

Review

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# Data-driven design of eutectic high entropy alloys

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## Abstract

Eutectic high entropy alloys (EHEAs) have attracted tremendous research interest over the past decade due to their superior physical and mechanical properties. Given the compositional complexity, there are no well-established phase diagrams for EHEAs. Therefore, the compositional design of EHEAs has been following a trial-and-error empirical approach, which is time-consuming, costly, and ineffective. To accelerate the search for EHEAs, data-driven approaches, particularly machine learning (ML) based modeling, have recently been utilized in lieu of the traditional empirical approach. In this article, we provide a critical overview of the recent efforts in the design and development of EHEAs, which covers the various empirical methods and the state-of-the-art machine learning models developed for EHEAs. In addition, we also briefly discuss the mechanical properties and plasticity strengthening mechanisms in EHEAs which are related to their heterogeneous microstructure, such as heterogeneous deformation induced strengthening, twinning induced strengthening, and phase transformation induced strengthening.

**Keywords:** Eutectic alloys, high entropy alloys, machine learning, alloy design, mechanical properties

## INTRODUCTION

Eutectic alloy, in which at least two phases form and grow in a coupled manner during solidification, has attracted immense attention and interest in both academia and industries in past decades<sup>[1,2]</sup>. The term



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“eutectic” was coined by Guthrie in 1884 to refer to easy melt (i.e., the minima on a liquidus curve)<sup>[3]</sup>. Since the melting temperature of a eutectic alloy is always lower than those of its constituent phases, eutectic alloys often exhibit good castability. Additionally, their eutectic microstructures can be readily tailored through thermomechanical processing for optimized properties, such as high rupture strength<sup>[4-6]</sup>, good high-temperature creep resistance<sup>[7-9]</sup>, high thermal conductivity<sup>[10,11]</sup> and superior wear and corrosion resistance<sup>[12-15]</sup>. Compared to single-phase alloys, eutectic alloys usually possess balanced mechanical properties that can meet demanding requirements in structural applications<sup>[1,16-18]</sup>.

To date, a number of eutectic alloy systems have already been developed for various applications, such as Sn-Pb as solder joints<sup>[19-21]</sup>, Sn-Ag for electronics<sup>[22-24]</sup>, Ni-Si for magnetics<sup>[25,26]</sup>, In-Ga for optics<sup>[27,28]</sup> and Ni-Al-Cr for aerospace engineering<sup>[29,30]</sup>. However, just like many other conventional alloys, the design of conventional eutectic alloys is usually based on one principal element. If the phase diagram is not available, alloying elements are then added in a trial-and-error manner to pinpoint a eutectic composition. While people have been following such a design strategy for decades, this approach is costly, time-consuming, and inefficient, particularly when it comes to compositionally complex alloys (CCAs), such as multi-principal-element alloys (MPEAs)<sup>[31-33]</sup> and high entropy alloys (HEAs)<sup>[34,35]</sup>, for which there is no phase diagram and the associated compositional space is too broad to navigate with the traditional design strategy.

In the past, the development of HEAs in their infancy was mainly focused on the formation of single-phase solid solutions<sup>[36-41]</sup>, while the recent trend has shifted to multi-phase HEAs with balanced mechanical properties<sup>[42-44]</sup>. One good example is the so-called eutectic high entropy alloys (EHEAs) comprising a biphasic or triphasic microstructure with a lamellar or rod morphology<sup>[1,45]</sup>. By carefully controlling the thermal and mechanical processing, EHEAs can exhibit a wide range of microstructural feature sizes, ranging from tens of nanometers to a few microns<sup>[46-49]</sup>. Owing to these heterogeneous eutectic micro- or nanostructures, EHEAs can attain high strength and good ductility<sup>[46,47,50,51]</sup>, remarkable creep resistance<sup>[45,52]</sup>, superior thermal stability at elevated temperatures<sup>[53-55]</sup> and good processability<sup>[56]</sup>.

Given the vast hyper-dimensional compositional space for EHEAs, the traditional design strategy becomes impractical. Therefore, several design methods were proposed recently to quickly locate the possible eutectic or near-eutectic composition in the compositional space. These include, the simple mixing method<sup>[57-60]</sup>, the grouping method<sup>[61-63]</sup>, the pseudo-binary methods<sup>[64-67]</sup> and the so-called “LEGO” method<sup>[68]</sup>. The pros and cons of these methods are listed in Table 1 and they can be categorized into two strategies, as summarized in Figure 1. However, we note that these methods are mostly empirical; therefore, the related experimental workload that one has to pay is heavy to verify these empirical predictions as the number of constituent elements increases. To improve efficiency, people usually turned to machine learning (ML) as an alternative to the traditional design strategy for the development of advanced alloys (i.e., titanium alloys<sup>[69,70]</sup>, copper alloys<sup>[71,72]</sup>, shape memory alloys<sup>[73-75]</sup> and even metallic glasses<sup>[76-78]</sup>). Recently, these efforts were extended to the design of EHEAs<sup>[79-81]</sup>. In the present work, we provide a critical overview of these recent efforts for the development of EHEAs covering the empirical design methods and the data-driven methods.

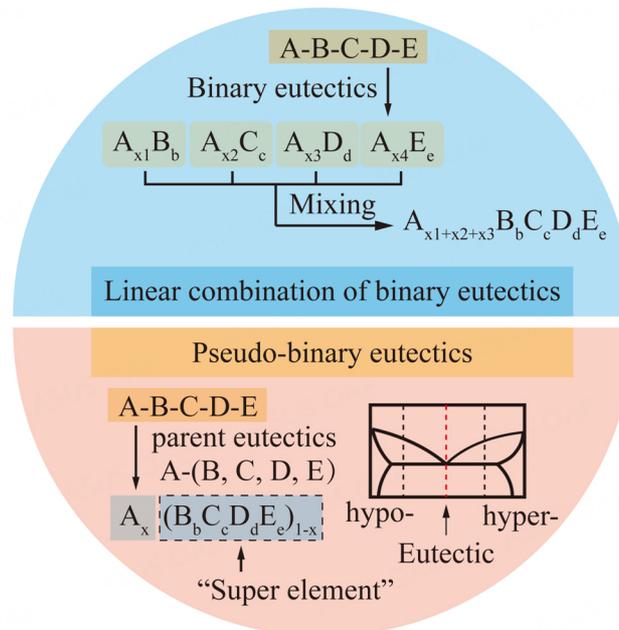
## EMPIRICAL DESIGN METHODS OF EUTECTIC HIGH ENTROPY ALLOYS

