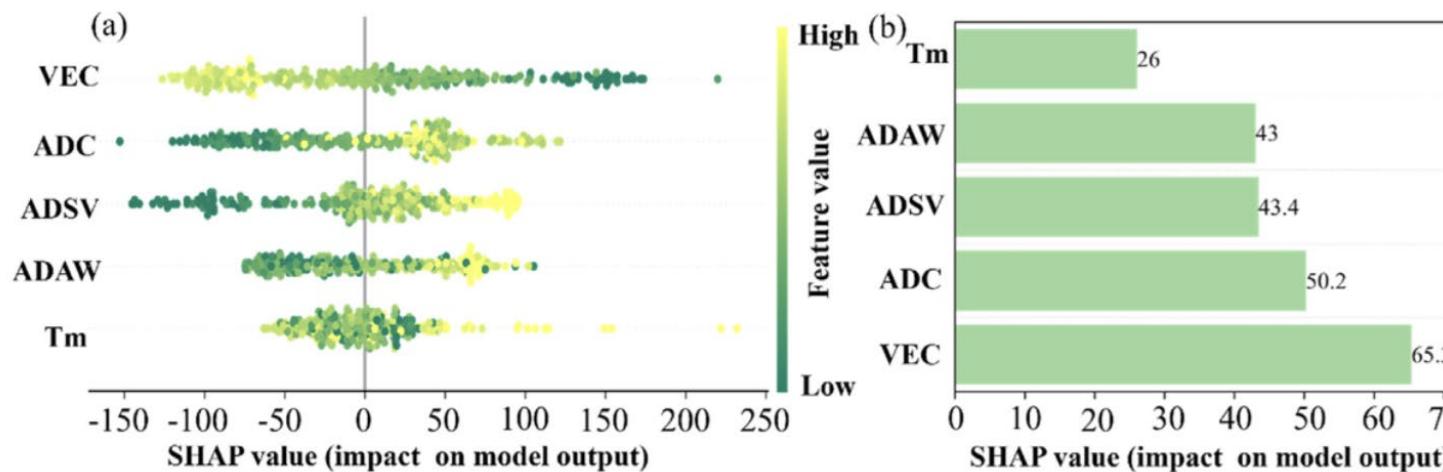
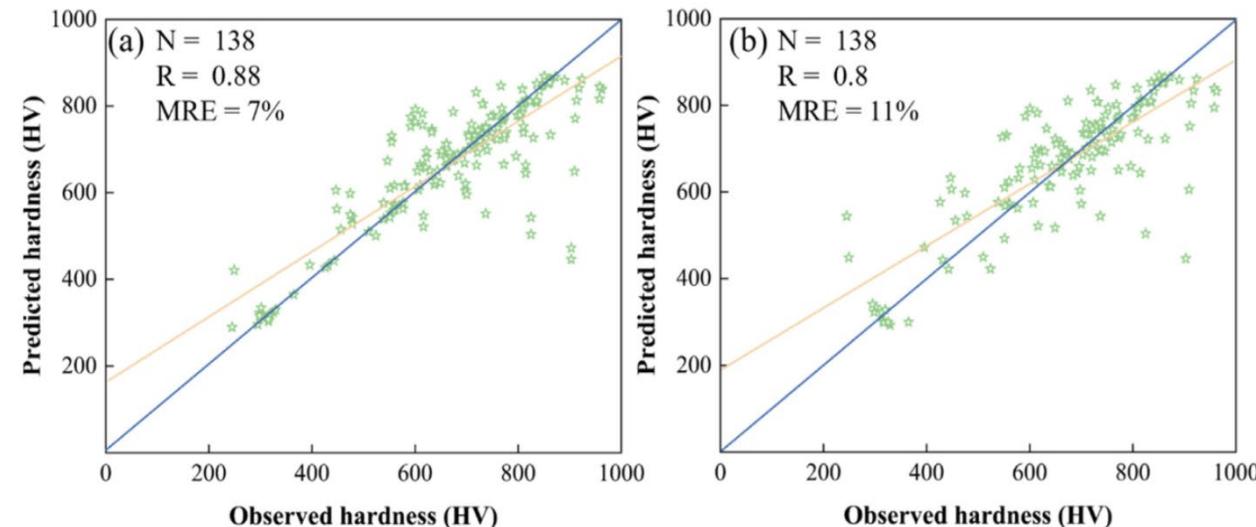
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Model interpretability gives us real insight into structure-property-processing links



