Phase Prediction of High-Entropy Alloys

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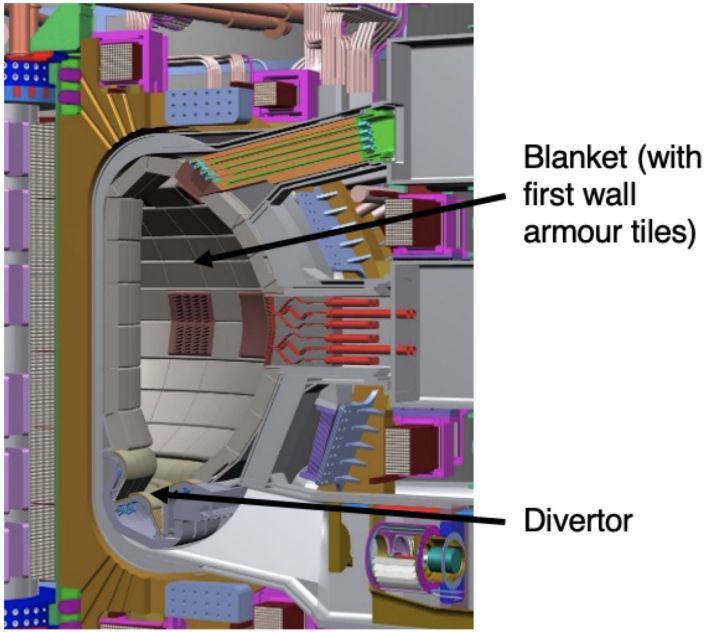
HEA Alloys



Unique metal alloys ideal for utilization in extreme environments due to their strength, structural stability, and resistance to corrosion and radiation

- Nuclear applications: radiation shielding

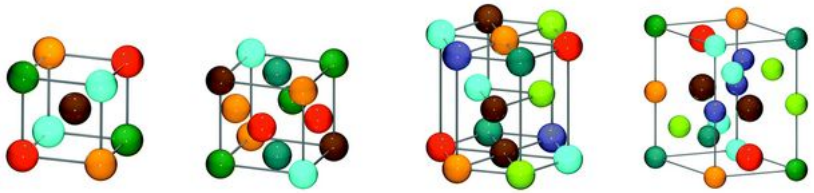
These alloys are composed of at least 5 elements with concentrations between 5 and 35 percent the atomic percentage, forming solid state solutions

[doi.org/10.3390/e23010098]

Motivation

Altering the elemental concentrations distorts the lattice structure, changing alloy resistance properties

- Can be used to an advantage
  - Achieving resistance to high temps



$\sim 10^8$ combinations of HEAs can be developed from only 64 elements in the periodic table using different weight percentages, with varying synthesis and processing routes

- Experimental process for testing characteristics is extremely time consuming
- Machine Learning models can be used to hasten the process and discover a general trend to identify alloy properties (e.g. phase prediction)

[doi.org/10.1039/D1EE00505G]

Goals

- To be able to determine crystal structures and the properties that HEAs typically have
- Analyze and figure out techniques for better data cleaning and learning
- Optimize each model with their hyperparameters for better performance
- Benchmark against the untuned versions of each model

Physical Parameters

Each HEA is described by a chemical composition tied to the following features:

ΔH_{mix}: Thermal Energy of the state

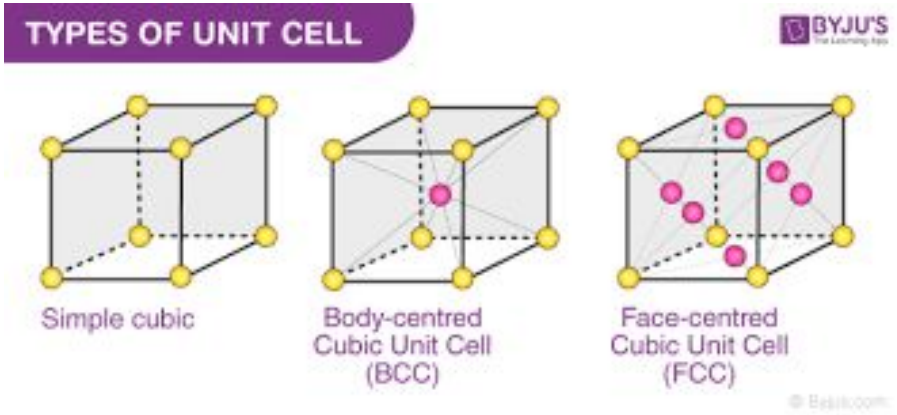
ΔS_{mix} : Deviation from perfect order of the state

