Chapter

Design and Development of High Entropy Alloys Using Artificial Intelligence

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Abstract

The conventional design approach of alloys initiates with one principal element and continues by adding several alloying elements to obtain desired properties. In this method, the intrinsic properties of the designed alloy are governed by the principal element. For example, in steel alloy, iron is the principal element, Aluminium in aluminium alloy, and so on. Compared to the conventional alloy, high entropy alloys do not have any dominating elements; all the elements present in these alloys either have an equal or near-equal ratio of elements. As reported in the literature, these alloys exhibit interesting material properties such as high strength, high hardness, improved elevated temperature strength, and magnetic properties. These characteristics make HEAs a suitable option for high-performance applications in the aero engine, aerospace structures, and machine tools. High entropy alloy has multiple principal elements as shown in schematic diagram 1; it leads to much higher possible compositions than conventional alloys. The huge compositional space provides an opportunity to improve desired mechanical properties. If it is explored through "trial and error," it will be challenging and cumbersome. Therefore, search schemes that can competently and promptly recognize particular alloys with desired properties are essential. Artificial Intelligence is a useful tool to model, discover, and optimize new alloys that enable predicting individual material properties as a function of composition. While the application of Artificial Intelligence is quite popular in many aspects of society, its usage in material informatics is still in the nascent stage. The algorithm used in artificial intelligence is trained to pick up predictive rules from data and create a material model quicker than a computational model and can even generate the model for which no physical model exists. Artificial Intelligence (AI) allows predicting a set of experiments to be conducted to detect new alloy having desired properties. Thus, AI can be used as a valuable tool to optimize the development of new alloys.

Keywords: high entropy alloy, Artificial Intelligence, mechanical properties

1. Introduction

Humans have been using alloys since ancient times. Arsenical Bronze, an alloy of copper and arsenic, was used as far back as the 5th millennium BCE. Sumerians first started alloying copper and tin to form Bronze in the third millennium BC. Bronze was much more challenging than its parent elements. Hence, societies wielding bronze weapons skillfully subjugated their neighboring communities. Meteoric iron, a naturally occurring alloy of iron and nickel, was in use from 3200 BCE.

Persians started using carbon steel in the 16th century BCE. With this began the Iron Age. Our modern society also relies heavily on steel.

Typically, alloys have one or two primary components and other components with small amounts. The primary ingredients are chosen for the alloy's leading properties, and the smaller components are selected for specific additional properties. High entropy alloys' development is essential in resolving the limitations of conventional alloys, especially in extreme temperature and loading conditions. High entropy alloys compared to traditional alloys do not have any dominating elements; all the elements present in these alloys either have an equal or near-equal element. As a result, these types of alloy features, as their name suggests, high entropy, i.e., reducing the Gibbs free energy of solid solution and their high configuration entropy mixing is stable at higher temperatures. These characteristics make HEAs a suitable option for high – performance applications such as gas turbines and other aerospace structures, tools, gears, and bearings.

High entropy alloys (HEA) or multi-component alloys are created from equal or similar proportions of multiple elements. There are generally two agreed-upon definitions of HEAs. It should have more than five elements with concentrations ranging from 5–35% of each component, or the entropy of mixing should be greater than 1.5R, where R is the gas constant. Due to each constituent element being a principal constituent, these alloys are characterized by larger configuration entropy. For a 2-component system, the entropy of mixing is given by Eq. (1).

$$\Delta_{mix} \mathrm{S} = -nR\left(\mathrm{x}_{\mathrm{A}} ln x_{\mathrm{A}} + \mathrm{x}_{\mathrm{B}} ln x_{\mathrm{B}}\right) \tag{1}$$

From **Figure 1**, the entropy increase is maximum when both the components are of equal proportions. It is easy to follow that for multi-component systems; configurational entropy increases with the increasing number of components. It is maximum for any given number of components when all the constituents' proportions are the same. High entropy alloy can be produced by processing in the gas phase, by methods like sputtering or molecular beam epitaxy, in the liquid phase by methods like arc melting, induction melting, and Bridgman solidification, or in the solid phase by methods like mechanical alloying.