### Linear combination of binary eutectics

Without a phase diagram, it is non-trivial and difficult to locate the eutectic compositions for multi-component alloys and HEAs. Therefore, to facilitate the design of EHEAs, one strategy is to resort to the phase diagrams of binary eutectics, which can be easily found in the literature<sup>[57,59,60,68]</sup>. In other words, there is a hypothesis that the eutectic microstructures of multi-component systems may inherit from some binary/ternary eutectics<sup>[68,82,83]</sup>. While this hypothesis remains to be verified theoretically, it indeed provides

**Table 1. Pros and cons of the empirical methods for EHEA design**

Methods	Pros	Cons
Simple mixing method "LEGO" method	Take binary eutectics as the constituents, which can be easily located from phase diagrams	(1) Lack of supportive physical and thermodynamic theories, which significantly reduces the efficiency
Pseudo-binary method Grouping method	With the help of CALPHAD, a pseudo-binary system can be quickly constructed and evaluated	(2) The explorable space following empirical methods is limited compared with the vast compositional space of HEAs

**Figure 1.** The schematic for the various empirical design methods for eutectic high entropy alloys.

an avenue to locate EHEA compositions. In practice, one can add the weighted compositions of binary eutectics as a candidate composition  $C_{eutectic}$  for a multi-component alloy, which may be formulated as follows:

$$C_{eutectic} = \sum x_i C_{A_i B_i} \quad (1)$$

where  $C_{eutectic}$  stands for the eutectic composition;  $A_i$  and  $B_i$  stand for the constituent element of the  $i^{th}$  binary eutectic, while  $x_i$  is the weighting factor. Following this line of reasoning, many EHEAs or multi-component eutectic compositions were discovered, including  $Nb_{0.45}CoCrFeNi$ <sup>[57]</sup>,  $Nb_{0.8}Co_{1.74}Fe_{2.82}Ni$ <sup>[60]</sup>,  $Nb_{0.5}CoFeNi$ <sup>[84]</sup> and  $Nb_{0.5}CoCrFeNi$ <sup>[85,86]</sup>, etc. Here we emphasized that the values of  $x_i$  are not fixed; however, in the literature, they were mostly set equal or nearly equal in a heuristic manner such that the overall composition was close to an equiatomic or near-equiatomic composition, as listed in [Table 2](#).

### Pseudo-binary eutectics

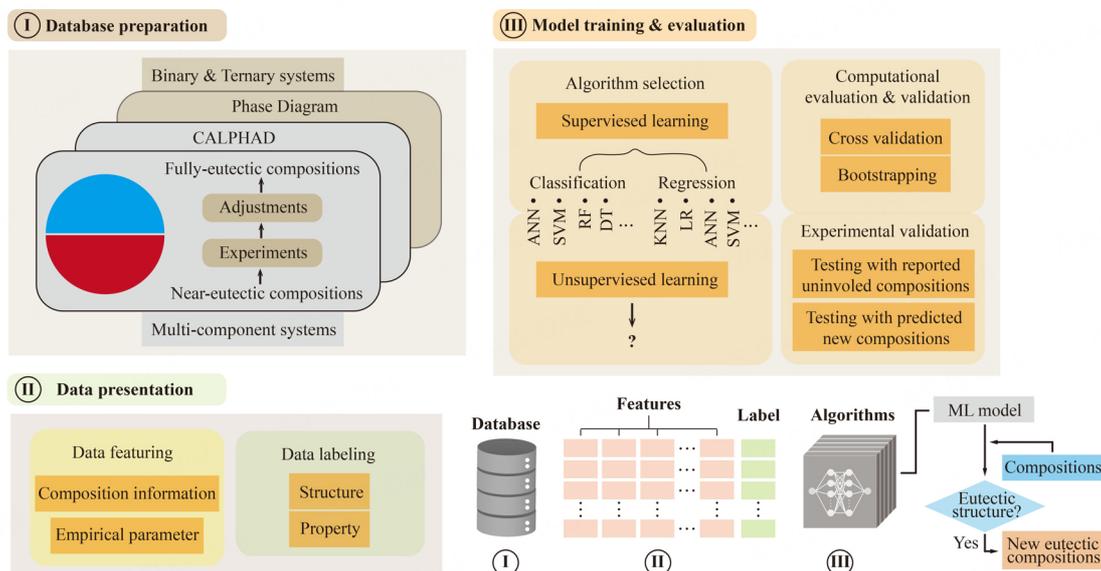
Since mixing of elements with a similar atomic size tends to form solid solutions<sup>[89,90]</sup>, such as  $CoCrNi$ <sup>[91,92]</sup>,  $CoCrFeNi$ <sup>[93]</sup>, and  $CoCrFeNi_2$ <sup>[94]</sup>, one alternative way to design EHEAs is to view these solid solutions as a