δ: Atomic Size Difference

Δχ: General form of electron cloud

VEC: % e^- in Valence Band

Phase: FCC, BCC, IM, FCC+BCC



https://byjus.com/chemistry/bcc-fcc-primitive-cubic-unit-cell/

Phase Determination

For a given composition, the phase is determined by the VEC value.

- BCC when VEC < 6.87
- FCC when VEC > 8.0
- BCC + FCC when VEC between 6.87 and 8.0

Composition	ΔH_{mix}	ΔS_{mix}	δ	$\Delta\chi$	VEC	Phase
$Al_{0.5}NbTaTiV$	-8.395062	13.145944	3.7375048	0.0496158	4.55556	BCC
$Hf_{1.375}Nb_{0.25}Ta_{0.25}Ti_{1.75}Zr_{1.375}$	0.98	11.449449	4.4954539	0.1147168	4.1	BCC
$HfMo_{0.5}NbTiV_{0.5}Si_{0.5}$	-31.11815	14.53503	9.4420482	0.2565635	4.53191	IM

Data Set

- 1200 alloys in total
- 30 metallic elements
  - Lots of NaN values

Out[8]:

	Al	Co	Cr	Fe	Ni	Cu	Mn	Ti	V	Nb	...	Pt	Y	Pd	Au	dHmix	dSmix	δ	Δχ	VEC	Phases
0	0.111	NaN	NaN	NaN	NaN	NaN	NaN	0.222	0.222	0.222	...	NaN	NaN	NaN	NaN	-8.395	13.146	3.738	0.050	4.556	1
1	0.158	NaN	NaN	NaN	NaN	NaN	NaN	0.215	0.215	0.215	...	NaN	NaN	NaN	NaN	-9.352	13.333	3.863	0.233	4.684	1
2	0.588	NaN	NaN	NaN	NaN	NaN	NaN	0.235	0.235	0.235	...	NaN	NaN	NaN	NaN	-4.042	12.708	4.003	0.243	4.882	1
3	0.588	NaN	NaN	NaN	NaN	NaN	NaN	0.235	0.235	0.235	...	NaN	NaN	NaN	NaN	-4.817	12.708	3.832	0.050	4.647	1
4	0.476	NaN	NaN	NaN	NaN	NaN	NaN	0.239	0.239	NaN	...	NaN	NaN	NaN	NaN	-3.356	12.569	4.018	0.244	4.905	1