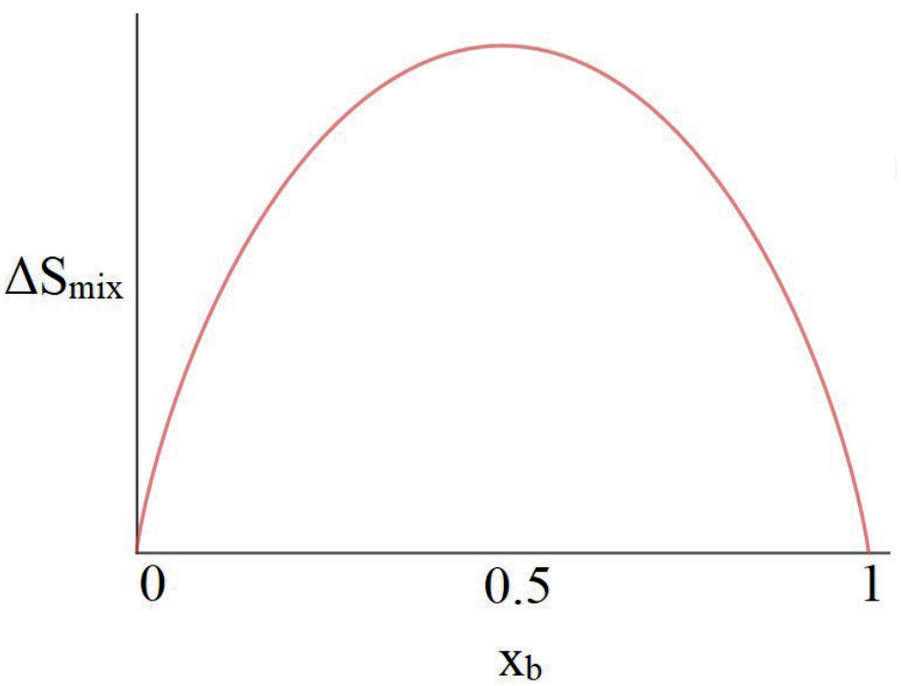

Figure 1.
The illustrated concept of random mixing of elements.

Figure 2 shows tetrahedral compositional space for a 4-element alloy [1]. The corner points represent pure elements. The alloys typically studied lie near the corner points or the edges. One or two primary elements have significant concentration and other elements of minimal concentrations. HEAs are a novel class of materials that lie near the centroid of the compositional space. Research accelerated in this field after the independent publication of Jien-Wei Yeh and Brian Cantor's papers in 2004. HEAs show great strength to weight ratios, fracture resistance, tensile strength, corrosion, and oxidation resistance. HEAs alloy properties are affected by the high-entropy effect, lattice distortion effect, sluggish diffusion effect, and cocktail effect. The crystal structure of HEAs consists of either face-centered cubic (fcc), body-centered cubic (bcc) crystal structure, and hexagonal close-packed (hcp) structure or a mix of any two. VNbMoTaW has excellent refractory properties, CoCrFeMnNi has excellent low-temperature mechanical properties, TaNbHfZrTi shows superconductivity. The reasons for such exceptional properties of HEAs are their microstructures. The most common structures are the Multiphase, single-phase FCC, and single-phase BCC. Generally, when a single-phase HEA is formed, it tends to create a BCC structure, as BCC lattices can accommodate a more extensive range of atomic sizes.

Many techniques are used to explain and guess the different phases of the HEAs based on their parameters, like Valence Electron Concentration (VEC), thermodynamic effects of enthalpy of mixing, and atomic sizes. Phases can be predicted using CALPHAD, molecular dynamics simulations, and kinetic mechanisms. However, using the traditional methods to predict phase formation is very time-consuming and computationally expensive. Density Functional Theory (DFT) is traditionally use to indicate phases by correlating parameters and fitting to existing data. Nevertheless, this method is inadequate given that not much is known about the compositional spaces' central regions. Traditional experimentation with HEAs is done by exploring the compositional and thermodynamic space based on trial-and-error strategies. Recently, Artificial Intelligence (AI) has been used to predict the formation of HEAs. AI use self-learning mechanisms to find patterns in given data. AI is not bound to follow fixed rules in order to obtain the solutions of a specified problem; instead, it solves each unique situation in its way. It is this unique feature