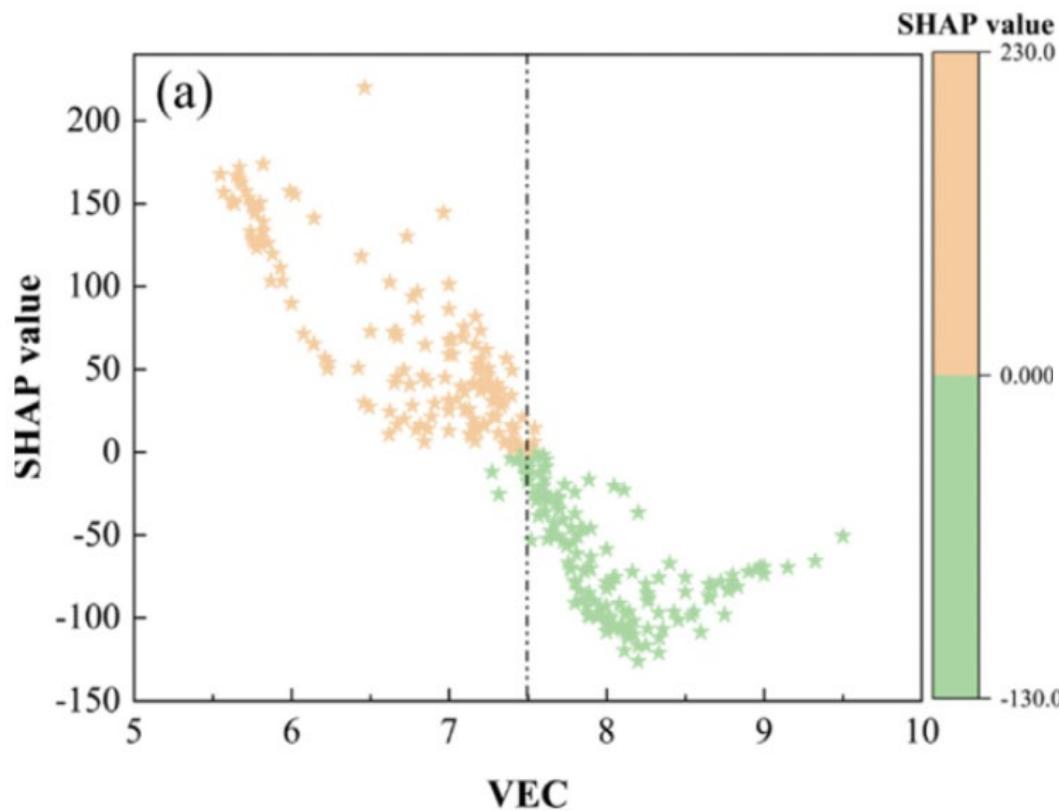
Yeh et al Nanostructured high-entropy alloys with multiple principal elements: novel alloy design concepts and outcomes, Adv Eng Mater, 6 (5) (2004), pp. 299-303

Most models rely on feature importance scores to provide interpretability



Yang et al, A machine learning-based alloy design system to facilitate the rational design of high entropy alloys with enhanced hardness, Acta Materialia (2022)

SHAP analysis for valence electron concentration & melting point



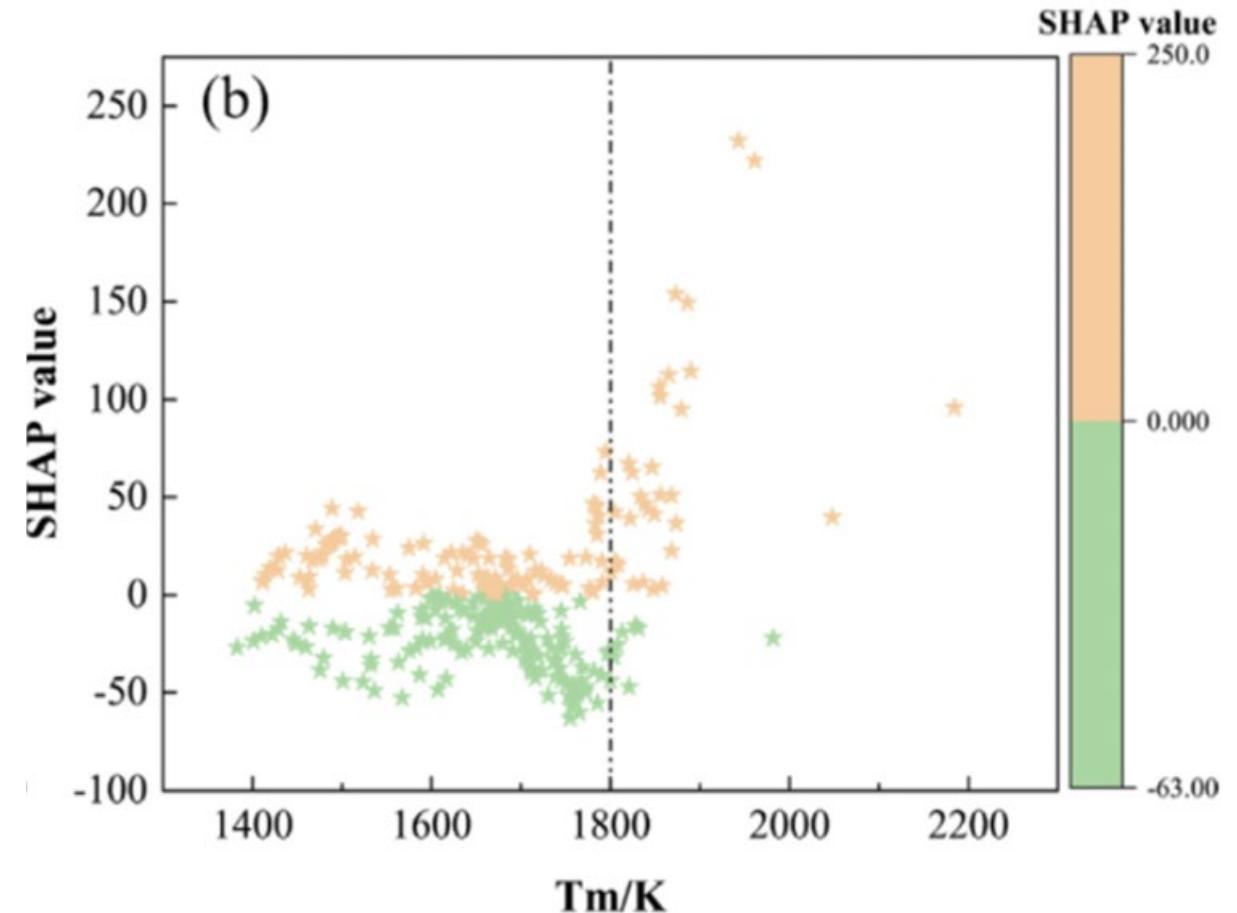
VEC exerts positive influence on hardness when VEC < 7.5, negative otherwise because BCC solid solution phase is more stable when VEC < 6.87!

