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Science and technology in high-entropy alloys

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ABSTRACT As human improve their ability to fabricate materials, alloys have evolved from simple to complex compositions, accordingly improving functions and performances, promoting the advancements of human civilization. In recent years, high-entropy alloys (HEAs) have attracted tremendous attention in various fields. With multiple principal components, they inherently possess unique microstructures and many impressive properties, such as high strength and hardness, excellent corrosion resistance, thermal stability, fatigue, fracture, and irradiation resistance, in terms of which they overwhelm the traditional alloys. All these properties have endowed HEAs with many promising potential applications. An in-depth understanding of the essence of HEAs is important to further developing numerous HEAs with better properties and performance in the future. In this paper, we review the recent development of HEAs, and summarize their preparation methods, composition design, phase formation and microstructures, various properties, and modeling and simulation calculations. In addition, the future trends and prospects of HEAs are put forward.

Keywords: high-entropy alloys, multiple principal components, microstructures and properties, phase formation, modeling and simulation calculations

INTRODUCTION

Metals and alloys have a long history, and they have been playing an irreplaceable role in the progress of human civilization. As shown in Fig. 1, the Bronze appeared as early as in Shang Dynasty, lasting over 1,000 years, and the Iron Age lasted about 3,000 years starting from the Spring and Autumn Warring States period [1]. The application of Al alloys has also been developed for one century [2], even Ti alloys for more than sixty years [3]. Till now, metals and alloys have been closely involved in all aspects of our lives, including agriculture, housing, transportation, food, machinery, and defense industries.

As illustrated in Fig. 1, in the early stage of alloy, one principal element was fused with other elements in trace amount to improve the specific properties. Today, the design concept of many essential alloys is considered as the traditional alloy-design strategy, such as Fe alloys [4,5], Cu alloys [6,7], Al alloys [8], Mg alloys [9,10], Ti alloys [11], and Ni alloys [12]. However, the chemical composition of modern alloys is greatly expanded, for example, Inconel 718 superalloy, which is a representative of Ni alloy, contains a variety of other elements [13] in addition to Ni. One of the three basic principles of bulk amorphous formation gives a description that an alloy contains at least three elements [14,15]. Through careful analysis, it is found that the alloys in Fig. 1 are still deeply influenced by the traditional alloy-design concept. While for further improving the properties, the traditional design paradigm meets with the bottleneck. Fortunately, the complexity of the elemental composition in alloys has been increasing steadily with time as illustrated in Fig. 1.

Crystalline multi-principal element alloys and highentropy alloys (HEAs) were first independently reported in 2004 by Cantor et al. [16] and Yeh et al. [17], which refer to the same concept. Different from the traditional alloy-design concept, HEAs are not based on only one or two elements but contain at least five principal elements in an equal or near-equal atomic percentage (at.%) with no obvious difference between the solute and solvent [17]. According to the existing physical-metallurgy and phase diagrams, such multi-element alloys may produce many phases and intermetallic compounds, resulting in complex and brittle microstructures that are difficult to analyze and engineer, but probably have finite practical values [18]. Beyond expectations, experimental results indicate that the higher mixing entropy in these alloys enhances the formation of random solid-solution phases

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Figure 1 Rising trend of alloy chemical complexity *versus* time (IMs: intermetallics or metallic compounds, HEA: high-entropy alloy). Reproduced with the permission from Ref. [1]. Copyright 2017, Springer.

with simple structures, such as face-centered-cubic (FCC), body-centered-cubic (BCC), or hexagonal-closepacking (HCP) structures, and thus reduces the number of phases [19–29]. Since then, the new alloy-design strategy opens a huge, unexplored field of multi-component alloys. The strategy of alloy design has obtained unimaginable successes and great efforts have been devoted to the development and application of many HEAs [30,31] in various fields due to their excellent performance, such as the unique wear resistance [32], excellent strength and thermal stability at elevated temperatures [33,34], superior high elongation [35,36], great fatigue and fracture resistance [37–39], etc.

Up to now, thousands of reports on HEAs have been published, including the excellent book "High-entropy Alloys: Fundamentals and Applications" [40], and several review papers [19,20,41–45] covering almost every aspects of current research on HEAs. However, our understanding and research on HEAs is just the tip of the iceberg, and more unexplored secrets of HEAs are disclosed continuously. Due to their remarkable properties and promising research prospects, many new findings in HEAs are emerging beyond the existing review papers and books [40,41]. This paper summarizes the recent progress of HEAs, provides a supplement to the research content of HEAs, and forecasts the future development of HEAs.

DEFINITIONS

There are two definitions of HEAs by composition and entropy, which lead to some confusion and controversy that whether the multi-component alloy could be regarded as HEAs [30].

Composition-based definition

The earliest composition-based definition was published in 2004 [17]. HEAs were preferentially defined as alloys containing at least 5 principal elements, each with an atomic percentage (at.%) between 5% and 35%. It is worth mentioning that the atomic percentage of each minor element, if any, is even smaller than 5%. The definition is expressed as follows [18,20,40]:

$$n_{\text{major}} \ge 5, 5 \text{ at } .\% \le c_i \le 35 \text{ at.}\%$$

and $n_{\text{minor}} \ge 0, c_j \le 5 \text{ at.}\%$, (1)

where n_{major} and n_{minor} are the numbers of major and minor elements, respectively. c_i and c_j are the atomic percentages of the major element, *i*, and the minor element, *j*, respectively. From this definition, HEAs need not to be equimolar or near-equimolar, and even contain minor elements to balance various materials properties, such as the ductility, toughness, strength, creep, oxidation, etc. [32,46,47].