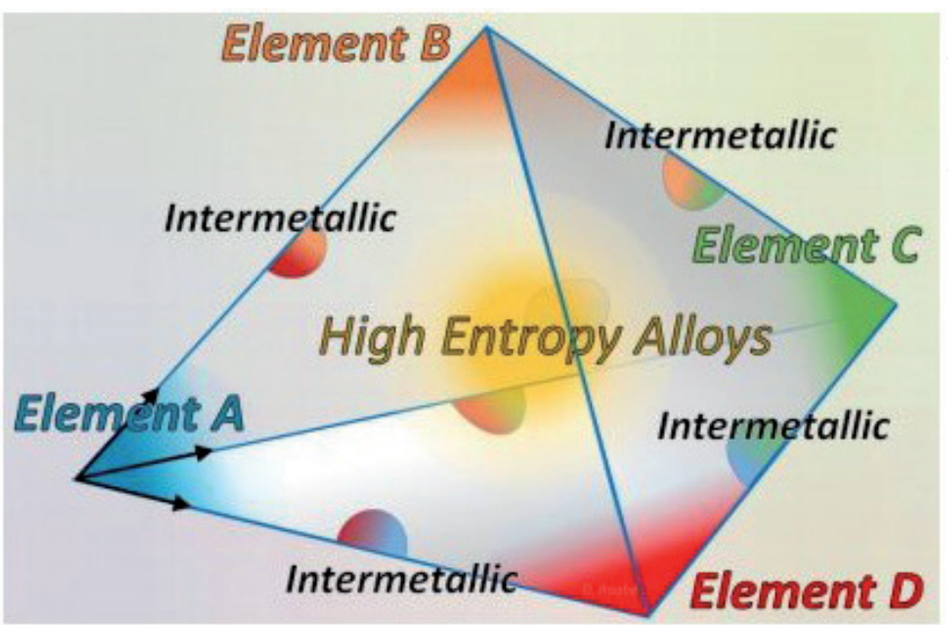

Figure 2.
Compositional space for 4-element alloy [1].

of AI, which can be used to tackle the vagueness of a given system. Phase formation in HEAs is also such a vague system. AI and its subsets, Machine Learning, and Deep Learning can significantly reduce the time component involved in designing HEAs with desired properties by quickly traversing the compositional space.

Adaptive Neuro-Fuzzy Interface System (ANFIS) is an AI algorithm, which is used for phase prediction in FCC, BCC, and multi-phase HEAs. ANFIS is constructed using Artificial Neural Network (ANN) and fuzzy logic. ANN mimics the working of a human brain. Fuzzy logic is a mathematical model that works on degrees of truths, not just absolute truth and false. ANFIS method is suitable to predict the phases in HEAs [2]. Generally two approaches as depicted in **Figures 3** and **4** are used in practice to model the AI problem. The first approach used the constituent elements

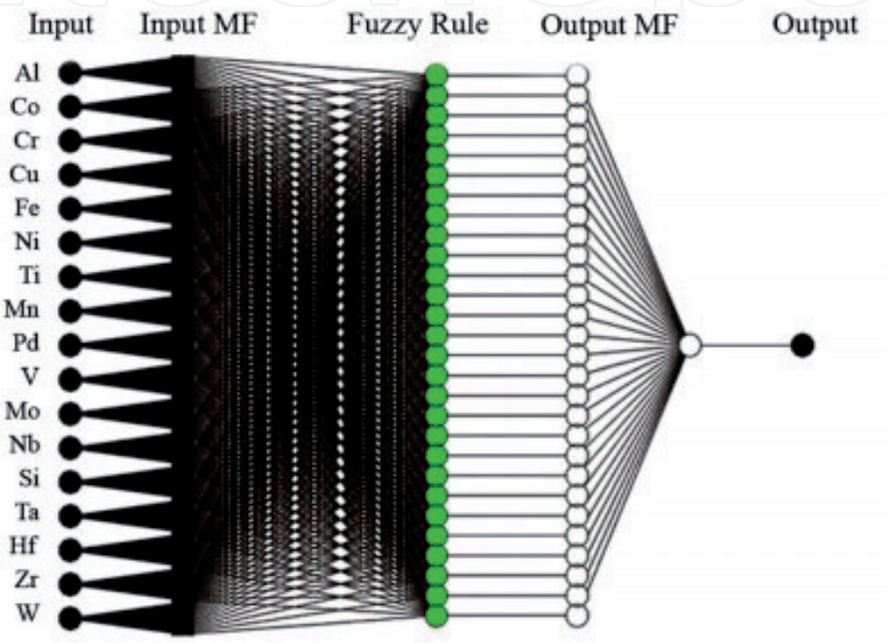

Figure 3.
A framework of ANFIS: Composition-based modeling [2].