BCC structure possesses more severe lattice distortion compared to FCC leading to more solid solution strengthening. Elements such as Al, Cr, Ti, Mo, and V, due to their low VEC, an increase in their content would decrease the VEC of the alloys, which might give rise to an increase in hardness.

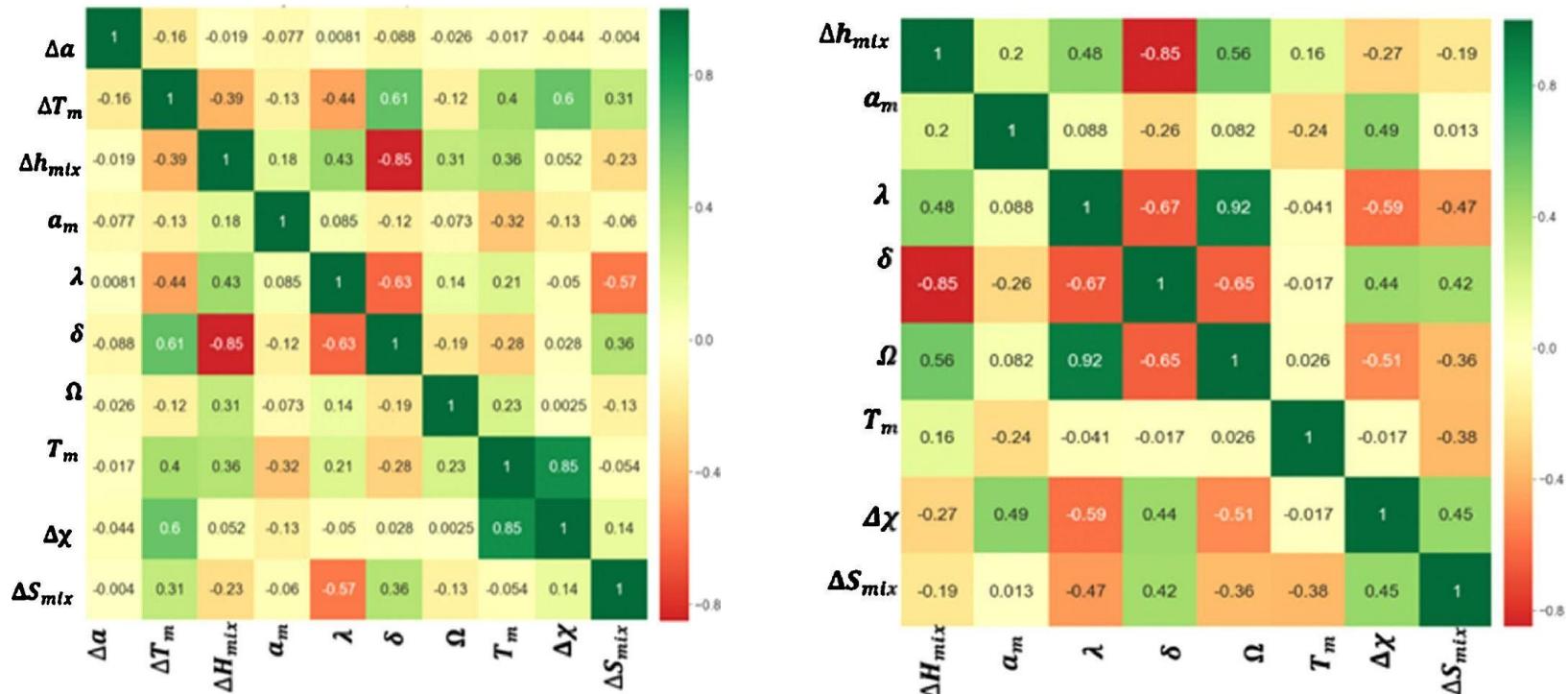
SHAP analysis for valence electron concentration & melting point

SHAP shows melting temperature benefits hardness when above 1800K, but is indifferent below this temperature