Entropy-based definition

Entropy is a thermodynamic state function, and the essence of entropy is "inherent chaos" of the system [20]. According to the Boltzmann's thermodynamic statistics principle, the quantitative relationship between the entropy and randomness of the system is given by:

$$\Delta S_{\rm conf} = k \ln w, \tag{2}$$

where *k* is the Boltzmann's constant, and *w* is the number of distinguishable ways of arranging the atoms in the solution. The total mixing entropy has four contributions: configurational entropy, $\Delta S_{\text{mix}}^{\text{conf}}$, vibrational entropy, $\Delta S_{\text{mix}}^{\text{vib}}$, magnetic dipole entropy, $\Delta S_{\text{mix}}^{\text{mag}}$, and electronic randomness entropy, $\Delta S_{\text{mix}}^{\text{elec}}$, and the relationship among them is given by:

$$\Delta S_{\rm mix} = \Delta S_{\rm mix}^{\rm conf} + \Delta S_{\rm mix}^{\rm vib} + \Delta S_{\rm mix}^{\rm elec} + \Delta S_{\rm mix}^{\rm mag}.$$
 (3)

The configurational entropy is dominant over the other three contributions. Hence, the configurational entropy often represents the mixing entropy in order to avoid complex calculations to determine the other three contributions [40]. For an ideal random *n*-component solid solution, its ideal configurational entropy per mole is approximately [19]:

$$\Delta S_{\text{conf}} = -R[c_1 \ln c_1 + \dots + c_n \ln c_n]$$

= $-R \sum_{i=1}^n c_i \ln c_i,$ (4)

where *R* is the gas constant, and c_i is the mole fraction of the *i*th element, and *n* is the number of the components. According to the extreme theorem, when $c_1 = c_2 = \ldots = c_n$, the entropy of the system reaches its maximum value.

Considering an equi-atomic alloy in its liquid state or regular solid-solution state, its configurational entropy per mole could be calculated as [30]:

$$\Delta S_{\rm conf} = R \ln n. \tag{5}$$

It defines that HEAs have a configurational entropy in a random state larger than 1.5R, no matter they are single phase or multiphases at room temperature. This definition could be expressed as [48]:

$$\Delta S_{\rm conf} > 1.5R.$$
 (6)

Developing definitions

Although each definition of HEAs contains a wide range

of alloys, both definitions overlap for the most part. Despite these definitions, the compositions in non-overlapping regions are also regarded as HEAs. For example, the maximum configurational entropy of a 5-component HEA is 1.61R (ΔS_{conf} =Rln5=1.61R) for the equimolar alloy, and the minimum value of the configurational entropy is 1.36R for an alloy with 35% A, 35% B, 20% C, 5% D, and 5% E (atomic percentage, $\Delta S_{conf} = -R(2 \times 0.35 \times 10^{-6})$ ln0.35+0.2×ln0.2+2×0.05×ln0.05)=1.36*R*). The latter alloy is also considered as an HEA by the composition-based definition but cannot fit into the entropy-based definition. Another example is an equimolar alloy containing 25 elements, with a concentration of 4 at.% for each element. This alloy is still considered as an HEA since the value of the configurational entropy is 3.219R ($\Delta S_{conf} = -R$ $(25\times0.04\times\ln0.04)=3.219R$). An alloy with a certain composition fitting only one of the two definitions is regarded as an HEA. In addition, sometimes the quaternary equimolar alloy also considered to be an HEA in literature because its composition and configurational entropy are close to the lower limits of both definitions. Thus, the definitions of HEAs are simply approximate guidelines, not strictly laws [19,40].

From the two definitions and the description of nonoverlapping regions of HEAs, it implies that the basic principle behind HEAs with multiple principal elements is to achieve the high mixing entropy in order to enhance the formation of solid-solution phases. As 1.5R is a lower limit for HEAs, we further define medium-entropy alloys (MEAs) and low-entropy alloys (LEAs) to differentiate the power of the mixing entropy effect for all alloys in the nature. Herein, 1R is the boundary between MEAs and LEAs since the mixing entropy less than 1R is expected to be noncompetitive with a larger mixing enthalpy [40].

Thus, the alloys are divided into the following three categories and illustrated in Fig. 2 [48]:

LEAs: $\Delta S_{conf} < 1R$, including traditional alloys based on one or two elements;

MEAs: $1R \le \Delta S_{conf} \le 1.5R$, including alloys based on two to four elements;

HEAs: $\Delta S_{conf} > 1.5R$, including alloys based on five elements at least or some quaternary equimolar alloys.

It should be noted that among various thermodynamic factors, such as the mixing enthalpy, mixing entropy, atomic-size difference, valence-electron concentration, and electronegativity, the mixing entropy is the sole factor, which rises with increasing the number of principal elements. The name "high-entropy alloys" is pertinent for MPEAs (multiple principal elements alloys) [32,49–51]. Various names have appeared with the development of



Figure 2 Alloys world based on the configurational entropy.

HEAs, such as complex concentrated alloys (CCAs) [19,52], complex multicomponent alloys (CMAs) [46,53], compositionally complex alloys (CCAs) [40], baseless alloys (BAs) [19], metal buffets (MBs) [45], etc.

PREPARATION OF HEAs

The discovery of HEAs is related to the study of bulk amorphous alloys [20]. Therefore, the preparation method of HEAs has inherited from the preparation technique of amorphous alloys, which can be divided into three major routes [40], as shown in Fig. 3. The main route is liquid mixing, including arc melting, electricresistance melting, inductive melting, laser melting, laser cladding, and laser engineered net shaping (LENS) [52]. The second route is solid mixing, which mainly contains the mechanical alloying and subsequent consolidation process. Another route is gas mixing, including sputter deposition, pulse-laser deposition (PLD) [20], atomiclayer deposition (ALD) [40], molecular-beam epitaxy (MBE) [54], and vapor-phase deposition.

The aforementioned methods could fabricate three-dimensional bulk, two-dimensional film-like, and one-dimensional fibrous HEAs, and the three-dimensional sample block-body-study is the most widely used [36,39,55,56]. Among the reported preparation methods, arc melting is the dominant fabrication route to produce bulk HEAs [20,53,57,58]. More recently, gradient bulk samples of HEAs were produced by elemental power mixes with LENS apparatus [52]. Due to the inherent compositional complexity and the huge difference in the melting points between the constituent elements, the preparation of HEAs is challenging. Significant elemental segregation occurs during melt solidification and cooling



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Figure 3 Fabrication routes of HEAs.



Figure 4 Schematic of multi-target co-deposition for HEAs.