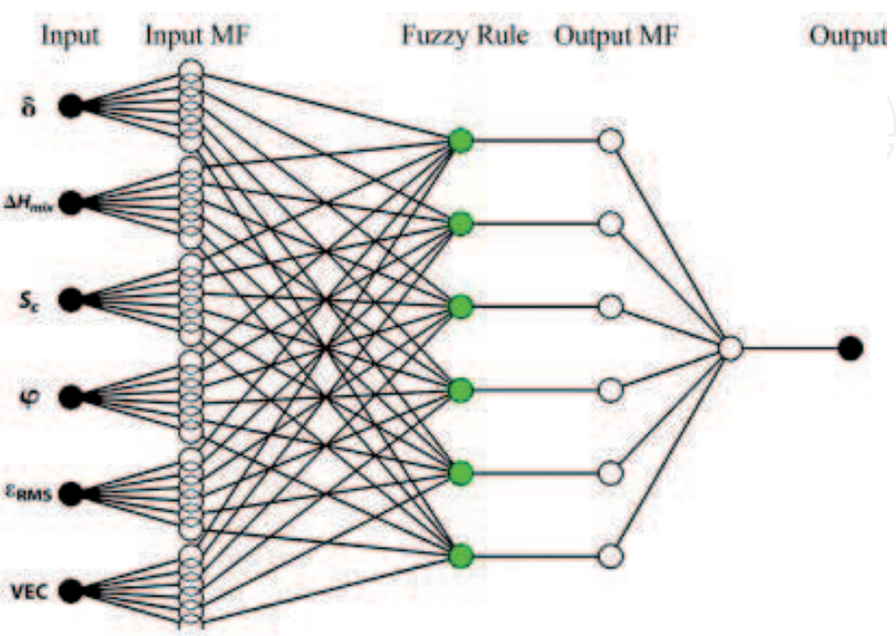

Figure 4.
A framework of ANFIS: Parameter-based modeling [2].

as the inputs, while the second used a set of six crucial parameters in the formation of HEAs. In the first approach, the input elements chosen are Fe, Ni, Ti, Mn, Pd, V, Mo, Nb, Si, Ta, Hf, Zr, and W. In the second approach, atomic size parameter (δ), enthalpy of mixing ($\Delta\Theta_{mix}$), configurational entropy (S_c), single dimensionless thermodynamic parameter (Φ), intrinsic strain (ε_{rms}) and valence electronic concentration (VEC) are the six input parameters. Φ is the thermodynamic parameter; δ and VEC are the parameters of atomic scales; $\Delta\Theta_{mix}$ and S_c are the parameters for nuclear interactions. The first model's accuracy was 84.21%, while it was 80% for the second.

2. Machine learning and design of experiments

Machine Learning (ML) is a subset of Artificial intelligence. ML offers flexibility; as new data becomes available; it is more rapidly able to construct relations between input and output data. Kevin Kaufmann and Kenneth S. Vecchio used an "ML-HEA" to predict the solid solution-forming ability of the HEA using thermodynamic data from ThermoCalc and chemical features with a random forest machine-learning model [3]. They also compared the ML-HEA model with other traditional models like CALPHAD and LTVC, as mention in **Table 1**.

The ML-HEA model correctly predicted the phase for 134 alloy systems with a known phase. This model is in 94% agreement with the CALPHAD model, and 82.1% agreement with the DFT based LTVC model. However, until the central region of the compositional space is further explored, it will be impossible to truly know which of these three models (ML-HEA, CALPHAD, and LTVC) is more accurate in predicting the phase of the HEAs.

Chang et al. used ML with ANN to predict the composition of non-equimolar AlCoCrFeMnNi to get the highest hardness HEAs [4]. In their limited data set of 91 alloys, they also included the alloys of Cu and Mn. The input for the network is the eight elements present in the alloy, and the target output is the hardness of the HEAs. **Figure 5** shows good agreement between the experimental hardness and predicted hardness. Pearson's correlation coefficient = 0.97, R-squared correlation = 0.94, and mean absolute error = 36 Vickers Hardness, and **Table 2** shows good agreement between the predicted hardness and experimental hardness from ML3 to ML5. Here ML3, ML4 and ML5 are high entropy alloy.