**Table 2. EHEA compositions identified via Equation (1) compared with the experimental verifications**

Alloy system	$C_{A_i B_i}$	$x_i$	$C_{eutectic}$ (calculated)	$C_{eutectic}$ (experimental)	Ref.
Nb-Co-Cr-Fe-Ni	$Nb_{13.9}Co_{86.1}$	0.25	$Nb_{0.6}CoCrFeNi$	$Nb_{0.45}CoCrFeNi$	[57]
	$Nb_{12}Cr_{88}$	0.25			
	$Nb_{10.6}Fe_{89.4}$	0.25			
	$Nb_{15.5}Ni_{84.5}$	0.25			
Ta-Co-Cr-Fe-Ni	$Ta_8Co_{92}$	0.25	$Ta_{0.47}CoCrFeNi$	$Ta_{0.4}CoCrFeNi$	[57]
	$Ta_{13}Cr_{87}$	0.25			
	$Ta_{7.5}Fe_{92.5}$	0.25			
	$Ta_{13.7}Ni_{86.3}$	0.25			
Zr-Co-Cr-Fe-Ni	$Zr_{9.5}Co_{90.5}$	0.25	$Zr_{0.51}CoCrFeNi$	$Zr_{0.55}CoCrFeNi$	[57]
	$Zr_{17.2}Cr_{82.8}$	0.25			
	$Zr_{9.8}Fe_{90.2}$	0.25			
	$Zr_{8.8}Ni_{91.2}$	0.25			
Hf-Co-Cr-Fe-Ni	$Hf_{11}Co_{89}$	0.25	$Hf_{0.49}CoCrFeNi$	$Hf_{0.4}CoCrFeNi$	[57]
	$Hf_{13}Cr_{87}$	0.25			
	$Hf_{7.9}Fe_{92.1}$	0.25			
	$Hf_{12.5}Ni_{87.5}$	0.25			
Nb-Co-Fe-Ni	$Nb_{13.9}Co_{86.1}$	0.29	$Nb_{0.62}Co_{1.22}Fe_{1.98}Ni$	$Nb_{0.62}Co_{1.22}Fe_{1.98}Ni$	[60]
	$Nb_{10.6}Fe_{89.4}$	0.46			
	$Nb_{15.5}Ni_{84.5}$	0.25			
Nb-Co-Fe-Ni	$Nb_{13.9}Co_{86.1}$	0.29	$Nb_{1.30}Co_{1.74}Fe_{2.82}Ni$	$Nb_{0.80}Co_{1.74}Fe_{2.82}Ni$	[60]
	$Nb_{10.6}Fe_{89.4}$	0.46			
	$Nb_{40.5}Ni_{59.5}$	0.25			
Zr-Co-Fe-Ni	$Zr_{9.5}Co_{90.5}$	0.29	$Zr_{0.43}Co_{1.19}Fe_{1.94}Ni$	$Zr_{0.53}Co_{1.19}Fe_{1.94}Ni$	[60]
	$Zr_{9.8}Fe_{90.2}$	0.47			
	$Zr_{8.8}Ni_{91.2}$	0.24			
Hf-Co-Fe-Ni	$Hf_{11}Co_{89}$	0.28	$Hf_{0.47}Co_{1.22}Fe_{2.11}Ni$	$Hf_{0.47}Co_{1.22}Fe_{2.11}Ni$	[60]
	$Hf_{7.9}Fe_{92.1}$	0.48			
	$Hf_{12.5}Ni_{87.5}$	0.24			
Nb-Co-Fe-Ni	$Nb_{13.9}Co_{86.1}$	0.33	$Nb_{0.49}CoFeNi$	$Nb_{0.5}CoFeNi$	[84]
	$Nb_{10.6}Fe_{89.4}$	0.33			
	$Nb_{15.5}Ni_{84.5}$	0.34			
Nb-Co-Cr-Fe-Ni	$Nb_{13.9}Co_{86.1}$	0.25	$Nb_{0.6}CoCrFeNi$	$Nb_{0.5}CoCrFeNi$	[85,86]
	$Nb_{12}Cr_{88}$	0.25			
	$Nb_{10.6}Fe_{89.4}$	0.25			
	$Nb_{15.5}Ni_{84.5}$	0.25			
$Al_2O_3$ - $Y_2O_3$ - $ZrO_2$	$(Al_2O_3)_{43}(Y_2O_3)_{57}$	0.35	$(Al_2O_3)_{44}(Y_2O_3)_{16}(ZrO_2)_{19}$	$(Al_2O_3)_{65}(Y_2O_3)_{16}(ZrO_2)_{19}$	[87]
	$(Al_2O_3)_{63}(ZrO_2)_{37}$	0.65			
$Al_2O_3$ - $Gd_2O_3$ - $ZrO_2$	$(Al_2O_3)_{77}(Gd_2O_3)_{23}$	0.57	$(Al_2O_3)_{102}(Gd_2O_3)_{19}(ZrO_2)_{23}$	$(Al_2O_3)_{58}(Gd_2O_3)_{19}(ZrO_2)_{23}$	[88]
	$(Al_2O_3)_{63}(ZrO_2)_{37}$	0.43			

“super element” and to substitute them for a normal element in a binary eutectic. In doing so, the candidate composition  $C_{eutectic}$  may be expressed as:

$$C_{eutectic} \in [(C_I)_{1-x}(C_{II})_x], x \in (0, 1) \quad (2)$$



**Figure 2.** Schematic of a data-driven approach for the design of eutectic high entropy alloys.

where  $C_I$  denotes the “super element” (e.g., the mixture of elements that may form a solid solution) and  $C_{II}$  the single element in a pseudo-binary system. Following Equation (2), a number of EHEA compositions were discovered with the aid of CALPHAD<sup>[64-67,95-102]</sup>, as tabulated in Table 3. Here,  $x$  is the composition yet to be determined via experiments or CALPHAD.

With CALPHAD, one can obtain the so-called pseudo-binary phase diagram as a function of  $x$  and temperature, through which fully-eutectic compositions can be identified, if any, for  $C_{eutectic}$ . For example, Wu *et al.*<sup>[109]</sup> identified the near-eutectic composition  $Al_{19.4}Co_{20.6}Cr_{20.6}Ni_{39.4}$  for the pseudo-binary  $(CoCrNi)_{1-x}(AlNi)_x$  alloy, which is very close to the eutectic composition of  $Al_{17.4}Co_{21.7}Cr_{21.7}Ni_{39.2}$  verified experimentally. In light of the Scheil solidification theory, Yurchenko *et al.*<sup>[110]</sup> also successfully found the  $Al_{28}Cr_{20}Nb_{15}Ti_{27}Zr_{10}$  EHEA. However, we note that all these above-mentioned methods are semi-empirical since CALPHAD is also based on the available database. Regardless of the difference in these methods, a more general method is always desirable, which can be applied to a wide range of compositions. In addition to the above-mentioned eutectic high entropy alloys, people also developed a number of eutectic refractory high entropy alloys<sup>[105-107]</sup>, eutectic soldering high entropy alloys<sup>[108]</sup> and eutectic high entropy ceramics<sup>[87,88]</sup>. Interestingly, some of these eutectics could also be designed based on the aforementioned empirical rules. Therefore, we also list them in Tables 2 and 3 for the sake of completeness.