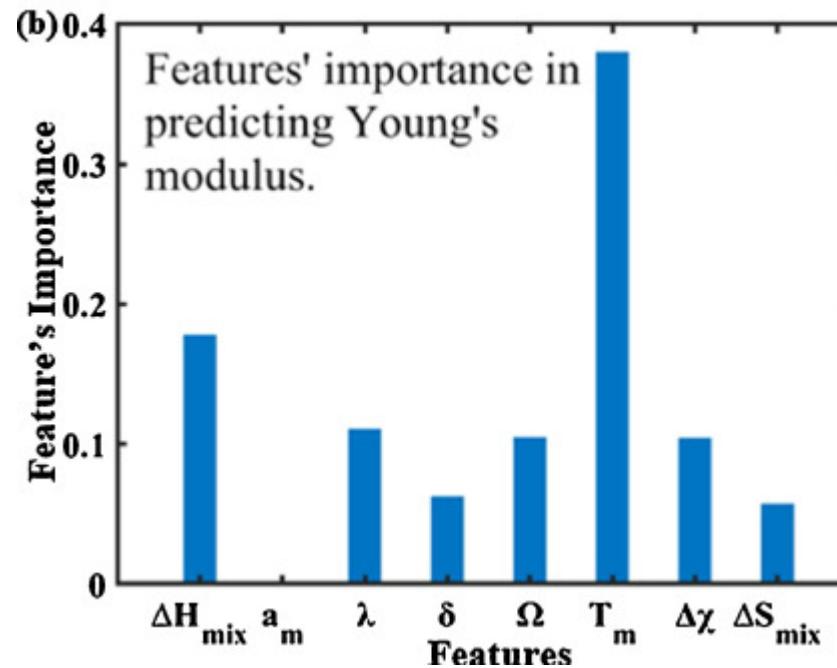
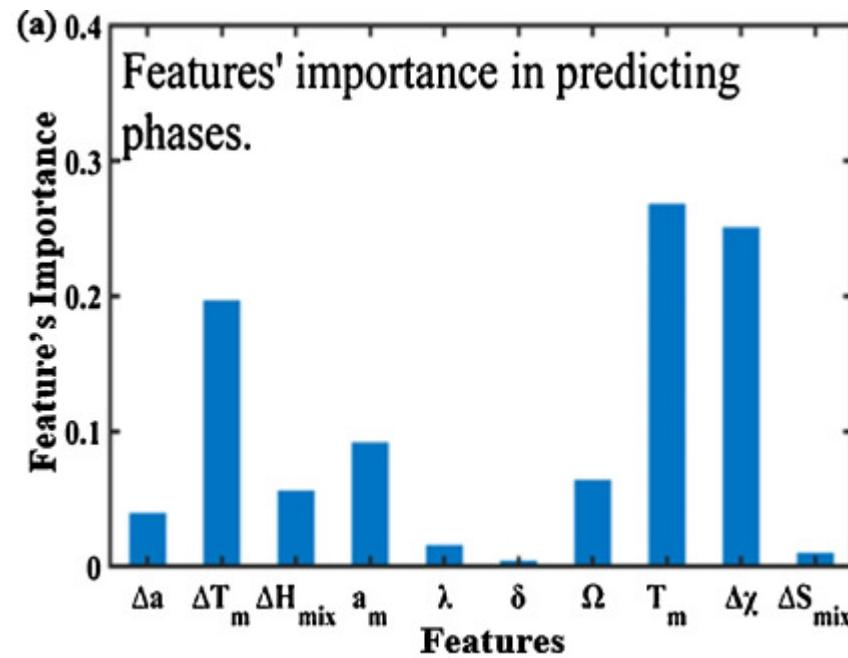
The composition of these alloys contained Ti, Mo, V, Cr which have high melting points leading to higher strength and hardness



Feature importance can provide counterintuitive findings – for example MoTaTiZrW system



Feature importance can provide counterintuitive findings – for example MoTaTiZrW system



Mean melting point and electronegativity difference exert the strongest contributions to the phase formation, and melting temperature and enthalpy of mixing are the key features impacting Young's modulus of these materials. Entropy of mixing only negligibly influences the phase or the Young's modulus, reigniting the issue of its actual impact on the material phase and properties of HEAs.

There are a growing number of excellent HEA data sets!

Borg et al. "Expanded dataset of mechanical properties and observed phases of multi-principal element alloys." *Scientific Data* 7.1 (2020): 1-6. <https://doi.org/10.1038/s41597-020-00768-9>