Cheng Wen et al. [5] used ML to predict hardness in an AlCoCrCuFeNi HEA system. They used two approaches using the compositions and other descriptive factors such as atomic radii, the difference in electronegativity, VEC, mixing enthalpy, and configurational entropy. They used hardness data of 155 HEAs, with the highest hardness of 735 HV. Pei et al. [6] used machine learning to identify the three most important physical properties of the elements (besides the Hume-Rothery rules) that describe the HEAs' formation. They developed a new method based on these additional features to predict the FCC, BCC or HCP structure of the HEAs.

Material systems	Known solid solution (Neither, FCC, or BCC)	CALPHAD	LVTC
Binaries	117 of 117 (100%)	110 of 117 (94%)	102 of 117 (87.2%)
Ternaries	N/A	362 of 441 (82.1%)	279 of 441 (63.3%)
Quaternaries	8 of 8 (100%)	N/A	690 of 1110 (62.2%)
Quinaries	9 of 9 (100%)	N/A	94 of 130 (72.3%)

Table 1.
Comparison of ML-HEA with CALPHAD and LVTC [3].

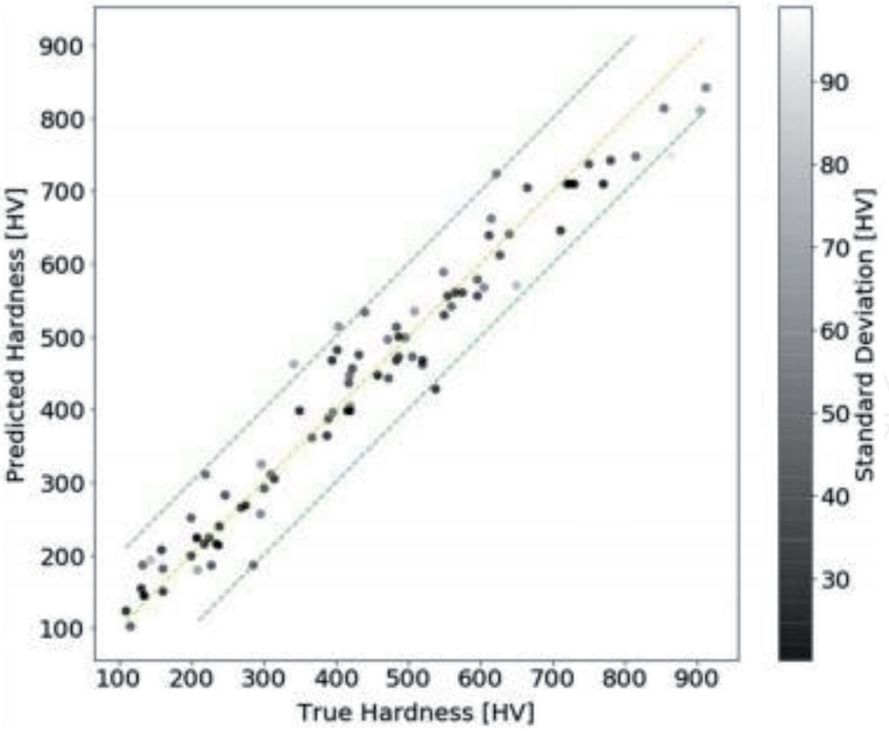

Figure 5.
Predicted versus proper hardness for HEAs, where different colors indicate the variation of the standard deviation [4].

Alloy	Al	Co	Cr	Fe	Mn	Ni	Experimental hardness	Predicted hardness (HV)
Al03	5.66	18.87	18.87	18.87	18.87	18.87	125 ± 3	242 ± 98
ML1	11	18	22	22	5	22	198 ± 6	303 ± 38
ML2	30.5	16	18.5	16.5	5	13.5	522 ± 8	505 ± 35
ML3	30	6	35	6	18	5	605 ± 14	670 ± 94
ML4	25.5	9	35	10	15.5	5	628 ± 13	670 ± 111
ML5	24	18	35	10	7.5	5.5	650 ± 12	670 ± 98

Table 2.
Nominal composition (%) of Al03 and ML alloys and their hardness values [4].