## DATA-DRIVEN METHODS FOR THE DESIGN OF EUTECTIC HIGH ENTROPY ALLOYS

### Database

In recent years, ML has been widely used to accelerate the search for advanced alloys<sup>[69-78]</sup>. As a data-driven approach, the performance of ML models is highly dependent on the quantity and quality of data<sup>[111,112]</sup>. Figure 2 illustrates the workflow of a typical ML approach to designing EHEAs. While EHEAs are important and very useful, we note that only a limited number of EHEA compositions are located out of vast compositional space with the ML approach [Table 4]<sup>[79-81,113]</sup>. While one can easily find the data of binary and ternary eutectics from their corresponding phase diagrams, the data for EHEAs mainly comes from the literature, including those found through the empirical methods illustrated in Figure 1 and/or the results of CALPHAD calculations. Here, we note that the CALPHAD calculations are performed with

**Table 3. EHEA compositions identified via Equation (2)**

$C_I$	$C_{II}$	Parent binary eutectics	Pseudo-binary system	$C_{eutectic}$ (experimental)	Ref.
CoCrNi	Ta	(Co, Cr, Ni)-Ta	$(CoCrNi)_{1-x}Ta_x$	$CoCrNiTa_{0.4}$	[67]
	Nb	(Co, Cr, Ni)-Nb	$(CoCrNi)_{1-x}Nb_x$	$CoCrNiNb_{0.4}$	[100]
CoFeNi	NiAl	(Co, Ni)-Al	$(CoFeNi)_{1-x}(NiAl)_x$	$CoFeNi(NiAl)_{0.92}$	[101]
CoCrFeNi	Ta	(Co, Cr, Fe, Ni)-Ta	$(CoCrFeNi)_{1-x}Ta_x$	$CoCrFeNiTa_{0.75}$	[66]
				$CoCrFeNiTa_{0.43}$	[95]
	Nb	(Co, Cr, Fe, Ni)-Nb	$(CoCrFeNi)_{1-x}Nb_x$	$CoCrFeNiNb_{0.65}$	[64]
	Zr	(Co, Cr, Fe, Ni)-Zr	$(CoCrFeNi)_{1-x}Zr_x$	$CoCrFeNiZr_{0.5}$	[102]
	Hf	(Co, Cr, Fe, Ni)-Hf	$(CoCrFeNi)_{1-x}Hf_x$	$CoCrFeNiHf_{0.4}$	[103]
CoCrNi <sub>2</sub>	(V, B, Si)	(Co, Ni)-Mo	$(CoCrFeNi)_{1-x}Mo_x$	$CoCrFeNiMo_{0.8}$	[99]
		(Co, Ni)-V	$(CoCrNi_2)_{1-x}(V, B, Si)_x$	$CoCrNi_2(V_2B)_{0.43}$	[97]
		(Co, Cr, Ni)-B (Co, Cr, Ni)-Si		$CoCrNi_2(V_3B_2Si)_{0.2}$	[97]
CoCrFeNi <sub>2</sub>	(V, B, Si)	(Co, Ni)-V	$(CoCrFeNi_2)_{1-x}(V, B, Si)_x$	$CoCrFeNi_2(V_2B)_{0.51}$	[97]
		(Co, Cr, Fe, Ni)-B (Co, Cr, Fe, Ni)-Si		$CoCrNi_2(V_6B_3Si)_{0.149}$	[97]
	$Ni_{0.8}Al_{1.2}$	(Co, Fe, Ni)-Al	$(CoCrFeNi_2)_{1-x}(Ni, Al)_x$	$CoCrFeNi_2(Ni_{0.8}Al_{1.2})$	[65]
Co <sub>2</sub> CrFeNi	$Ni_{0.8}Al_{1.2}$	(Co, Fe, Ni)-Al	$(Co_2CrFeNi)_{1-x}(Ni, Al)_x$	$Co_2CrFeNi(Ni_{0.8}Al_{1.2})$	[65]
CoCrFe <sub>2</sub> Ni	$Ni_{0.8}Al_{1.2}$	(Co, Fe, Ni)-Al	$(CoCrFe_2Ni)_{1-x}(Ni, Al)_x$	$CoCrFe_2Ni(Ni_{0.8}Al_{1.2})$	[65]
Ni <sub>2</sub> AlTi	V	Ni-V	$(Ni_2AlTi)_{1-x}V_x$	$(Ni_2AlTi)_{68}V_{32}$	[104]
CrNbTiZr	Al	(Nb, Zr)-Al	$(CrNbTiZr)_{1-x}Al_x$	$(CrNbTiZr)_{0.25}Al_{0.75}$	[105]
HfMo <sub>0.5</sub> NbTiV <sub>0.5</sub>	Si	(Hf, Mo, Nb, Ti, V)-Si	$(HfMo_{0.5}NbTiV_{0.5})_{1-x}Si_x$	-	[106]
HfCo	NbMo	Co-(Nb, Mo)	$(HfCo)_{1-x}(NbMo)_x$	$(HfCo)_{0.75}(NbMo)_{0.25}$	[107]
GaInSn	Zn	(Ga, In, Sn)-Zn	$(GaInSn)_{1-x}Zn_x$	-	[108]

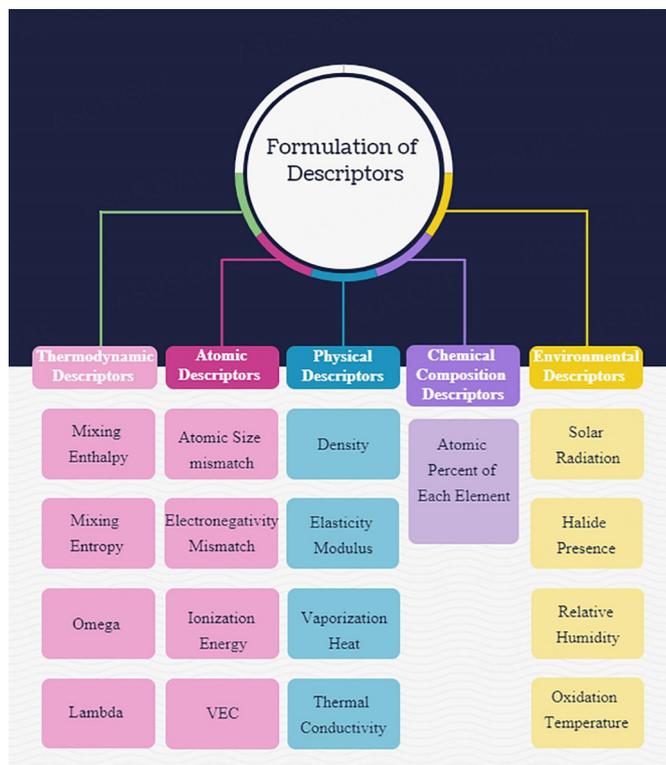
**Table 4. EHEA compositions identified via machine learning**