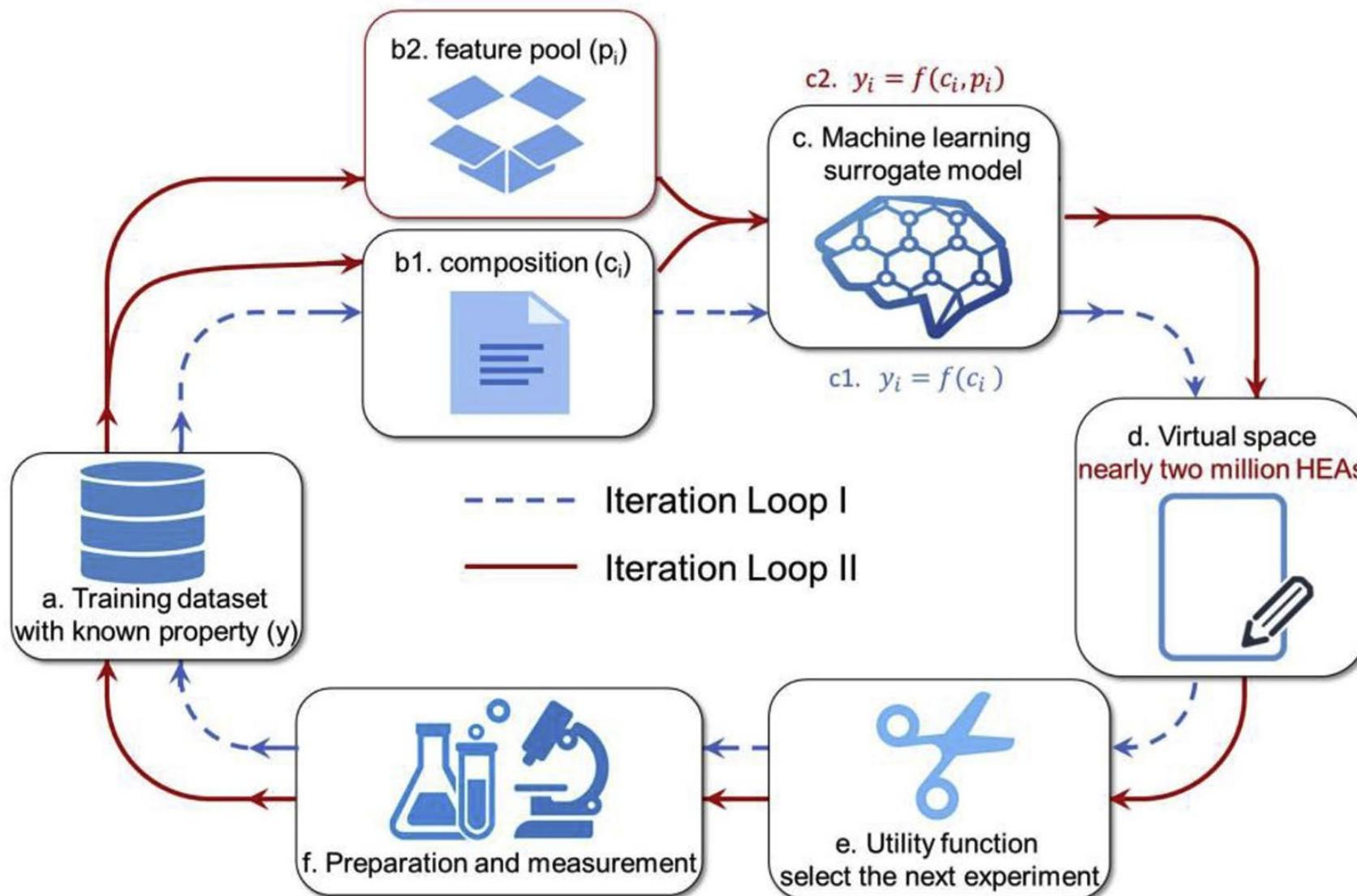
Han et al. "Data-driven based phase constitution prediction in high entropy alloys." *Computational Materials Science* 215 (2022): 111774.
<https://doi.org/10.1016/j.commatsci.2022.111774>

Machaka et al. "Machine learning-based prediction of phases in high-entropy alloys: A data article." *Data in Brief* 38 (2021). <https://doi.org/10.1016/j.dib.2021.107346>

Wen et al. "Machine learning assisted design of high entropy alloys with desired property." *Acta Materialia* 170 (2019): 109-117. <https://doi.org/10.1016/j.actamat.2019.03.010>

Detor et al. "Refractory high entropy alloy dataset with room temperature ductility screening." *Data in Brief* 45 (2022): 108582. <https://doi.org/10.1016/j.dib.2022.108582>