They used a large dataset of 1252 HEAs to train a model to predict the formation of phases with an accuracy of 93%. For each constituent element of each HEA, they considered 85 elemental properties. From these 85 properties, they can identify the three most essential quantities: molar volume, bulk modulus, and melting temperature. They used these quantities to develop a model based on the physical nature of the elements. This new model was 73% accurate in predicting the phase of the HEAs. **Figure 6** is a hardness map plotted with the variation of the element Al, Co, and Mn contents. The availability of data machine learning technique for alloy design can be a valuable tool to predict HEAs' properties and constituent elements.

Similarly, **Figure 7** used ML and the Design of experiment approach for the accelerated design of HEAs. The first machine-learning model is trained with an existing dataset consisting of hardness and composition of constituent elements. The model is applied to a search in the materials space for unknown value hardness. Based on hardness predictions, a function to select the next set of experiments is performed—this helps to choose an alloy for experimentation.

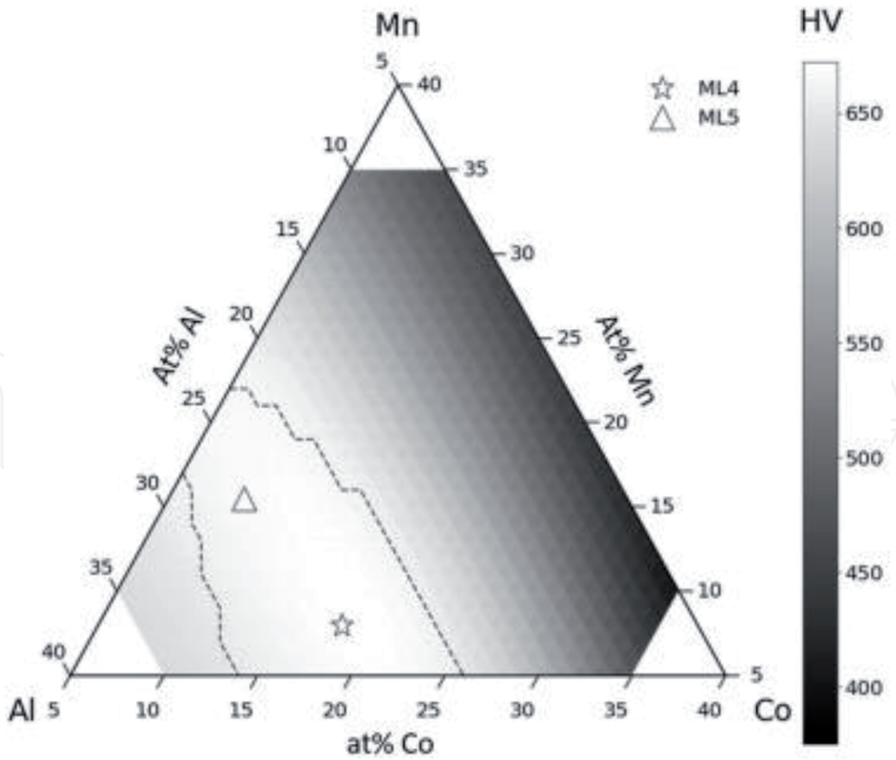

Figure 6.
Hardness plot with a variation of Al, Co, and Mn contents and fixed Co content (black dashed line is the boundary of<30% risk) [6].