[59]. Thus, compared to conventional alloys, the as-cast samples of HEAs may have obvious casting defects, such as cracks, pores, and residual stresses along with compositional gradients and abnormal grain-size distributions. In order to obtain organized and well-behaved HEAs, subsequent processing is essential [60–63].

As the development of HEAs, the elements with FCC or BCC crystalline structures were generally considered as the matrix elements. Thereafter, appropriate modification elements were added to improve the required performances. The "trial and error" traditional alloying method leads to the consumption of a large number of human and material resources, research-cycle extension, low efficiency, etc. These shortcomings are more conspicuous for multi-element HEAs [64]. The development of highthroughput experiments can well solve these problems [65]. High-throughput experiments have been developed in the preparation of thin films and bulk alloys, especially for the fabrication of thin films. In addition, the multitarget co-deposition method is suitable for the preparation of HEA films with various constituent elements [66]. As shown in Fig. 4, co-deposition uses a different distance between the substrate and the target, and different targets provide a certain concentration gradient on the substrate during deposition to produce a HEA film with a continuous concentration gradient [66]. Subsequently, combining with high-throughput characterization techniques, we can achieve rapid screening of HEAs, and then the bulk-like selected components are prepared. According to the composition design of HEAs, many single-element or alloy targets can be prepared, with the atomic percentages of elements controlled by adjusting the target sputtering power. The HEA film with a continuous composition gradient is obtained and then the bulk-like selected alloy can be manufactured. Nowadays, the high-throughput highway to the computational HEAs design is a great opportunity and challenge for the development of HEAs [67].

PHASE FORMATION

Although there are more than four or five elements in HEAs [20], they tend to form a relatively-simple phase after solidification, such as FCC [21], BCC, or HCP [23,68] structures. However, with the extensive and deep research on HEAs, it is found that the alloys also contain ordered intermetallics, amorphous and nanocrystalline precipitates [19,47,69].

Simple solid-solution structure in an HEA

HEAs are easy to form phase structures with simple FCC, BCC and HCP solid-solution structures [9,20,23]. One famous HEA is the Cantor alloy [20], containing Fe (BCC), Co (HCP), Cr (BCC), Mn (BCC), and Ni (FCC) in an equimolar ratio, with only an FCC solid-solution phase when solidified dendritically in the as-cast sample. Then the 5-component alloy was expanded to a 6component alloy by adding Cu (FCC), Nb (FCC), or V (BCC), showing the simple FCC solid-solution structure in the as-cast alloy with different lattice parameters. When the HCP-type Ti was added, a BCC structure formed within the FCC solid-solution phase [16]. Another typical example is the Al, CoCrFeNi (molar percentage, $0 \le x \le 2$) system prepared by arc melting [70]. The as-cast CoCrFeNi alloy has a pure FCC solid-solution phase. As increasing the Al molar percentage from 0 to 2, the Al_xCoCrFeNi system changes the crystal structure from FCC to FCC+BCC phases, and finally to a single BCC phase [55]. A typical alloy TaNbHfZr has a solely BCC structure [71].

Although the HCP phase is often referred to as a typical simple solid-solution structure of HEAs, there is few discovery for this phase. Tsau [72] found the HCP solid-solution phase in the alloy of TiCrZrNb (in atomic proportion), but the HCP phase did not exist independently, and it was in the interdendritic region, and the matrix

 Table 1
 Mixing enthalpy data (kJ mol⁻¹) between the elements of Fe, Ni, Cr, Cu, and Zr

	Fe	Ni	Cr	Cu	Zr
Fe	-	-2	-1	13	-25
Ni	-	-	-7	4	-49
Cr	-	-	-	12	-12
Cu	-	-	-	-	-23
Zr	-	-	-	-	-

was BCC phase. In addition, Qiao *et al.* [23] found that the as-cast alloy GdHoLaTbY (in atomic proportion) formed an HCP phase.

Mesophase and other complex phases in HEAs

The high entropy of HEAs hinders the formation of intermetallic compounds and the occurrence of phase separation, therefore the formation of solid-solution phases is promoted [52]. However, there are many factors that affect the formation of HEAs, including the mixed enthalpy, atomic-size difference, valence-electron concentration, and so on [50]. The mesophase or complex multi-phase coexistence forms in HEAs due to the existences of some chemically-compatible elements. Li et al. [73] studied the alloy of FeNiCrCuZr. They found that apart from the BCC phase, intermetallic compounds also precipitated in the alloy; and the formation of intermetallic compounds was due to the strong compoundformation tendency of Zr with other metals, consistent with measurements that the mixing enthalpies between Zr and other metals are too negative (Table 1). Thus, the mixing enthalpy became another dominant factor to facilitate the formation of intermetallic compounds in Fe-NiCrCuZr. In some systems, the high mixing entropy cannot completely overcome the contribution of the high enthalpy of mixing to the free energy.

Nanocrystalline and amorphous phases in HEAs

Conventional alloys or bulk amorphous alloys can only precipitate nanocrystals under special heat-treatment conditions. The lattice-distortion effect and the slow-diffusion effect in HEAs seriously hinder the nucleation and growth of the grains. Hence, some HEAs precipitate nanometer phase and even amorphous phase during the as-cast or complete-tempering condition [20,40,47]. As illustrated in Fig. 5, a series of heat-treatment conditions were performed on $Al_{0.5}$ CoCrFeNi alloy [74], and the bright-field image of the acicular precipitate and the corresponding diffraction patterns are present, indicating that such precipitates have the same BCC (B2) phase as



Figure 5 The bright-field image and corresponding diffraction pattern of the acicular precipitate in the $Al_{0.5}$ CoCrFeNi alloy (the precipitate possesses an ordered BCC crystal structure). Reproduced with the permission from Ref. [74]. Copyright 2014, Elsevier.

the interdendritic structure. Zhao *et al.* [75] developed an amorphous alloy, which can be uniformly deformed without generating shear at room temperature. Mean-while, its critical size is greater than 3 mm, and the amorphous transition temperature of the alloy is close to room temperature, with low density, high specific strength, and low elastic modulus.