Systematic search for new HEAs via active learning

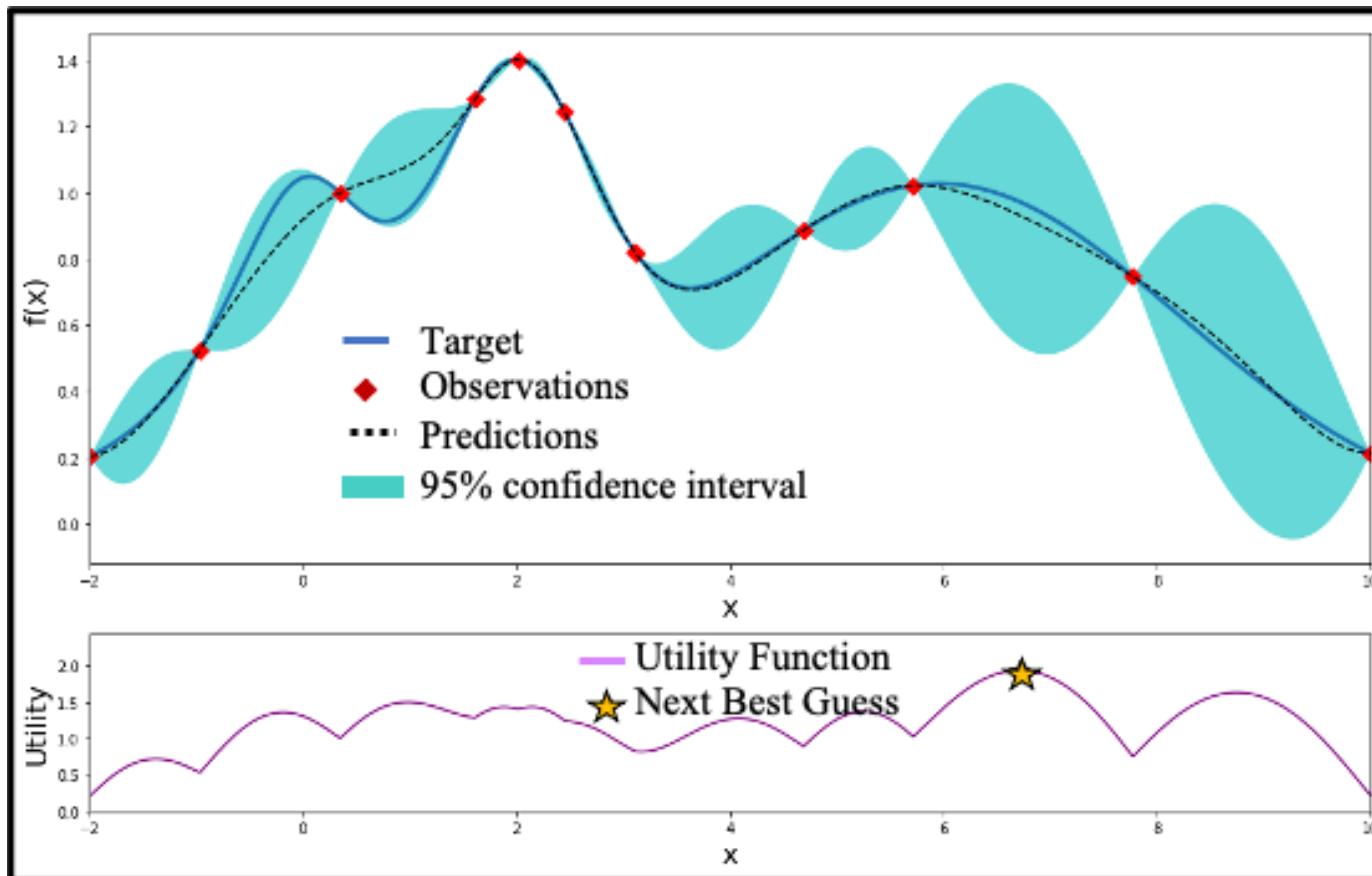


AlCoCrCuFeNi

Iteration Loop 1 :
Chemical Composition as
features

Iteration Loop 2:
Chemistry of elements as
features

Active learning often leverages a utility function to balance exploitation vs exploration



Expected Improvement as utility function

$$EI(x) = \underbrace{\sigma(x)z\Phi(z)}_{\text{Exploitation}} + \underbrace{\sigma(x)\phi(z)}_{\text{Exploration}}$$
$$z = [\mu(x) - f(x^+) - \varepsilon]/\sigma(x)$$