Alloy	Database	Features	Label	Algorithm	Ref.
$Al_{18}Co_{30}Cr_{10}Fe_{10}Ni_{32}$	10 (Experiment) + 311(CALPHAD)	Compositions	Primary phase fraction	ANN	[79]
$Al_{19}Co_{16}Cr_{16}Ni_{49}$	4 (Experiment) + 96(CALPHAD)	Compositions	Primary phase fraction	SVM	[80]
$Hf_{0.34}Co_{1.33}Cr_{0.74}Fe_{0.20}Ni_{0.75}$	20 (Experiment)	Content of Co, Cr, Fe, Ni	Content of Hf	ELM	[81]
$Hf_{0.30}Co_{0.80}Cr_{1.40}Fe_{0.82}Ni_{0.16}$					
$Hf_{0.37}Co_{0.42}Cr_{0.81}Fe_{1.29}Ni_{0.82}$					
$Hf_{0.36}Co_{0.16}Cr_{0.76}Fe_{0.81}Ni_{1.38}$					
AlCrFe2.5Ni2.5 (Near-eutectic)	66 (Experiment)	Compositions, phase volume fractions	Melting range	GRNN	[113]

ANN: Artificial neural network; ELM: extreme learning machine; GRNN: generalized regression neural network; SVM: support vector machine.

respect to equilibrium phases, which might differ from the actual EHEA compositions that are metastable because of the fast cooling. In addition, one needs to be cautious while resorting to Scheil simulations for metastable phases, which are usually considered to represent as-cast phases<sup>[63,110,114,115]</sup> because it is performed by only considering atom diffusion in liquids (i.e. completely ignoring solid-state diffusion)<sup>[111]</sup>.

To improve the data fidelity and also the performance of ML modeling, the data may need to be screened or pre-processed. Generally, data pre-processing includes (1) deletion of repetitive and incompatible data; (2) data normalization; and (3) data undersampling or oversampling<sup>[116-118]</sup>. However, randomly oversampling may result in model overfitting while randomly undersampling may cause loss of useful data, both of which



**Figure 3.** A typical categorization of data descriptors in the high entropy alloy design. Reproduced with permission from Roy et al.<sup>[124]</sup>. Copyright 2021, Elsevier.

could jeopardize the validity of the ML predictions<sup>[119]</sup>. Also, we note that metastability or the thermal history is another issue that may affect data fidelity. In such a case, one may obtain different microstructures and properties from the same alloy composition, such as AlCoCrFeNi<sub>3</sub> EHEA<sup>[120,121]</sup>.

### Data features & labels

After data collection, one needs to develop proper data features (or descriptors) and labels for the subsequent training of the ML models. Table 5 lists the commonly used features for the design of HEAs. Ideally, data features should be uncorrelated while containing all relevant information. In data-driven design of HEAs<sup>[122,123]</sup>, alloy composition is usually the first data feature to be included. However, it is believed that only alloy composition alone is not sufficient. Therefore, other complementary data features, which are of physical relevance and significance, should be considered<sup>[112]</sup>. To date, nearly a hundred data features have been employed in the training and optimization of the ML models, which include the so-called atomic parameters<sup>[124-126]</sup>, the environmental parameters<sup>[123]</sup>, and the thermodynamic parameters which can all be derived from alloy composition<sup>[125-128]</sup>, as represented in Figure 3 and Table 5.

The formulation of the complementary data features requires domain knowledge in material science and physical metallurgy<sup>[124]</sup>. To date, data features for eutectic alloys can be divided into two groups: (1) those related to eutectics formation and growth; and (2) those correlated with mechanical properties<sup>[1]</sup>. However, unlike the Hume-Rothery rules for solid solution HEAs<sup>[125,126,129]</sup>, there still lacks a well-established general theory that can underpin the correlation between alloy compositions and eutectics, if there is any. Therefore, most ML models for EHEAs reported in the literature are solely based on the data feature of alloy composition [Table 4], which may compromise their performance. In practice, one can find the most important features using different approaches, such as Pearson Correlation Coefficient (PCC)<sup>[130,131]</sup> and

**Table 5. List of the commonly used features of HEAs with the corresponding formula**

	Data feature	Formula
Compositional feature	Molar fraction of components	$c_i$
Atomic features	Mean atomic radius	$a = \sum_{i=1}^n c_i r_i$
	Atomic size difference	$\delta = \sqrt{\sum_{i=1}^n c_i \left(1 - \frac{r_i}{a}\right)^2}$
	Valence electron concentration	$VEC = \sum_{i=1}^n c_i VEC_i$
	Electronegativity	$\chi = \sum_{i=1}^n c_i \chi_i$
Thermodynamic features	Mixing enthalpy	$\Delta H_{mix} = 4 \sum_{i \neq j} c_i c_j H_{ij}$
	Ideal mixing entropy	$S_{id} = -k_B \sum_{i=1}^n c_i \ln c_i$
Physical features	Melting temperature	$T_m = \sum_{i=1}^n c_i T_{mi}$
	Elastic modulus	$E = \sum_{i=1}^n c_i E_i$
	Bulk modulus	$K = \sum_{i=1}^n c_i K_i$

Shapley Additive Explanation (SHAP) value<sup>[132]</sup>. We believe that data miners need to develop physics-informed data features, which can be derived from the fundamental theories for eutectic formation, such as the Jackson-Hunt theory<sup>[133]</sup>, to improve the predictability of the machine learning models. At present, these are still the ongoing effort of active research for EHEAs. By comparison, the design of data labels for EHEAs is relatively more straightforward, which is either the characteristics of a eutectic-related microstructure (i.e., the volume fraction of eutectic phases<sup>[79,80]</sup>) or the targeted properties for regression ML modeling. For instance, Qiao *et al.*<sup>[113]</sup> used the difference between the solidus and liquidus temperature (i.e. the so-called melting range termed in Ref.<sup>[113]</sup>) as the data label, and the composition and phase fraction as the data feature in the search of EHEAs, which led to the discovery of a near-eutectic composition of AlCrFe<sub>2.5</sub>Ni<sub>2.5</sub>.