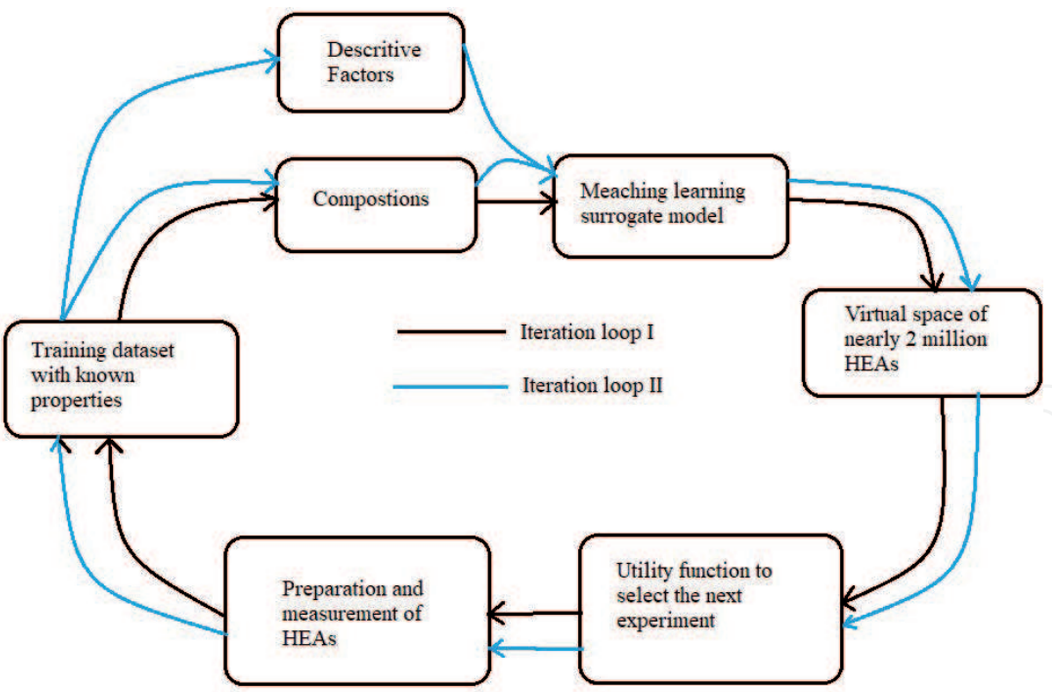

Figure 7.
A schematic of the ML and DOE based iterative design loop for accelerated design of HEAs.

3. Methodology for implementation of AI in HEAs

Figure 8 described the proposed methodology for the implementation of AI to develop a new high entropy alloy. AI algorithm create an AI model by training on the experimental dataset for training, and then this trained AI model is fed with the

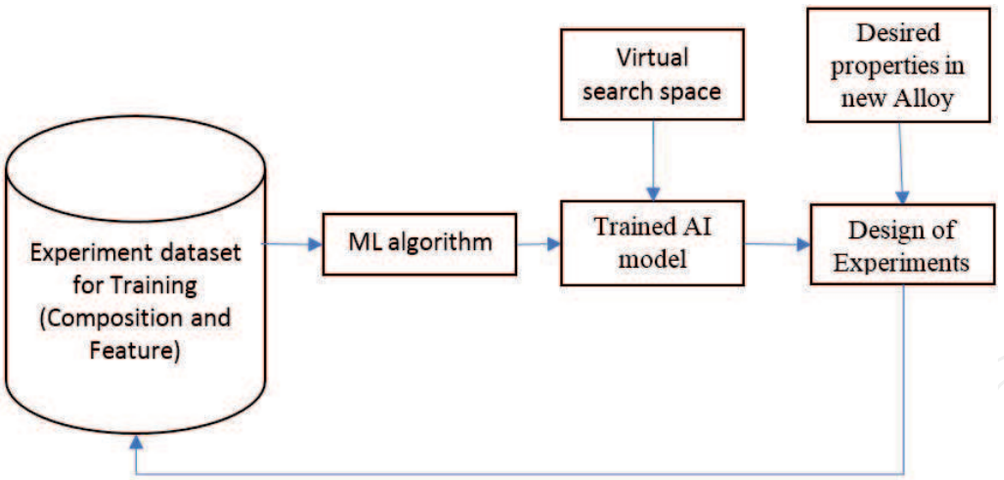

Figure 8.
Methodology for the implementation of AI for the discovery of new alloy.

desired properties of the new model. The output of the AI model is a set of experiments for the creation of a new alloy. The set of experiments suggested by AI model is a subset of an extensive set that will result from all possible combinations of input parameters. Presently trial and error method is used to narrow down this search space, but the use of AI model can lead to selecting a more appropriate subset from this search space. Further, to evolve the AI model with time output of each suggested experiment should be added to the experimental dataset for training and a new AI model is created after each update of the dataset.