MICROSTRUCTURES

In the case of multicomponent alloys, there are more chances of segregation of elements due to the difference between the constituent elements in melting points, densities, and other physical properties. Depending on the growth conditions, grains may grow into planar, cellular, or dendritic morphologies [76]. For HEAs, arc melting limits the fabrication of HEA ingots, compared to induction melting. The melting furnace with non-consumable tungsten electrode is only useful for making ingots with a limited size and shape (button shaped or slender-rod shaped as shown in Fig. 6). After the arc melting or induction melting, the alloy exhibits as-cast dendrites. The dendrites and inter-dendrites have significant component segregations [40]. Solidification/ cooling rates have a significant influence on the microstructural evolution of as-cast HEAs [77]. The AlCoCr-CuFeNi HEA fabricated by splat quenching with rapid solidification rate $(10^6 - 10^7 \text{ K s}^{-1})$ possessed a single BCC phase with fine polycrystalline structures, whereas the ascast alloy had multi-phases with serious inter-dendritic segregation [78].



Figure 6 Schematic of arc melting.

The microstructure of the alloy can also be changed by controlling the growth direction and the growth rate of crystal during the solidification process of the alloy. Zhang *et al.* [79] successfully prepared Al_{0.3}CrFeNiCo columnar crystals by Bridgman primary and secondary directional solidification. The effects of directional solidification on the microstructure, crystal orientation, and mechanical properties of the Al_{0.3}CrFeNiCu₂ alloy were studied. In addition, eutectic and peritectic structures can be observed in HEAs of different systems. For example, Al_xCrCeFeNi₂ HEAs exhibit eutectic clusters similar to sunflower shapes [80], while AlCoCrFeNb_{0.5}Ni [81] and CoFeNi₂V_{0.5}Nb_{0.75} [58] alloys exhibit lamellar eutectic structures.

PROPERTIES

Hardness

HEA is widely studied not only because it forms a unique multi-component solid-solution phase but also because it possesses high hardness and strength. Fig. 7 shows the hardness of a variety of alloys in the as-cast and fully-annealed states. Compared with other traditional alloys, such as the 316 stainless steel, Hastelloy, HEAs have higher hardness and better anti-annealing softening performance [17].

Compressive property

The mechanical properties of HEAs are evaluated by compressive loading on the cylindrical samples [82,83], which could be prepared easily by arc melting. The HEA systems generally possess an FCC phase and have great



Figure 7 Comparison of hardness between HEAs and conventional alloys before and after annealing. The data are from Ref. [17].

combination of strength and ductility in compression, when compared to those with a BCC phase, in Fig. 8a. However, the yield strength of HEAs with a BCC phase is very high and comparable to bulk metallic glasses [46]. With increasing number of principal elements, the strength of the HEAs system increases due to solidsolution hardening, with reduced ductility [20]. Miracle and Senkov [19] reviewed the compression yield stresses between the refractory HEAs and other elevated-temperature alloys. The compression yield stress reduces with increasing temperature, and the inflection point is around 1,000 K for almost all alloys. However, NbMoTaW and NbMoTaWV exhibit outstanding behaviour and remain useful strengths up to 2,000 K (Fig. 8b).

For the Al_xCoCrFeNi and Al_xCoCrCuFeNi systems [17,70], increasing the amount of Al results in the formation of a BCC phase in the FCC matrix, which yields an increase in the compressive strength at the expense of ductility. The HEAs with an FCC phase, such as Al_{0.1}CoCrFeNi [84], CoCrFeNi, and CoCrFeNiMn [85], exhibits excellent plasticity and work-hardening behaviour with low yield strength under compressive loading. The Al_{0.75}CoCrFeNi alloy with FCC + BCC dual phases exhibits an excellent yield and ultimate strengths of 1,938 MPa and 2,221 MPa, respectively, but a limited ductility of 7.6% [86].

Tensile property

Few studies were focused on the tensile properties of HEAs. The crystal structure of the HEAs has a large in-fluence on the tensile properties [36].

He *et al.* [87] conducted a systematic study on the influence of the progressive addition of Al on the mechanical behaviour of the FCC-phase CoCrFeNiMn HEA, illustrated in Fig. 9. The FCC alloy of Al_x CoCrFeNiMn system exhibits excellent plasticity and low strength, whereas the dual phase alloys exhibit a better combination of strength and ductility. The AlCoCrFeNi_{2.1} alloy with dual phases also presents promising tensile properties with an elongation and fracture strength of 23% and 1,200 MPa, respectively [88].

In the range of low-temperature to room-temperature, the yield strength and the deformation of the alloy de-



Figure 8 (a) Compressive behaviour of various HEAs; (b) compressive yield strength *versus* temperature. Reproduced with the permission from Ref. [19]. Copyright 2017, Elsevier.



Figure 9 Tensile strength, yield strength, and elongation of the as-cast Al_xCoCrFeNiMn alloys. Reproduced with the permission from Ref. [87]. Copyright 2014, Elsevier.

crease as the temperature drops. However, Li et al. [36] found that the Al_{0.3}CoCrFeNi alloy's tensile strength and elongation in the liquid nitrogen temperature reached the maximum. The curves of the quasi-static tensile engineering of the alloy in the range of 77-298 K are shown in Fig. 10. The Al_{0.3}CoCrFeNi HEA at low temperature has good plastic-deformation behavior for the formation of deformation twins, which is similar to the alloy of CoCrFeMnNi studied by Otto et al. [62]. The yield strength and shaping deformation were measured in the range of 77-1,073 K. At the liquid-nitrogen temperature, the yield strength reaches the maximum, indicating that the alloy with a fine grain structure has very good mechanical properties over the entire testing temperatures, similar to conventional alloys, and exhibits serrated flows at intermediate temperatures. Diao et al. [45] summarized the yield strength of HEAs and conventional alloys from room temperature to 1,900 K. The excellent mechanical properties of HEAs over a wide range of temperatures under tensile loading is evident, promising them ideal candidates for structural applications, compared to conventional superalloys and stainless steels [45,62,89,90].