### Machine learning model

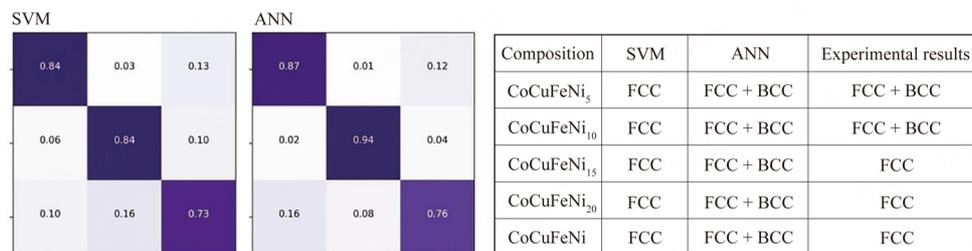
After data are collected with their descriptors/labels being developed, the next task for the data-driven based alloy design is to select a proper ML algorithm. To date, there are a number of ML algorithms that have been used for the design of HEAs, such as support vector machine (SVM)<sup>[116,125,126,128,134]</sup>, artificial neural network (ANN)<sup>[125,126,130,135]</sup>, random forest (RF)<sup>[126,136,137]</sup>, decision tree (DT)<sup>[138,139]</sup> and k-nearest neighbors (KNN)<sup>[130,140]</sup>. The selection of the ML algorithms can be either heuristic<sup>[116,141,142]</sup> or through benchmarking<sup>[125,135]</sup>.

Once the ML algorithm is selected, the ML model will be trained and the reliability of the training results is usually evaluated against the issues, such as overfitting and underfitting, through cross-validation (CV)<sup>[143,144]</sup> and bootstrapping<sup>[122,131]</sup>. To be more specific, the testing accuracy<sup>[116,126]</sup>, the Kappa index<sup>[128]</sup>, the confusion matrix<sup>[118,145]</sup>, and/or the receiver operating characteristic (ROC) curves<sup>[144]</sup> are usually used as the metric for the evaluation of classifiers, while the coefficient of determination (R<sup>2</sup>)<sup>[146]</sup> and the root mean square error

**Table 6. List of the ML models based on SVM or ANN with good performance on small-sized databases**

Target	Size of database	Algorithm	Performance	Ref.
Phase prediction	118	ANN	Accuracy = 0.992	[127]
Phase prediction	401	ANN	Accuracy = 0.943	[130]
Phase prediction	550	SVM	Accuracy = 0.887	[134]
Phase prediction	322	SVM	Accuracy = 0.9384	[139]
Phase prediction	391	ANN	Accuracy = 0.92	[142]
Phase prediction	407	SVM	Accuracy = 0.9743	[149]
Phase prediction	209	ANN	Accuracy = 0.9297	[150]
Hardness prediction	155	SVM	RMSE = 31	[122]
Hardness prediction	214	SVM	R <sup>2</sup> = 0.873	[146]
Hardness prediction	370	SVM	R <sup>2</sup> = 0.8836	[147]
Hardness prediction	53	ANN	R <sup>2</sup> = 0.8575	[151]
Strength prediction	231	ANN	R <sup>2</sup> = 0.9702	[152]
EHEA Design	321	ANN	R <sup>2</sup> = 0.9663	[79]
EHEA Design	100	SVM	R <sup>2</sup> = 0.916	[80]

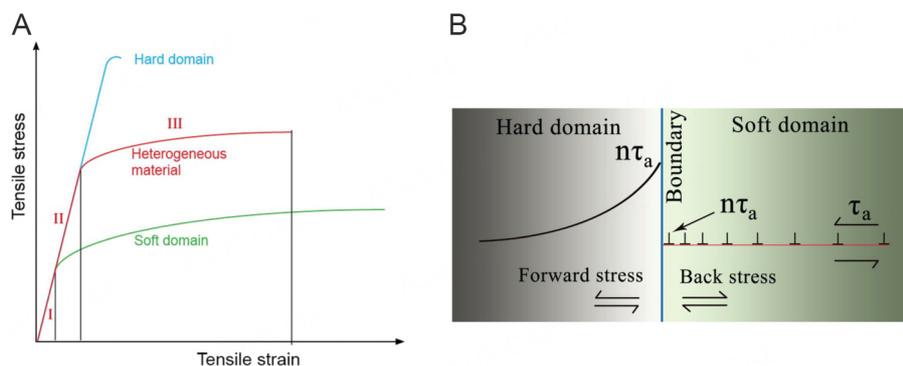
ANN: Artificial neural network; EHEA: Eutectic high entropy alloys; RMSE: root mean square error; SVM: support vector machine.



**Figure 4.** Confusion matrix of the SVM, ANN models, and the predicted results. Reproduced with permission from Jaiswal *et al.* [126]. Copyright 2021, Elsevier. ANN: artificial neural network; SVM: support vector machine.

(RMSE)<sup>[147]</sup> are usually used for regressors. It is noteworthy that the performance of the ML models should be judged not only by existing data (i.e. data in the database) but also by the “unseen” data (data out of the database). In addition to the above-mentioned numerical evaluations, experimental validation is therefore needed that produces unseen data to test the predictability of the ML models. For instance, Jaiswal *et al.* [126] used two different ML models (i.e., SVM and ANN) for the phase prediction of the CoCuFeNi<sub>x</sub> system. While both ML models achieve a similar numerical accuracy (~0.85), it appears that the ANN model can predict the result being consistent only with the experimental observation for low Ni content ( $x = 5, 10$ ). In contrast, the SVM model can predict the results correctly only for high Ni content ( $x = 15, 20, 25$ ), as illustrated in Figure 4.

