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Functional properties and promising applications of high entropy alloys

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ABSTRACT

High-entropy alloys are a novel class of complex materials discovered near the center of phase diagram, and show exceptional performances over the traditional alloys. The ability of high-entropy alloys to be designed unique combinations of mechanical and functional properties in unlimited space of alloy compositions is encouraging. The unconventional chemical structures hold promise for achieving unprecedented functional performances, making it a potential functional material in the field of soft-magnetic, anti-radiation, catalyst, photothermal conversion materials, etc. Here, the recent progress in understanding the salient features of high-entropy alloys is reviewed. The functional performances and underlying mechanisms are carefully discussed.

Introduction

The future development of high-entropy alloys (HEAs) should not be focused on the reproduction of properties achieved by conventional alloys or slight improvements in existing performances. Factually, the high content of alloying elements inevitably increases alloy costs, but usually obtains minor performance improvements [1,2]. In this case, the significance of HEAs should not be limited to the mechanical performances, but should be designed an unprecedented combination of specific functional properties and mechanical properties utilizing the novel alloy compositions in the center of phase diagram. With reference to current researches, HEAs have achieved salient breakthroughs in functional applications, including soft-magnetic materials [3–5], irradiation resistance materials [6–9], superconducting materials [10,11], and photothermal conversion materials [12,13], diffusion barrier films [14–16].

The novel alloy design concept and high-concentration solid solution structure give HEAs many unique kinetic and thermodynamic characteristics, which makes it possible to develop special functional properties [17,18]. Moreover, the properties of HEAs can be more easily adjusted than conventional alloys due to the large compositional variation [19,20]. Generally, the functional properties are available from traditional materials. However, traditional materials usually fail to provide a well-service at extreme conditions, and cannot simultaneously possess excellent mechanical properties [1]. In this case, the excellent mechanical properties of HEAs under extreme environments provide a major boost for making them excellent functional materials [21,22]. It is imperative to carry out the researches on functionally oriented HEAs, and establish a relationship between the structure and functional properties.

Key research topics

Categories, mechanisms and advantages

Compared to initial definition that alloys containing more than five components in equal or near equal atomic percent (at.%), the current definition of HEAs has been extended to alloys with dual or multiple phases in non-equal molar components [23]. The promising goal for HEAs is no longer to develop the perfect HEAs that matches well with the definition, but to utilize the "entropy" to regulate the composition to obtain unprecedented combinations of specific functional properties and mechanical properties. According to the current researches, the categories, mechanisms and advantages of HEAs are summarized from both mechanical and functional performances, as shown in Fig. 1. There are several superior performances that have been proved for HEAs, namely, overcoming the trade-off of strength and ductility, excellent low-temperature plasticity, good thermal stability, exceptional