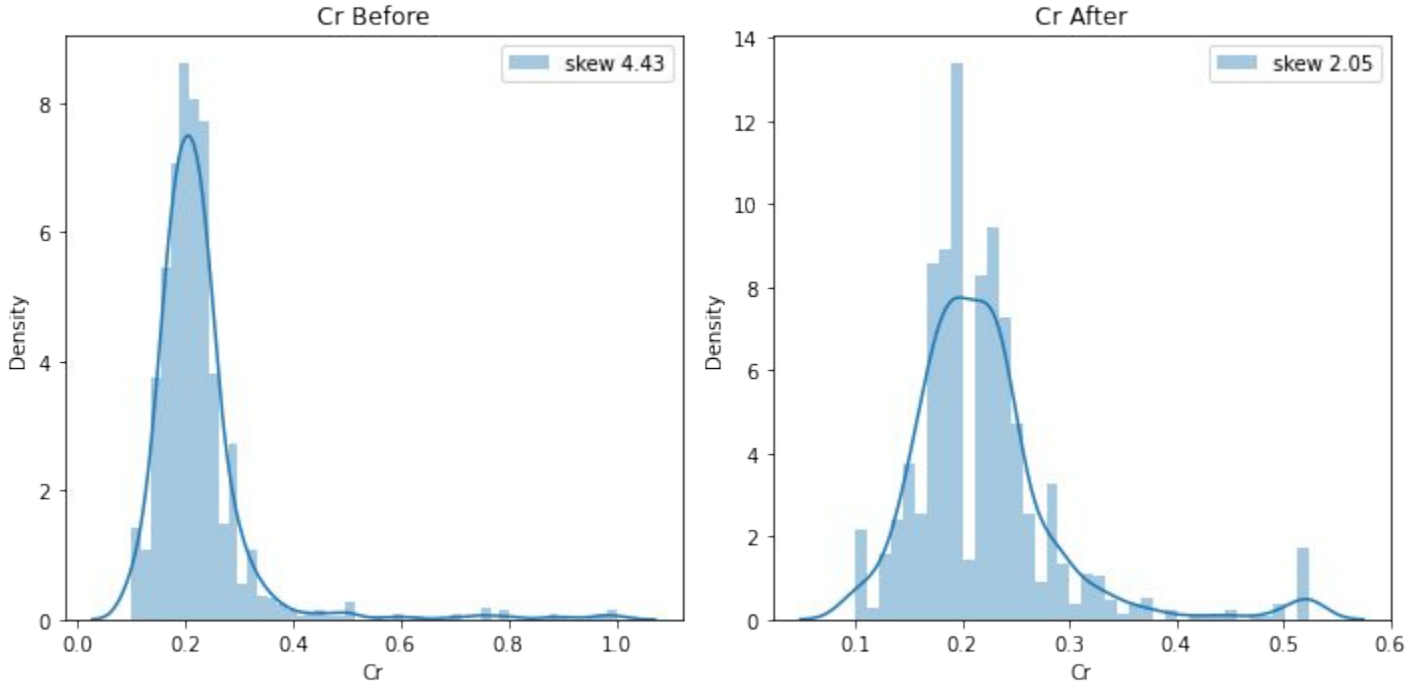
5 rows × 36 columns

- Compiled from: Materials for Design Open Repository. High Entropy Alloys
- IM: 0, BCC: 1, FCC: 2, BCC+FCC: 3

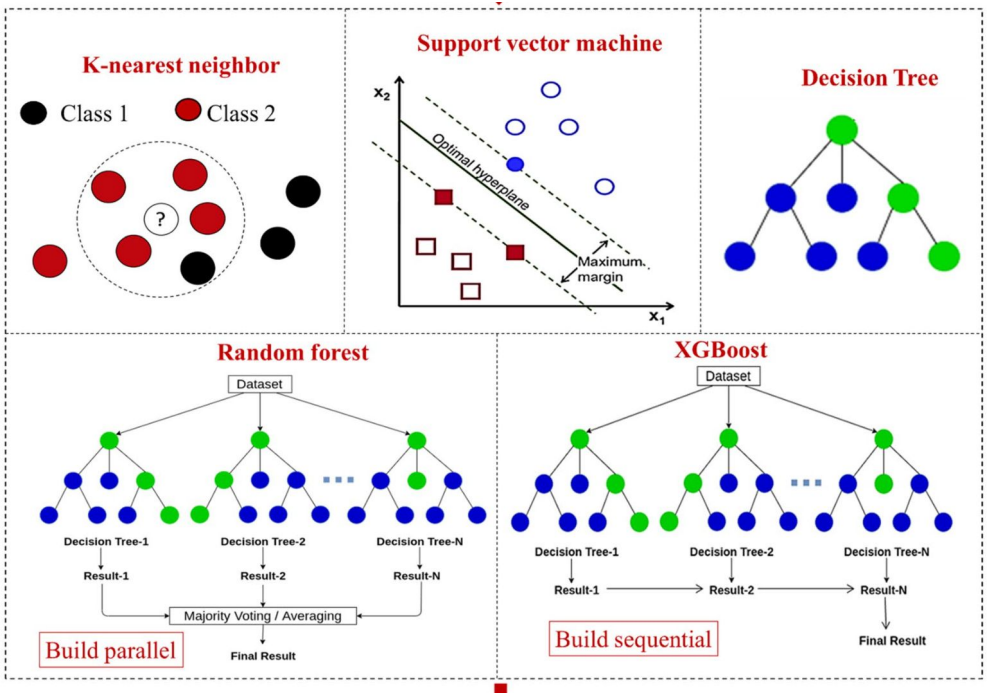
Data Pre-processing

- The data is compiled from multiple sources so some values are NaN
- Data needs to be imputed
    - One technique we used is getting the closest neighbor’s values and using the mean
- Feature scaling:
    - We need to minimize the skewing by introducing a cap on data points
    - One solution is to use Z-Score capping, we essentially set a max of 3 standard deviations that a data value can go up to