μ predicted mean

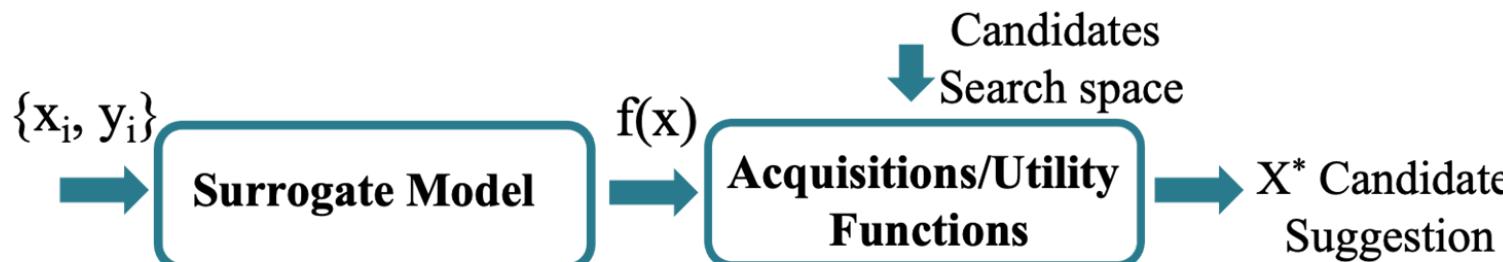
σ standard deviation

$f(x^+)$ maximum value of the target observed in training dataset

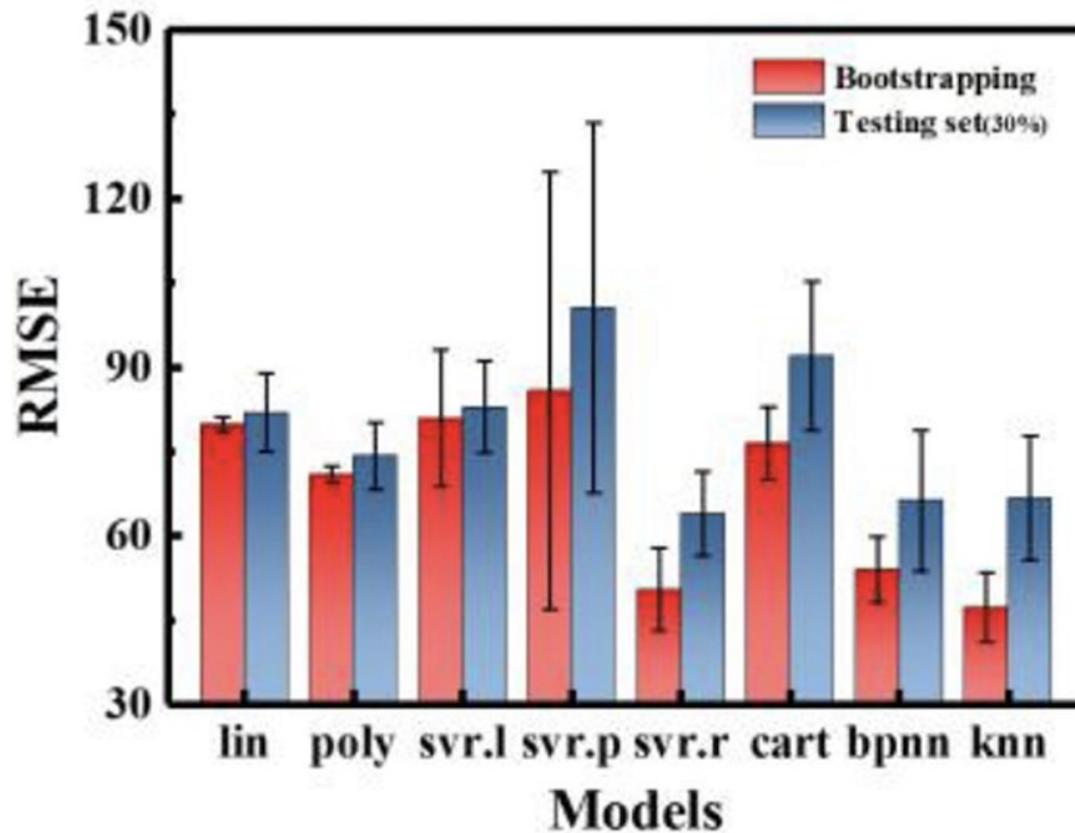
Φ cumulative distribution function

ϕ probability distribution function

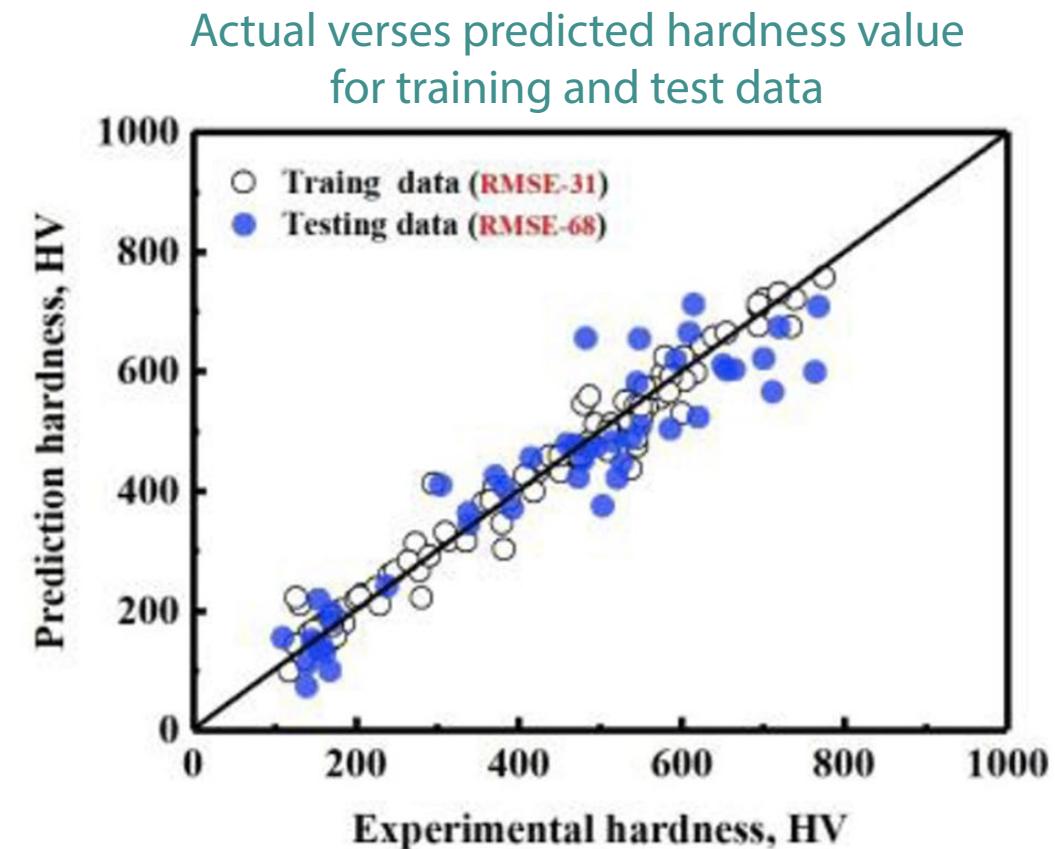
ε regulates the exploration, higher ε more exploration