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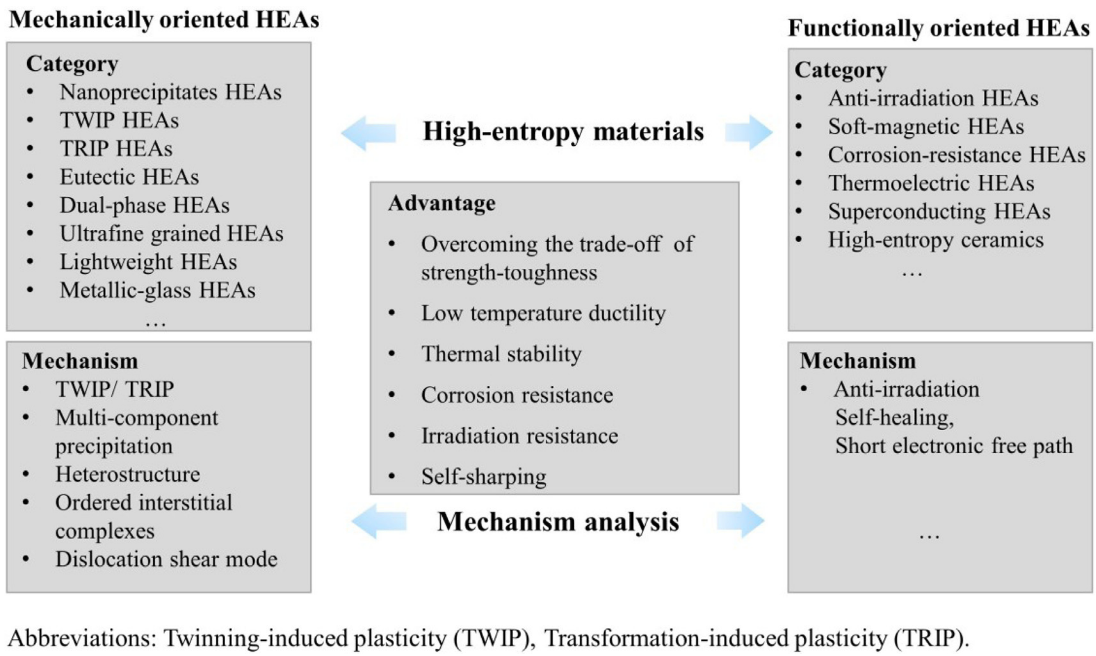

Fig. 1. The categories, mechanisms and advantages of high-entropy materials.

corrosion resistance, radiation resistance and self-sharping [24–27]. Fig. 1 also indicates that researches on HEAs are mainly focused on the mechanical performances in the past few years. Compared with the mechanically oriented HEAs, the studies on functionally oriented HEAs are weak, and mechanism analysis is more ambiguous.

Functional performances

Irradiation resistance properties

The unexpected stability of phase structure and mechanical performances in extreme environments makes HEAs attractive candidates for irradiation resistant materials. The integrated experiment and modeling work indicate that HEAs show a significantly lower volume swelling rate and defect density than that of traditional alloys [28–30], which may be attributed to the effective self-healing mechanisms of HEAs under irradiation conditions. The possible self-healing mechanisms of HEAs is illustrated in Fig. 2.

It is well known that particle irradiation could cause atomic displacements, which induces the irradiation defects such as vacancies and interstitials, and also is accompanied with thermal spikes. Generally, the interstitials in conventional alloys migrate rapidly along the direction of the Burgers vector in a long-range one-dimensional mode, while the short-range three-dimensional (3D) motion of interstitial clusters occurs in the HEAs [31]. The short-range 3D motion of interstitial clusters significantly increases the probability of vacancy-interstitial recombination, further reduce the defect density and void swelling in materials, as shown in Fig. 2-I. The novel short-range 3D migration paths caused by the chemical disorder and compositional complexity of HEAs could promote the disappearance of radiation damage, and further improve the radiation tolerance.

Another self-healing mechanism is attributed to the high atomic-level stresses that originated from mixing of elements with different atomic sizes in HEAs. The simulation results indicate that the atomic-level stresses, which destabilize the solid solution, will facilitate amorphization of alloys. The thermal spikes caused by particle irradiation bring local melting and recrystallization, which can improve the orderliness of the alloy, and significantly reduce the density of defects, as shown in Fig. 2-II [32,33]. In addition, the energy transfer mechanism of HEAs is also discussed from the complex electronic correlation [8]. The electron mean free path decreases significantly with the increase of the number of component elements, meaning a lower energy consumption efficiency in HEAs. This action can prolong thermal spike and strongly promote the recovery from the injured state. Briefly, the recent progress in HEAs demonstrates the possibility of obtaining high radiation tolerance through unique damage self-healing mechanisms. It is desirable to design high performances HEAs for serving as irradiation resistance materials.