Corrosion resistance

Under high concentrations of sulfuric acid, hydrochloric acid, nitric acid and other corrosive solution condition, HEAs show excellent corrosion resistance, especially the one with Cu, Ti, Cr, Ni, or Co [43,91,92]. Some HEAs present outstanding corrosion resistance even better than the traditional stainless steel. The CoCrFeNiCu_x HEA system shows that the CoCrFeNi alloy has good pitting



Figure 10 Tensile strength (a) and elongation (b) as a function of fiber diameter, respectively. Reproduced with the permission from Ref. [36]. Copyright 2017, Elsevier.

corrosion resistance, similar to 304 stainless steel [43,91– 93]. The polarization curves of the $Al_{0.5}$ CoCrCuFeNi alloy and 304 stainless steel in 1 mol L⁻¹ H₂SO₄ solution showed that the corrosion potential of HEAs is higher, with better corrosion resistance [43,91–94].

The relationship between pitting potential (E_p) and corrosion current densities (I_{corr}) of HEAs (Al_x CoCrFeNi in which x = 0.3, 0.5, and 0.7) and conventional alloys (such as stainless steels, Al-, Ti-, Cu-, and Ni-based alloys) [91] are displayed in Fig. 11. The data were measured in the environment of 3.5 wt% NaCl solution at room temperature. It suggests that HEAs have higher E_p and lower I_{corr} than Al-, Cu-, and some of Ti-based alloys. Therefore, the HEAs show excellent localized and general corrosion resistance.

Thermal stability

Refractory HEAs [27] are a category of emerging multi-

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Figure 11 Comparison of E_p and I_{corr} between HEAs of Al_xCoCrFeNi (x = 0.3, 0.5, and 0.7) and other conventional alloys in 3.5 wt% NaCl solution at room temperature. Reproduced with the permission from Ref. [91]. Copyright 2017, Elsevier.

component alloys, showing superior mechanical properties at elevated temperatures which is important for application [33,95]. The NbMoTaW alloy has better performance retention than the refractory metal tungsten after a heat exposure at 1,373 K for 3 days [33]. Meanwhile, the HEAs with these excellent properties drive a new class of materials in nanoscale devices potentially in high-stress and elevated-temperature applications. Sathiyamoorthi et al. [96] found that ultrafine-grained CoCrFeNi HEAs show the exceptional thermal stability upon the exposure of sintered compacts to high temperature (973 to 1,173 K) and prolonged duration of 600 h, as illustrated in Fig. 12. Samples exposed to 973 K for 600 h show negligible change in hardness. Meanwhile, the fractional decrease in hardness after 600 h at 1,073 K and 1,173 K is 9.5%, and 25%, respectively.

Irradiation property

In general, the interaction between energetic ions and atoms in the material causes lattice damage, resulting in a variety of defects, including point defects, dislocation loops and holes [97], etc. The research shows that the HEA still has high phase stability, even with the Au ion-irradiation dose exceeding 50 dpa (displacement per atom). At the same irradiation dose (50–70 dpa), compared to other commonly-used irradiation-resistant materials, such as M316 stainless steel and pure Zr, an HEA has a relatively-low volume swelling rate [98]. Nagase *et al.* [99] studied the *in situ* electron irradiation of ZrHfNb with an amorphous structure, and Jin *et al.* [100] studied



Figure 12 Diagram of composite microstructures observed in the assintered condition. Reproduced with the permission from Ref. [96]. Copyright 2017, Elsevier.

the CoCrFeNiMn with an FCC structure. It was found that the CoCrFeNiMn HEA did not change significantly after 773 K high-temperature and 3 MeV Ni ions irradiation, and the grains did not coarsen. High performance of HEA radiation-resistant materials provides a new idea for nuclear materials, and it has been used as the catalyst in the nuclear energy [97,101–103]. Waseem *et al.* [68] considered that the W_x TaTiVCr as low/reduced-activation alloys have a promising future for fusion power plants.

Thermoelectric property

Thermoelectric materials capable of the direct conversion between heat and electricity have attracted tremendous attention in the past several decades for the engine-waste heat recovery to improve the fuel efficiency. HEAs have a high degree of chaos in its atomic arrangement, resulting in the enhanced scattering of phonon and effectively reducing its lattice thermal conductivity. The PbSnTeSe [104] HEA was discovered, possessing a quite low lattice thermal conductivity of 0.6 W m⁻¹ K⁻¹ at room temperature. By minor additions of La to substitute Pb, the thermoelectric performance of PbSnTeSe could be further enhanced, as indicated in Fig. 13a. The series of Pb_{1-x}SnTeSeLa_x alloys containing various miscellaneous contents of La, x = 0, 0.02, 0.04, 0.06, 0.08, and 0.10, aredesignated hence as base, 0.5La, 1.0La, 1.5La, 2.0La, and 2.5La, respectively. The Pb_{1-x}SnTeSeLa_x alloys show a lattice thermal conductivity below 1 W m⁻¹ K⁻¹ at room temperature, which is far lower than that of binary compounds with the same crystal structure, for example, SnTe, PbTe, and PbSe, due to the strong phonon scattering resulted from the severe lattice-distortion of the



Figure 13 (a) XRD patterns of Pb_{1-x}SnTeSeLa_x HEAs; (b) the combined lattice and bipolar thermal conductivity for all the samples. The lattice thermal conductivity of PbTe, PbSe, and SnTe at room temperature is also shown in (b). The inset in (b) presents $\kappa - \kappa_e$ as a function of 1,000/*T*. Reproduced with the permission from Ref. [104]. Copyright 2017, Taylor & Francis Group.

PbSnTeSe HEA. Unfortunately, the lattice thermal conductivity of the base alloy starts to rise when the testing temperature exceeds 400 K due to the strong bipolar effect. For all the La-doped alloys, the bipolar effect is suppressed to a certain degree but still exists. The inset of Fig. 13b analyzes the lattice thermal conductivity without bipolar contribution, $\kappa - \kappa_e vs. 1,000/T$ (κ : thermal conductivity, κ_e : electronic thermal conductivity). It is obvious that the lattice thermal conductivity at 873 K would be lower than 0.5 W m⁻¹ K⁻¹ for the base alloy, if the bipolar effect was entirely eliminated.