Among the above-mentioned ML algorithms, the SVM and ANN are the ones that are widely used in the design of HEAs, including EHEAs, due to their good performance on small-sized databases<sup>[148]</sup>, as shown in Table 6<sup>[79,80,122,127,130,134,139,142,146,147,149–152]</sup>. Here, we note that the reported ML models for EHEAs with good performance are mostly regressors, outperforming the classifier. This phenomenon could be attributed to the data imbalance in the EHEA database (i.e. the number of EHEAs is significantly smaller than that of non-eutectic HEAs)<sup>[153]</sup>. In practice, data shortage and/or imbalance could be an issue, particularly for the design of EHEAs. To mitigate the negative effect, people proposed a few methods, including (1) data rebalancing with the Synthetic Minority Oversampling Technique (SMOTE)<sup>[154]</sup> and the Tomek links for



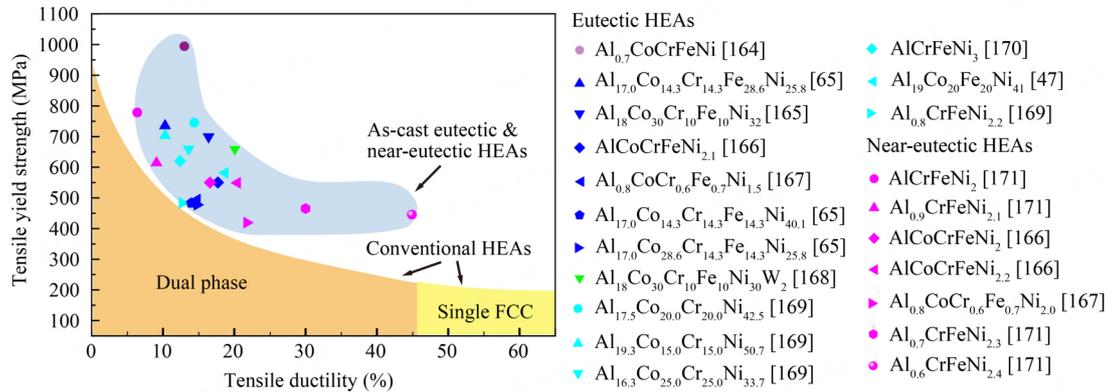
**Figure 5.** (A) Three-stage deformation of heterogeneous materials. Reproduced from Ref.<sup>[161]</sup>. CC BY 4.0; (B) schematic illustration of the heterogeneous deformation strengthening mechanism. Reproduced from Ref.<sup>[162]</sup>. CC BY 4.0.

data cleaning<sup>[155]</sup>; (2) data augmentation with binary/ternary eutectics; (3) engineering of data features which may lead to improved predictability of the ML models even from the small dataset, such as Fuzzy C-means clustering function (FCM)<sup>[156]</sup> and Genetic Programming-based feature extraction using Rough Set Theory (GPRST)<sup>[157]</sup>. In the literature, Bhowan *et al.*<sup>[158]</sup> proposed new parameters to mitigate the issue of imbalanced data. It includes (1) the average mean square error (*AMSE*), which uses the average *MSE* for each data class instead of the overall *MSE* for all data, (2) the incremental-reward-assigned accuracy (*Incr*), which can differentiate different models with similar accuracy by assigning a higher weighted factor to the outputs closer to the target value, and (3) the correlation-ratio-based parameter (*Corr*), which uses the separability of outputs for different data classes to evaluate the classifier performance. In our opinion, it is plausible to extend the finding of Bhowan *et al.* to the data-driven design of EHEAs, which, however, remains to be an open issue.

Currently, the design of eutectics is still limited in the dual-phase structure while only a few multi-phase eutectics were found, e.g. triple-phase eutectics<sup>[59]</sup>, which makes the database significantly biased towards dual-phase eutectics and makes it difficult to find multi-phase eutectics using supervised machine learning models. To solve this problem, one method is to enlarge the database by including more multi-phase eutectics, which, however, is time-consuming. The other one is to use generative machine learning models, such as Variational Autoencoder (VAE)<sup>[159]</sup> or Generative Adversarial Network (GAN)<sup>[160]</sup>, to generate multi-phase eutectics even with the data from binary eutectics. To our best knowledge, this has not been explored yet for EHEAs.

## MECHANICAL PROPERTIES OF EUTECTIC HIGH ENTROPY ALLOYS

Similar to conventional eutectic alloys, EHEAs usually show lamellar or rod-like microstructure with alternating soft and hard phases. Such a heterogeneous microstructure (HS) can provide a unique strain hardening capability through the asynchronous plastic deformation of the soft and hard phases during plastic deformation, which is termed the heterogeneous deformation induced (HDI) strengthening mechanism<sup>[161-163]</sup>. To rationalize the HDI effect, it was proposed that geometrically necessary dislocations (GNDs) will be generated during the plastic flow in a heterogeneous microstructure, which pile up along the interface between the hard/soft phase to maintain the overall deformation compatibility, as illustrated in Figure 5. As a result, this produces the back stress in the soft phase and forward stress in the hard phase, leading to the synergy that a more plastic flow is facilitated even at higher flow stress. Consequently, EHEAs usually show a balanced combination of strength and ductility, as shown in Figure 6<sup>[47,65,164-171]</sup>. To characterize the effect of HDI strengthening, various experimental techniques have been utilized, such as electron back scattered diffraction (EBSD)<sup>[172-174]</sup>, transmission electron microscopy (TEM)<sup>[175,176]</sup>, digital



**Figure 6.** The superior mechanical properties of eutectic high entropy alloys overcome the strength-ductility trade-off in conventional high entropy alloys<sup>[47,65,164-171]</sup>.