Soft-magnetic properties

The conventional soft-magnetic materials, such as silicon steels and Fe-Al alloys, have a relatively high degree of structural order which results in poor deformability. The amorphous soft-magnetic materials possess excellent corrosion resistance and high electrical resistivity. However, harsh manufacturing conditions make it impossible to obtain bulk amorphous alloys. Furthermore, the crystallization temperature of amorphous alloys is low, which further limits working temperature. As pointed out recently, HEAs have become one of the most promising soft-magnetic materials due to their flexible forming process, excellent deformability, corrosion resistance, and acceptable soft magnetic properties [1,3]. Fig. 3 shows the comparison of soft-magnetic properties between HEAs and conventional materials, including electrical resistivity (Ω), saturation magnetization (M_s), and coercivity (H_c). Fig. 3a indicates that the M_s and H_c values of HEAs are mainly located in the region of soft and semi-hard region, and the properties of partial alloys are accepted by the soft-magnetic materials. In addition, HEAs generally possess higher electrical resistivity in comparison with traditional alloys due to large lattice distortion, as shown in Fig. 3b.

Generally, the soft-magnetic HEAs are developed on the basis of ferromagnetic elements Fe, Co and Ni. Ferromagnetic elements that excite the magnetic behaviors can be treated as the main elements, while components designed to improve the corrosion resistance, mechanical properties, and resistivity are regarded as the auxiliary elements, such as the Si, Al, Ni and B. From composition

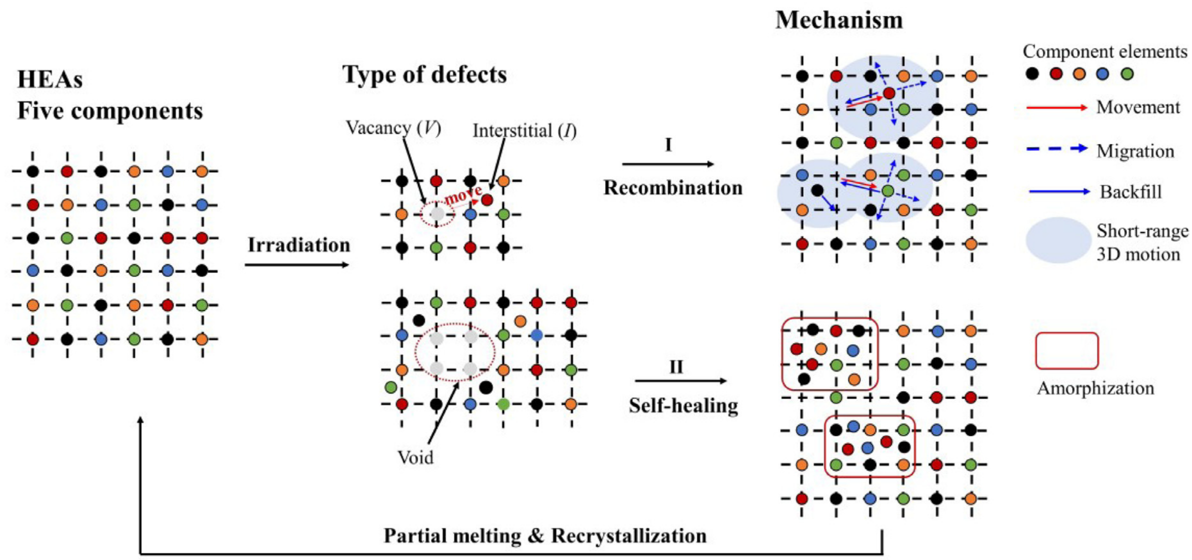

Fig. 2. Schematic diagram for the self-healing mechanism of irradiation resistance HEAs.

design perspective, there are several advantages of HEAs over traditional magnetic materials: (i) The increase of principal element number can reduce the structural order of alloy system, thereby the deformability is improved. (ii) The auxiliary properties, such as corrosion-resistance, thermal stability and mechanical properties, are easier to adjust due to the large compositional variation. (iii) High mixing entropy contributes to the stability of simple solid-solution phases, which can avoid magnetic domain wall pinning caused by phase boundaries. (iv) The large lattice distortion in HEAs increases the resistance of electron movement, resulting in high electrical resistivity and low eddy current loss.