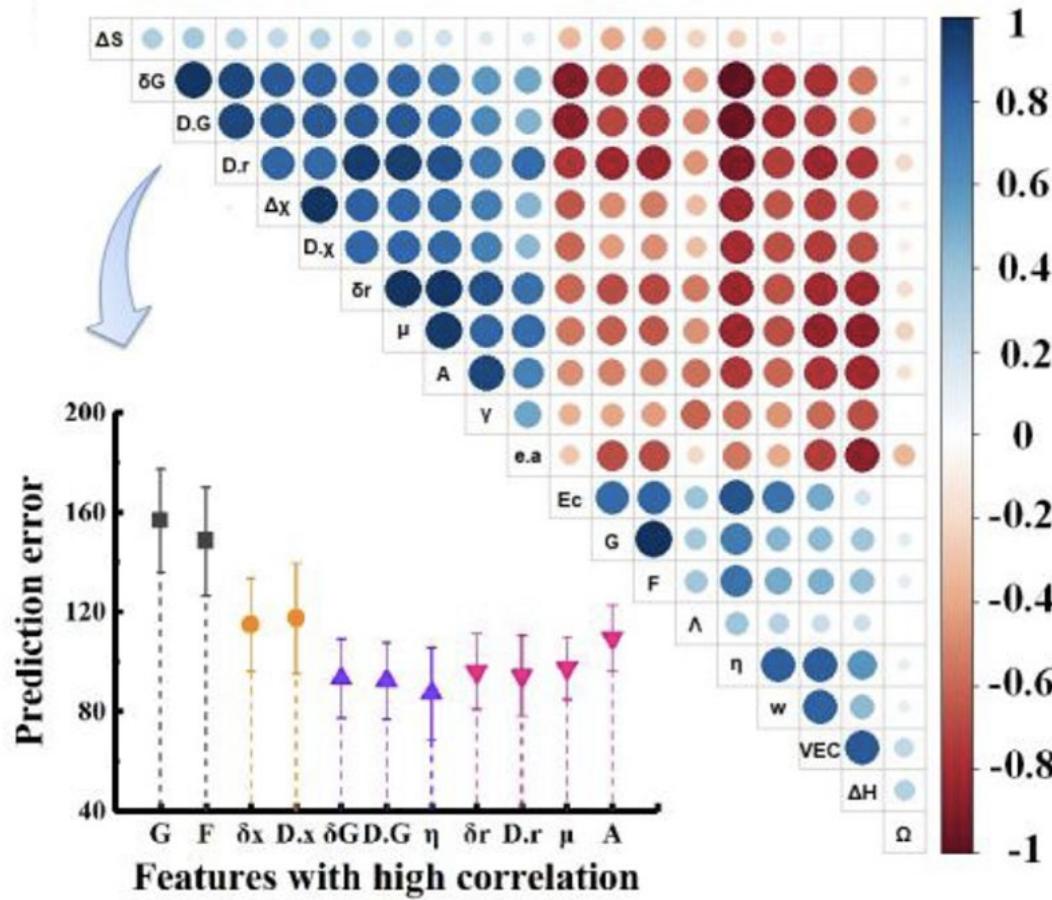
Surrogate model selection (iterative loop 1, composition only)



RMSE Score for different models for training (70%) and test data (30%). The training data are sampled by using Bootstrapping and 100 models are trained. The figure shows the mean RMSE value.

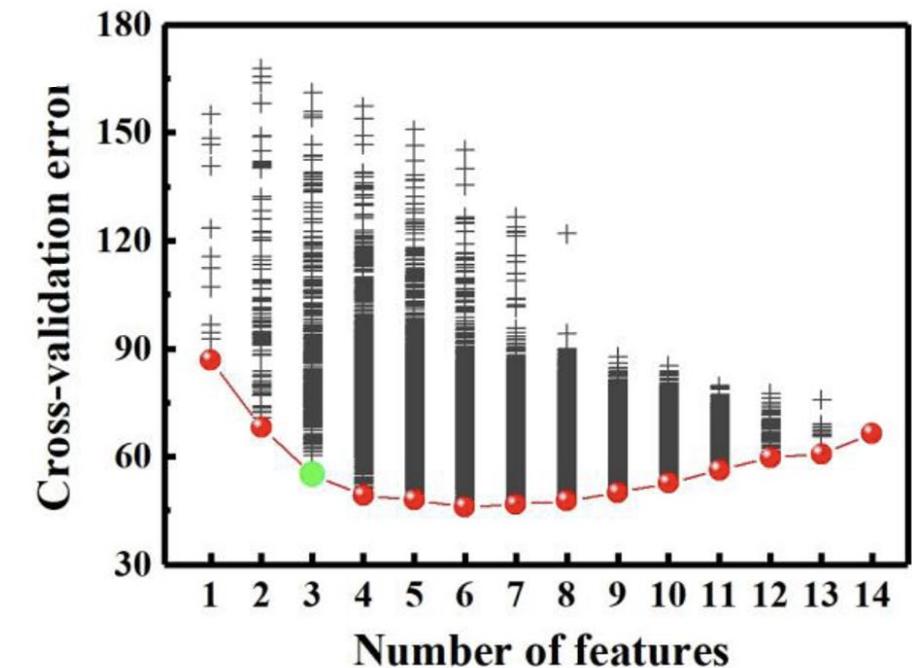


Feature selection (iterative loop 2)



Pearson Correlation map of the initial
20 features

The cross-validation error of each possible svr.r model containing a subset of preselected features



We then define a candidate search space

- ❑ Virtual Alloy compositions in the form : $\text{Al}_x\text{Co}_y\text{Cr}_z\text{Cu}_u\text{Fe}_v\text{Ni}_w$
- ❑ Mole Fraction : $x + y + z + u + v + w = 100\%$

With varying concentrations of the elements in atom percent with a step size of 1 atom %.

Training Dataset hardness value varies from 110 - 770 HV

Based on the composition, Upper and lower compositional limits for alloys with hardness exceeding 500 HV in the training dataset

$$15 < x < 47, 5 < y < 22$$

$$6 < z < 34, 5 < u < 16$$

$$5 < v < 31, 5 < w < 22$$

Total 1.8 Millions Candidates, (1331 quaternary,
218324 quinary, 1.6 Million Six component alloys)

Finding the optimal candidate