The strategy to design and develop of high entropy alloy for achieving the desired strength in the material using the artificial intelligence approach is described below with a detailed explanation:

a. Dataset creation: Data of experiments conducted to create new HEA can be obtained from available literature to create a dataset. This database need to be divided into three subsets (60:20:20): training, validation, and test dataset.

b. Selection of machine learning (ML) algorithm: For ML algorithm, various options like linear regression model, support vector machine, artificial neural network, and deep neural networks should be explored. Starting with the most straightforward algorithm viz. linear regression, each algorithm's performance after training should be evaluated on training and validation dataset (via a loss function like RMSE for expected and actual value of hardness) using k-fold cross-validation. Based on its performance and complexity the ML algorithm can be selected.

c. Training of AI model: AI model created based on selecting the ML algorithm is to be fed with known composition data from the training dataset. The output of this model is the estimated value for the hardness of HEA. Several epochs of training is to be needed to arrive at an acceptable trained AI model.

d. Apart from composition data several physical properties of constituent elements like the difference in atomic radii between composition elements, the difference in electronegativity between components, the valence electron concentration, the mixing enthalpy, the configurational entropy, the U parameter (which is related to the entropy, enthalpy, and the melting point), the L parameter (which is associated with an atom's configuration on a lattice

and its radius) and the g parameter (the solid angles of atomic packing for the elements with the most significant and most minor atomic sizes) are related to the intrinsic properties and affect the final hardness. So further AI model should also be trained by accepting both composition data and physical properties as inputs.

e. Training and test datasets with bootstrapping should be used for training the AI model. A bootstrap training set can be generated by resampling the original training data and testing data with replacement. On each made training set, an AI model is created. Each of these AI models can then run on the original training dataset to give different hardness values for each training sample. This give the output as estimated hardness and standard deviation for the estimated hardness.

f. Use of AI model in the design of experiments: By assuming an available range of values for each constituent element, a virtual search space can be created for experiments to conducted of creation of new HEA. Using this virtual space as input to generated AI models, an estimate for hardness and corresponding confidence (standard deviation) can be computed. Candidates in virtual space can then be selected for desired hardness and sorted standard deviation value for the design of experiments.

4. Summary

Artificial Intelligence is a valuable tool to model, discover, and optimize new alloys that enable predicting individual material properties as a function of composition. Artificial Intelligence (AI) allows the prediction of a set of experiments to be conducted to discover new alloy having desired properties. Thus, AI can be used as an effective tool to optimize the development of new alloys. The use of AI for predicting phase, hardness, and other HEAs properties is a gateway to many possibilities. It is the flexibility of AI to adapt to new data that enables further improvement of the models' accuracy, which will lead to a better understanding of phases of HEAs.

Synthesizing HEAs is a complicated and expensive process, so using traditional modeling to explore compositional space and synthesizing each alloy is impractical. AI can be used so that it is not necessary to explore the entire composition space. AI models can handle voluminous experiment data and are bound to outperform human counterparts. Therefore, AI can be effectively employed to competently and promptly recognize particular High Entropy alloys with desired mechanical properties.

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References

[1] E. P George, D. Raabe, and R O Ritchie: High Entropy alloy, Nature Review, Vol.4, (2019), pp, 515-534

[2] A.Agarwal, and A. K. Prasada Rao: Artificial Intelligence Predicts Body-Centered-Cubic and Face-Centered-Cubic Phases in High-Entropy Alloys. JOM , Vol. 71, (2019) pp.3424-3432.

[3] K Kaufmann, and K. S. Vecchio: Searching for high entropy alloys: A machine learning approach, Acta Materialia, Volume 198, 2020, pp. 178-222.

[4] Y J Chang, C Y Jui, W J Lee: Prediction of the Composition and Hardness of High-Entropy Alloys by Machine Learning. JOM, Vol. 71, 2019, pp.3433-3442

[5] Cheng Wen, Yan Zhang, Changxin Wang, Dezhen Xue, Yang Bai, Stoichko Antonov, Lanhong Dai, Turab Lookman, and Yanjing Su: Machine learning assisted design of high entropy alloys with desired property, Acta Materialia, Volume 170, 2019, pp. 109-117.

[6] Pei, Z., Yin, J., Hawk, J.A. et al. Machine-learning informed prediction of high-entropy solid solution formation: Beyond the Hume-Rothery rules. npj Comput Mater 6, Vol.50 2020, pp.1-8.