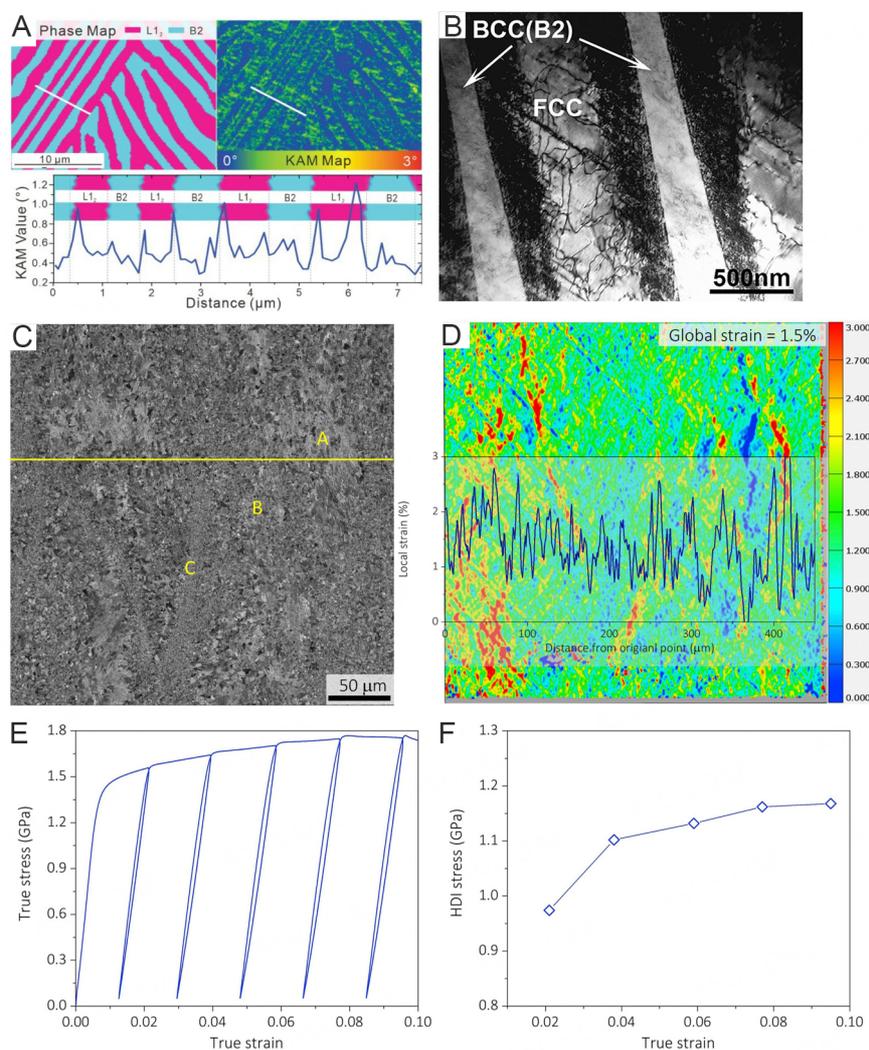
image correlation (DIC)<sup>[177]</sup> and the loading-unloading-reloading (LUR) tests<sup>[177,178]</sup>, as illustrated in **Figure 7**.

In addition to the HDI strengthening, twinning-induced plasticity (TWIP) is another plausible strengthening mechanism in EHEAs, although, in the first place, it applies to materials of low stacking fault energy (SFE) with enhanced strain hardening capability and delayed plastic instability<sup>[179,180]</sup>. Through the so-called dynamic Hall-Petch effect, twin boundaries can act as obstacles to dislocation motion while permitting some partial dislocations to glide<sup>[181]</sup>. Diao *et al.*<sup>[182]</sup> reviewed the deformation twinning (DT) mechanism in HEAs and proposed the preferential conditions for the activation of the DT mechanism, including (a) large deformation strain; (b) low deformation temperature; (c) high strain rate; and (d) large grain size. With the first three conditions being met, a sufficient high dislocation density will be generated, leading to a high local stress for twin nucleation; while the last condition is to ensure there is sufficient space for high twinning activities (i.e. twin thickening)<sup>[183]</sup>. For instance, Shi *et al.*<sup>[180]</sup> uncovered a sequentially activated DT mechanism in the ultrafine-grained  $\text{Al}_{19}\text{Fe}_{20}\text{Co}_{20}\text{Ni}_{21}$  EHEA which resulted in an outstanding combination of yield strength ( $\sim 1.2$  GPa) and tensile ductility ( $\sim 24.0\%$ ).

Besides DT, transformation-induced plasticity (TRIP) is another strengthening mechanism that can be activated in an alloy with an even lower SFE<sup>[184]</sup>. For conventional HEAs, the deformation induced phase transformation can be activated by heuristically adjusting the alloy chemical composition to lower SFE<sup>[184,185]</sup>. However, it is difficult to activate or control the TRIP effect in EHEAs through compositional tuning because of the narrow compositional range for a eutectic alloy. Liu *et al.*<sup>[186]</sup> reported the phase transformation from the BCC/B2 phase to the FCC phase in the near-eutectic  $\text{AlCoFe}_2\text{Ni}_2$  alloy and attributed this phenomenon to enhanced atom diffusion under high-temperature torsion. Wu *et al.*<sup>[187]</sup> found a phase transformation from B2 to BCT structure in the  $\text{Al}_{18}\text{Co}_{30}\text{Cr}_{10}\text{Fe}_{10}\text{Ni}_{30}\text{W}_2$  EHEA during tensile deformation, which was thought to be responsible for the deformability of the B2 phase. These results are interesting and warrant further research. In particular, it still remains open whether EHEAs with the TRIP and/or TWIP effect can be designed through the aforementioned data-driven approach.

## SUMMARY

To sum up, we provide a critical review of the recent development of EHEAs in this article by focusing on the various approach for compositional design, from the empirical to data-driven methods. Through the heuristic empirical methods based on binary eutectics (i.e., combination versus substitution), people have successfully developed a number of EHEAs; however, this trial-and-error approach is ineffective in



**Figure 7.** Characterization of heterogeneous deformation induced strengthening through various experimental techniques. (A) Phase map and kernel average misorientation map of electron back scattered diffraction (EBSD) results in the  $\text{Al}_{19}\text{Co}_{20}\text{Fe}_{20}\text{Ni}_{41}$  EHEA. Reproduced from Ref. [173]. CC BY 4.0; (B) bright-field images of transmission electron microscopy (TEM) in the  $\text{AlCoCrFeNi}_{2.1}$  EHEA. Reproduced with permission from Huang et al. [175]. Copyright 2021, Elsevier; (C) SEM image and (D) strain map of digital image correlation (DIC). A: deformed region; B: large grain; C: small grain; (E and F) loading-unloading-reloading (LUR) curves in the  $\text{Al}_{0.2}\text{CoCrFeNi}_2\text{Ti}_{0.24}$  HEA. Reproduced with permission from He et al. [177]. Copyright 2021, Elsevier.

navigating the multi-dimensional compositional space. Therefore, people turned to the data-driven approach, such as ML modeling, which is supposed to be more effective in locating the eutectic compositions in the complex compositional space. However, the lack of sufficient high-fidelity EHEA data, the imbalanced database, and the poor design of data descriptors can compromise the performance of the ML models, which warrants further research efforts in this field. Finally, we also discuss the various strengthening mechanisms derived from the eutectic microstructure and compositional complexity in EHEAs (i.e., low stacking fault energy). These prior works indicate that the data-based design of EHEAs is promising but still at its infant stage.

## DECLARATIONS

### Author's contributions

Supervised the project: Yang Y

Wrote the manuscript: Chen Z, Yang Y

### Availability of data and materials

Not applicable.

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### Conflicts of interest

Both authors declared that there are no conflicts of interest.

### Ethical approval and consent to participate

Not applicable.

### Consent for publication

Not applicable.

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