HEAs also have other excellent properties, such as excellent wear resistance [105], fracture toughness [36], as well as very high resistivity [56,106]. At the same time, the HEA has good soft magnetic properties [20,107]. For the FeCoNi(AlSi)_{0.2} HEA [106] prepared by arc-melting method, the saturated magnetic strength, coercivity, and electrical resistivity at room temperature reach 1.15 T, 1,400 A m⁻¹ and 69.5 m Ω cm⁻¹, which are promising in the high-frequency communication.

HEAS AND "MATERIALS GENOME INITIATIVE"

The purpose of the "Materials Genome Initiative" is to increase the speed of discovery, development, production, and application of new materials by means of calculations, database and experimentation, which changes the experiment-oriented "trial and error" research and design model [108]. Therefore, HEAs as a typical new material can meet its opportunity to develop rapidly in the subsequent days. The project emphasizes a change in the research and design culture of materials through the integration of the computational and experimental data, and the integration of high-throughput computations and multi-scale simulations to accelerate material-development methods and approaches [109,110]. The material simulation is widely used to predict material properties. It can be used to simulate the material from different scales. and qualitatively and/or quantitatively describe the characteristics of materials, and promote us to understand the material from multiple angles. Fig. 14 displays the relationship between the time and space. Computational materials science involves all aspects of the material, such as different levels of structures, various properties, etc. Thus, there are many corresponding calculation methods, the first-principles density functional theory (DFT), molecular dynamics (MD), discrete dislocation dynamics (DDD), phase-field method (PFM), thermodynamics model (TM), finite element method (FEM) [111], etc. For materials with different space scales, there are corresponding material calculation methods, including the calculation of phase diagram (CALPHAD) and high-throughput methods [26,65,69,112-117].

There are many formation rules and empirical criteria for HEAs, which may not apply to all HEAs. Zhang *et al.* [29] summarized the factors of the atomic-size difference, δ , and the enthalpy of mixing, ΔH_{mix} , of the multi-component alloys:

$$\delta = \sqrt{\sum_{i=1}^{N} x_i (1 - x_i / \sum_{j=1}^{N} x_j r_j)^2},$$
(7)

$$\Delta H_{\rm mix} = 4 \sum_{i=1, i \neq j}^{N} \Delta H_{ij}^{\rm mix} x_i x_j, \tag{8}$$

where N is the number of the elements in an alloy, x_i or x_j is the atomic percentage of the $i^{\rm th}$ or $j^{\rm th}$ component, r_j is the atomic radius of the $j^{\rm th}$ component, $\Delta H_{\rm mix}$ is the mixing enthalpy for AB alloys. Subsequently, to further understand the relationship between $\Delta H_{\rm mix}$ and $\Delta S_{\rm mix}$, Zhang and Yang [51] proposed a new parameter, Ω , defined by:



Figure 14 A map of time and spatial scales in computational materials.

$$\Omega = \frac{T_{\rm m} \Delta S_{\rm mix}}{|\Delta H_{\rm mix}|},$$
(9)

where $T_{\rm m}$ is the melting temperature of the *N*-component alloy. Zhang *et al.* [40] summarized the published HEAs and suggested a phase-formation rule using the δ and Ω with $\Omega \geq 1.1$ and $\delta \leq 6.6\%$. However, the FCC-type phase-forming δ shows a large overlap with that of the BCC-type phase, which means new rules or parameters need to be considered for the phase formation. Guo *et al.* [118] considered that the phase stability of FCC and BCC solid solutions could be well delineated by the valanceelectron concentration (VEC) defined by:

$$\text{VEC} = \sum_{i=1}^{N} x_i \text{VEC}_i, \tag{10}$$

where x_i and VEC_i is the atomic percentage and VEC of the i^{th} component. The simplified rule to identify the phases of HEAs is: VEC<6.87, BCC; VEC>8, FCC. Although the understanding of the prediction of phase stability from fundamental properties of constituent elements benefits the alloy design greatly, and then could be used to predict the mechanical or physical properties of the alloys, the accuracy of the previous parameters are limited [118]. Thus, the computation modeling is an excellent opportunity and challenge.

The HEA is a new type of materials that exhibits a disordered structure, and the alloy generally contains at least five composition elements, and the atomic ratio of each element is above 5% [17,20]. Such a multi-component alloy is a vast challenge for the computational modeling [50]. However, with the research and interest in the field of HEAs, and the gigantic development of material computation, there are many calculations and pre-

dictions to study the structures, properties, dynamics, and thermodynamics of HEAs [119–123].

DFT computations

Among the available predictive computing techniques, the DFT method is probably the ideal technique for solving multi-composition alloy systems [40,112], such as HEAs. Generally, the first-principle calculation and modeling are the solution of the Schrödinger equation, which only requires structures and atomic numbers as the input to predict physical properties; and no experiments are needed. The following equation is the many-body Schrödinger's equation [124]:

$$\widehat{H}\Psi(\vec{r}) = E\Psi(\vec{r}),\tag{11}$$

where the \widehat{H} is Hamiltonian operator, $\Psi(\overline{r})$ is the statewave function, and E is the energy. However, the Schrödinger equation is very difficult to describe a complex system, and can only handle simple electronic systems, such as hydrogen atoms [125]. Therefore, the researchers use some methods to scientifically and rationally approximate and simplify the Schrödinger equation. The most classical method is the DFT [126]. The solution of the bulk ground state is accurately reduced to the solution of the ground density distribution, which is given by the Schrödinger equation of a single particle. Kohn and Sham [40] considered that the particle density function of a multi-particle system can be obtained by a simple single-particle wave equation, and the Kohn-Sham equation [127] is a self-consistent equation.

The DFT mainly converts the problem of multi-electrons into that of a single electron, and describes the physical properties of the ground state through the ground state electron density. The DFT calculations usually only contain the basic physical constants, such as the speed of light, Planck's constant, electronic charge, etc. as input parameters [128]. Based on the most accurate theory, the predicted results are in good agreement with the experimental results [112]. The first-principles approach referred here deals with the DFT. Although the DFT has many simplifications of the Schrödinger equation, the computational process is still challenging due to the HEAs containing multiple principal components. Thus, hybrid Monte Carlo/molecular dynamics (MC/ MD) simulations, ab initio molecular dynamics (AIMD) simulations, special quasi-random structure (SQS) modeling, coherent potential approximation (CPA), even small sets of ordered structures (SSOS) calculations are used for the DFT calculation of HEAs [24,40,59,116,129-135].