Example of De-skewing



The 5 Models:



https://doi.org/10.1038/s41598-023-31461-7

Methodology - K Nearest Neighbors

Hyperparameters:

- K Neighbors: between 1 and 25
- Weight: uniform vs distance
- Metric:
  - Euclidean: straight line distance
  - Manhattan: sum of total difference of each component
  - Chebyshev: max of each components difference
  - Cosine: dot product definition

Methodology - Support Vector Machine

Hyperparameters:

- Regularization parameter: between 1e-3 and 1e3
- Kernel: linear, poly, rbf, sigmoid
- Degree (for poly): between 2 and 5
- Gamma (for poly, rbf, sigmoid): scale, auto

Methodology - Decision Tree

Hyperparameters:

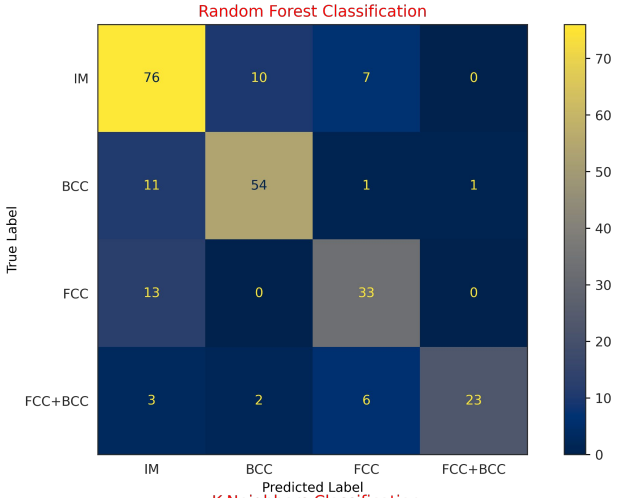
- Max depth: how many levels (“big”) the tree is. Need to optimize for over/underfitting
- Minimum Samples Split: Threshold of samples a node has before splitting.
- Minimum Samples Leaf: Counterpart of minimum samples. When splitting stops which is important for dictating size of the tree.

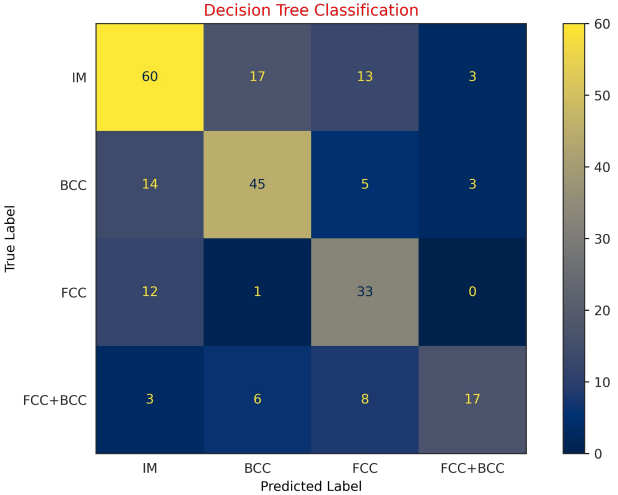
Methodology - Extreme Gradient Boost / Random Forest

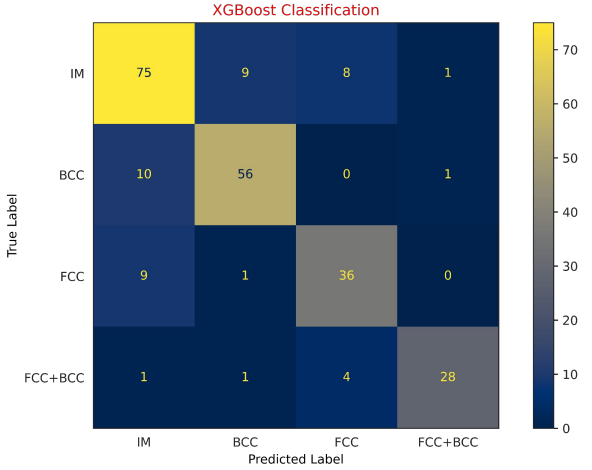
Hyperparameter:

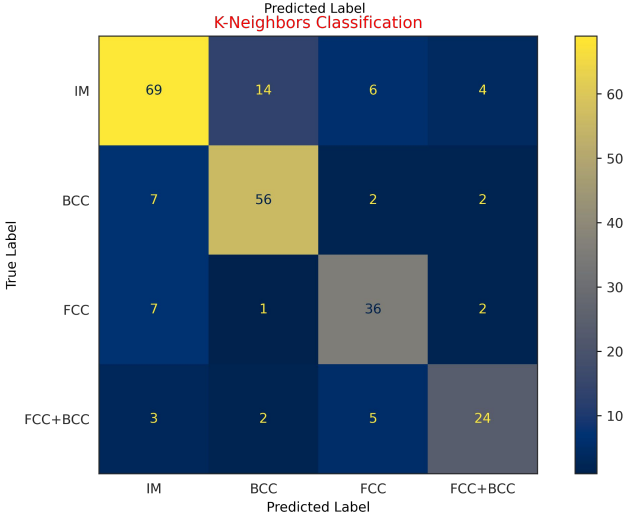
- Number of trees
- Criterion: Gini impurity
- Depth: height of the tree
- Min Sample leaf: least number of samples to be leaf node

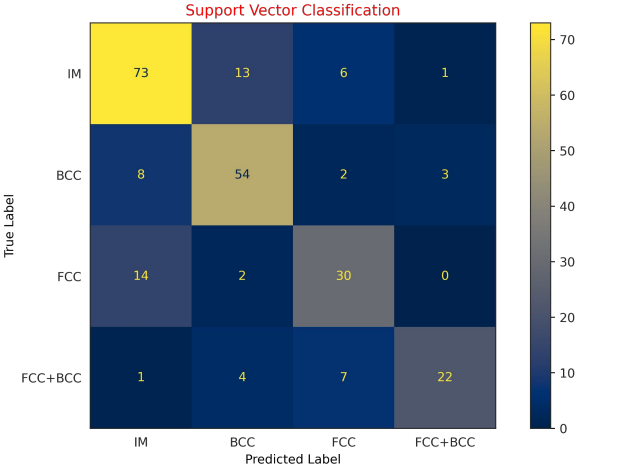
Results: Confusion Matrix



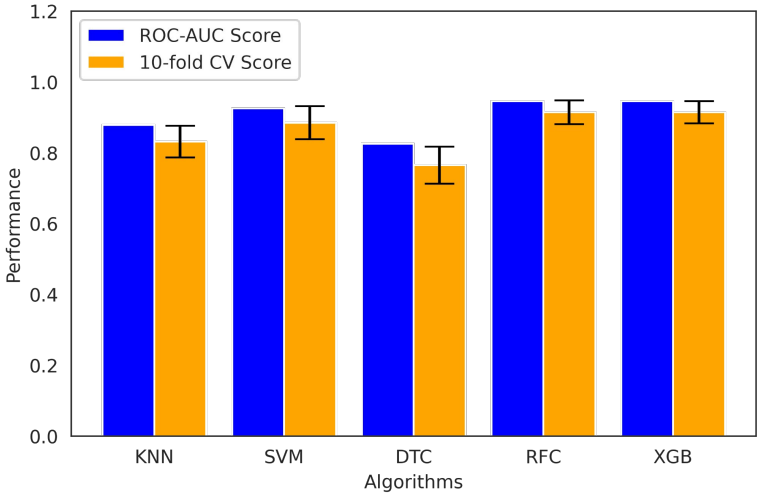


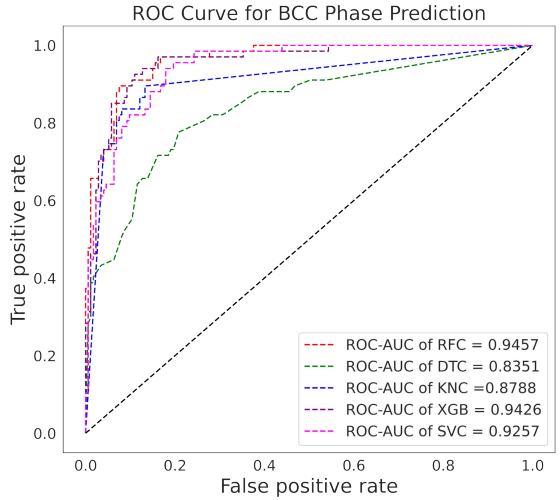


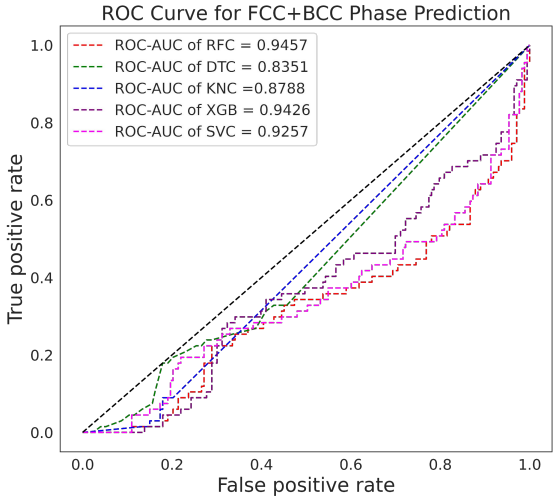


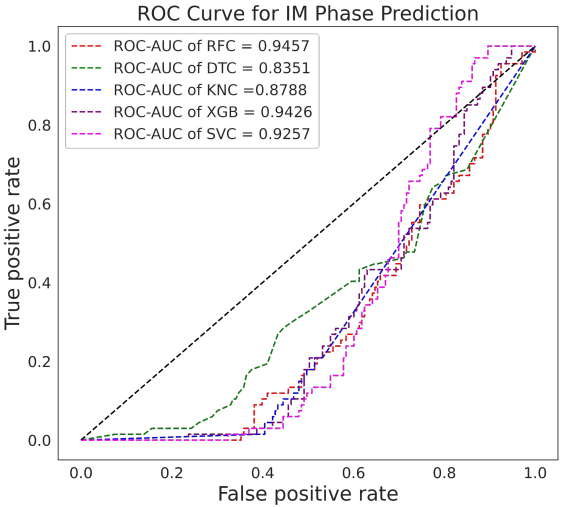


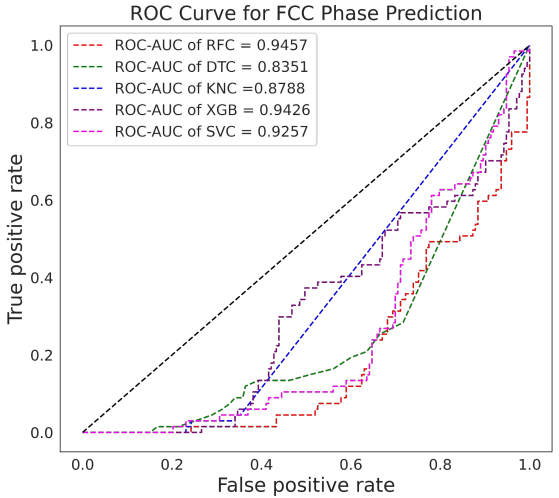
Model Performance











Summary

- HEA are metallic compounds with useful properties, but discovery of novel compositions is laborious and expensive experimentally
- We can use multi-classifiers to predict the phase structure of HEAs
- We clean and prepare the data by removing NaNs and substituting with synthetic values
- The resulting performance is best for the BCC phase

Next Steps

- Future work will focus on:
  - Reduce the complexity of each model to increase prediction accuracy
  - Increase the range over which hyperparameters are queried when tuning
  - Apply the best model towards compositions outside of the dataset
- Bigger picture:
  - Raise model confidence to discover new alloys and their applications
  - Incorporate more data to expand which phases the model can classify

Reference

Original paper:

- Singh, S., Katiyar, N.K., Goel, S. *et al.* Phase prediction and experimental realisation of a new high entropy alloy using machine learning. *Sci Rep* 13, 4811 (2023). https://doi.org/10.1038/s41598-023-31461-7

Data:

- Precker, C.E., Gregores Coto, A., Muíños Landín, S. (2021) Materials for Design Open Repository. High Entropy Alloys. (version 0) [Dataset]. Zenodo. https://zenodo.org/records/5155150