By the DFT calculations, the electron density near the Fermi surface is often used to reveal the stability of materials and some functional properties, especially the band gap, magnetic properties, etc. The element of the transitional (3d) HEAs is usually magnetic [40,44,106,128, 129,136]. The model of the transitional HEAs can be constructed by a supercell or SQS. Normally, only the spin orbit is considered or the magnetic force is not taken into account. In the DFT calculations, we need to consider the spin polarization. Zuo et al. [129] used the SQS to construct the structures of CoFeMnNi, CoFeMnNiCr, and CoFeMnNiAl, with the DFT calculations conducted at 0 K through the VASP (the Vienna ab initio simulation package). The electron density of the 3d HEAs with spin polarization is considered and illustrated in Fig. 15. The DFT calculations on the electronic and magnetic structures reveal that the anti-ferromagnetism of Mn atoms in CoFeMnNi is suppressed especially in the CoFeMnNiAl HEA, because Al changes the Fermi level and itinerant electron-spin coupling that leads to ferromagnetism.

MD calculations

The MD calculations are used for predicting the thermomechanical properties, which computes the materials from a molecular-scale and has a very extensive range of applications [137]. MD is well suited to reproduce the smart amplitude oscillations of atoms in the vicinity of crystal-lattice sites. The probability of an atom crossing the barrier from one lattice site to another is prohibitively low and will rarely occur on the MD time scale at low temperature [132]. The method mainly relies on Newtonian mechanics to simulate the motion of the molecular system. The samples are extracted from the whole system consisting of different states of the molecular system, and calculation is used to construct the configuration of the system.

Sharma *et al.* [24] investigated phase transformations (PTs) in Al_xCrCoFeNi using classical MD simulations. The influences of cooling rates, temperature, and Al contents on PTs were investigated by the MD simulation and experimental method in Al_xCrCoFeNi (Fig. 16). The cooling rates' effect could be neglected. Two transformations, molten/amorphous and amorphous/crystalline phases, were observed with the temperature decreased below the certain temperature. At last, the nucleation type was determined by the contents of Al. The nucleation type was the Al phase when x=10 and 20, while the amorphous phase when $x\geq 30$, respectively. This trend was consistent with the common neighbor analysis with lower Al contents [24].

CALPHAD modeling

The most important tools to design HEAs are the phase diagrams and thermodynamic properties depending on the thermodynamic databases. Gao et al. [40,44] summarized that the term, "thermodynamic database", means that the parameters for the Gibbs energies of a large number of binary and ternary systems are assembled, which are important for the intended compositional ranges. Gao also gave the schematic of the CALPHAD processing and suggested that the first step in developing a thermodynamic database of a HEAs system is to collect the thermochemistry and phase-equilibrium data from the articles about the lower-order systems, generally the related binaries and/or ternaries. However, if such data are not available unfortunately, it becomes necessary to design and carry out experiments. In this regard, the thermodynamic data obtained from the DFT calculations will be useful to fill up experimental data [31,116]. Thus, the reliable thermodynamic databases, containing a series of functions related to the composition and temperature, are assessed by the CALPHAD method, based on the reliable experimental data and accurate DFT results [40,50,113-117,119-122].

Previously, Zhang *et al.* [26] studied the phase stability of Al-Co-Cr-Fe-Ni HEAs and found that the experimental results are consistent with the CALPHAD modeling (Fig. 17). As one of the new-generation low-density structural materials for automobile and aerospace, the phase stability of Al-Co-Cr-Fe-Ni HEAs is a research focus, and the CALPHAD method proves a powerful tool to study the phase stability and transformation as well as the alloy design.

High-throughput methods

As the "Materials Genome Initiative " focuses on calculations, database, and experimentation in a three-in-one way, there are more examples of success in terms of computations and simulations as a way to speed up the material design [69,128]. Troparevsky *et al.* [112] used the high-throughput DFT to calculate the formation enthalpy between binary alloys to predict the formation of singlephase HEAs. The method correctly identifies all known single-phase alloys while getting rid of the similar elemental combinations that are known to form an alloy containing multiple phases. More importantly, they predicted plenty of potential single-phase alloy compositions and guided experimental studies. In Fig. 18, the highthroughput DFT calculation method demonstrates the higher accuracy in predicting new materials.

HEAs generally contain at least five alloying elements





Figure 16 (a) The influence of Al content on phase transition via E_{tot} -T (total energy per atom-temperature); (b) the phase type of nucleation via radial distribution at different temperature below 2,650 K. Reproduced with the permission from Ref. [24]. Copyright 2017, Elsevier.



Figure 17 Comparison of the CALPHAD calculations and experimental observation of $Al_{0.7}$ CoCrFeNi HEA. (a) Equilibrium calculation; (b) opticalmicroscope image of the specimen aged at 1,523 K for 1,000 h; (c) APT result: FCC_A1; (d) APT result: BCC_A2 + BCC_B2. Reproduced with the permission from Ref. [26]. Copyright 2016, Elsevier.

and every element with high content [20,40,45]. Such multi-component alloys are difficult to rely on the existing binary-alloy phase diagrams or ternary-alloy phase diagrams for the material design and formation prediction. Miracle and Senkov [19] used the CALPHAD calculations to predict the phase formation of the alloy and found that phase prediction is far from the experimental results, as shown in Fig. 19. Chen *et al.* [138] used CALPHAD calculations combined with DFT to establish a thermodynamic database especially for HEAs within a 15-element framework applied in many groups well.

FUTURE TRENDS AND PROSPECTS

HEAs have excellent properties, such as excellent me-



Figure 18 The predicted phase composition after one- or two- components added to FeCoNi and NbMoTa. Reproduced with the permission from Ref. [112]. Copyright 2015, the American Physical Society.



Figure 19 The number of formed phases for experiments and CAL-PHAD calculations. Reproduced with the permission from Ref. [19]. Copyright 2017, Elsevier.

chanical properties, high-temperature performance, as well as corrosion and irradiation resistance, which are the potential materials under extreme conditions. Although many studies focus on HEAs, our understanding of HEAs fundamental is rare. HEAs contain a variety of systems with more than five different elements in approximately equal-molar, which offer unlimited possibilities for the discovery and development of new alloys. Several future trends and prospects are proposed, based on the existing HEAs and materials-science literature, as following.

1. Among HEAs, CoCrFeMnNi [16], HfNbTaTiZr [139] and DyGdHoTbY [23] are classical representatives

of the equimolar single-phase HEAs in FCC, BCC, and HCP lattice structures, respectively. The two definitions of HEAs have been described, respectively. There are two restrictions for the former one, single-phase and equal molar fraction. Therefore, searching for HEAs with single-phase solid-solution structure with more than five principal elements is a research focus for decades. With the in-depth understanding of HEAs, these two restrictions have been broken. Now, the non-equimolar complex phases (the matrix is a solid solution) HEAs have been an important part of HEAs. Therefore, HEA can be divided into the first and the second generations, as illustrated in Fig. 20 and Table 2. The research scope of HEAs expands from the central region to the surroundings, which means the development from the equimolar single-phase solid-solution alloys to the non-equimolar multi-phase solid-solution alloys, which affects the definition of HEAs and the research scope. In other words, the evolution of HEAs conception facilitates the material design and development, meanwhile extends this idea to meet the specific needs, rather than rigidly adheres to the classical high-entropy definition. The non-equimolar complex-phases HEAs can be modified in the future to improve their performance by changing the component ratio and adding minor components to maintain a certain phase in special condition, or forming a second phase for precipitation strengthening or composite strengthening, such as the TRIP (transformation-induced plasticity) HEA Fe₅₀Mn₃₀Cr₁₀Co₁₀ [140], the precipitation-hardening HEA [74], and eutectic HEA [90].

2. At present, the common preparation methods of HEAs are traditional arc melting and induction melting. In order to shorten the development cycle and collect numerous data *via* a few experiments, new preparation methods, such as 3D printing, magnetron sputtering and directional solidification, etc. are applied to the study of HEAs. And with the application of high-throughput experimental method in HEAs research [141], a series of small samples with different composition or one sample containing composition gradient were fabricated. Combing with the highly developing technology of microscopy, material properties testing and computer, a lot of data and reliable relationship between composition and properties will be got in a short time.

3. As shown in the definition of HEAs, the elemental component, phase and thermodynamic properties (entropy) play the critical role on the properties of HEAs. Therefore, compared with experimental methods to design the HEAs, the alloy designing, based on the properties calculated by the DFT or MD method and the



Figure 20 The evolution of alloys.

Table 2 Characteristics for the two generations of HEAs



multi-component phase diagrams assessed by CAL-PHAD, is an effective method to save time and reduce the cost. However, the multiple elements (more than 5) and microstructures (solid solutions) of HEAs make the computational process much more complex and timeconsuming than that in conventional alloys. In addition, there is a certain gap between the phase composition of HEAs measured by the experimental method and the prediction by the CALPHAD method [69]. Therefore, it is crucial to develop reliable and robust databases for HEAs through the "Materials Genome Initiative". There is no doubt that advanced computing methodologies that can simulate the dislocation-related properties in HEAs are highly desirable nowadays.

4. There is a great opportunity for HEAs to overcome the bottlenecks of conventional alloys and to be applied in certain critical environment. For example, HEAs show great fatigue and fracture resistance [37–39], and superior high tensile strengths at cryogenic temperatures [36,39].

These outstanding properties make it possible for the HEAs to be used in turbine blades of aircraft [79], and cryogenic environments [36] as engineering alloys. HEAs possess low swelling after ion irradiation and some self-healing effects that has good application prospect in the field of nuclear materials [142].

5. The studies of HEAs are mainly focused on the bulk alloys, but less on the high-entropy films properties of corrosion resistance, wear resistance, irradiation properties, mechanical properties, and elevated temperatures stability. High-entropy nitride films have been applied in cutting tools [143]. Another significant potential application of high-entropy films is as diffusion barriers [144]. Sheng *et al.* [145] proposed huge application potentials of high entropy films in the fields of solar thermal conversion systems.

6. Although massive articles focus on the mechanical behavior of HEAs, while limited work focused on the elevated-temperature mechanical properties. More mechanical tests at room temperature and elevated temperatures with different strain rates should be done, which will be a precious guide to the broader application of HEAs, especially the tensile tests. Another interesting phenomenon found in the study of HEAs is that the stress-strain curve appears to be serrated in a specific temperature and strain rates range, and the occurrence of serration behavior will seriously affect the application of HEAs, because the unsmooth stress-strain curve is difficult to predict the material safety factor [146]. Serration behavior is closely related to the flow units in materials, and the flow units will change with the changes of testing temperatures and strain rates [147]. Since the serration behavior of HEAs reflects the deformation mechanism, it is necessary to study the serration behavior of HEAs more thoroughly.

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高熵合金材料研究进展

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摘要 根据人类开发材料的能力来看,合金成分经历了从简单到复杂的发展过程.合金的功能和性能不断改善,同时促进了人类文明进步. 具有多组分的高熵合金(HEAs)可以有效地改善合金的微观结构和性质.高熵合金具有诸如高强度和高硬度、优异的耐腐蚀性和热稳定 性、良好的抗疲劳强度及断裂强度、强耐辐射性等优异的性能,这是传统的合金无法比拟的.这些优异的性能也说明高熵合金未来具有 非常高的应用前景.近年来,高熵合金在各个领域也呈现出快速发展的趋势.为了更好地了解高熵合金的基础,未来快速开发出具有更加 优异性能的高熵合金,本文综述了近年来关于高熵合金的发展.高熵合金的发展已经经历了两个阶段,第一个阶段为等摩尔-单相固溶体 结构的高熵合金,第二阶段为非等摩尔比的多相固溶体高熵合金.本文主要讨论了高熵合金的制备方法、组分设计、相形成和微观结 构、优异的性能和高熵合金在计算模拟方面的应用,同时提出了高熵合金的未来发展趋势和前景.