It should be noted that the coercivity of HEAs is relatively high. The recent researches show that the directional solidification plays a positive role in reducing coercivity by controlling the morphology and crystallographic texture [4]. The coercivity value of $FeCoNiAl_{0.2}Si_{0.2}$ alloy manufactured by directionally solidified is reduced to 315 A/m, which is much lower than the as-cast alloy with 1400 A/m. To further understand the effect of alloying elements and structures on the magnetic performances of HEAs, the ab-initio and density functional theory (DFT) have been conducted [3,43,44]. Modeling results prove that the chemical short-range order in HEAs significantly changes the local environment of atomic, which further reduces the average magnetic moment of magnetic atoms [34]. Briefly, the excellent mechanical properties over a wide temperature-range and corrosion resistance guarantee a well-service of HEAs under the extreme environments. In addition, the superior ductility of HEAs provides the possibility of preparing thin plates, which can effectively reduce eddy current losses of magnetic devices [35]. The optional magnetic performances and mechanical properties provides a strong motivation for future development of soft-magnetic HEAs.

Thermoelectric properties

The ideal thermoelectric materials should have high Seebeck coefficient, high electrical conductivity, and low thermal conductivity [42]. However, these three parameters are intrinsically intercorrelated, meaning that optimizing one parameter will inevitably sacrifice others. As a potential thermoelectric material, the thermal conductivity of HEAs is several orders of magnitude lower than that of traditional alloys, but the electrical conductivity is also reduced inevitably. Furthermore, the highly symmetrical solid-solution structures of HEAs are more likely to contributes the convergence of electronic bands, thereby obtaining a higher Seebeck coefficient [45]. Clearly, HEAs are expected to be used as thermoelectric materials with high thermoelectric efficiency.

As pointed out recently, designing high-entropy thermoelectric materials by doping existing low-entropy thermoelectric materials is a viable and cost-effective approach, which could significantly reduce the thermal conductivity. Simultaneously, the increase of mixing entropy facilitates to improve the stability of simple solid-solution structure, and markedly optimizes the Seebeck coefficient [42]. For instance, the overall thermoelectric efficiency has been significantly improved by doping SnTe alloys with appropriate Mn and Ga, although at the cost of reduced electrical conductivity [46,47]. Similar conclusions have also been confirmed in multi-component thermoelectric materials, such as $(AgCu)(InGa)Te_2$- and $Cu_8Ge(SeTe)_6$-based thermoelectric materials [48].

Catalytic properties

The high overpotential, durability, selectivity and limited composition are the common problems in the application of single metal-based catalysts [49,50]. It is encouraging that the flexible composition design and solid-solution structure of HEAs provide a huge configuration space for designing new catalyst materials. Many studies have been conducted on the catalytic properties of HEAs, such as CO oxidation [51], ammonia oxidation [52], and oxygen reduction [53]. Taking ammonia oxidation catalyst as an example, the PtPdRhRuCe nanoparticles fabricated by carbothermal shock method show ~100% conversion of ammonia and more than 99% selectivity toward NO_x. The HEAs catalyst also exhibits good durability, and no degradation in catalytic activity or selectivity over ~30 h of continuous operation at 700 °C is observed in this work [52]. Similarly, the degradation in catalytic activity of $Co_{25}Mo_{45}Fe_{10}Ni_{10}Cu_{10}$ HEAs is negligible after 50 h of continuous operation at 500 °C in NH_3 decomposition reaction [54]. The modeling work indicates that solid-solutions structure possess higher energy barriers, thereby preventing the surface segregation induced by the adsorbate under electrocatalysis [52]. The high catalytic efficiency and durability are also observed in the CO_2 and CO reduction reactions [49].

For HEA catalysts, the adsorption energy distribution can be changed by adjusting the composition, which further optimizes the

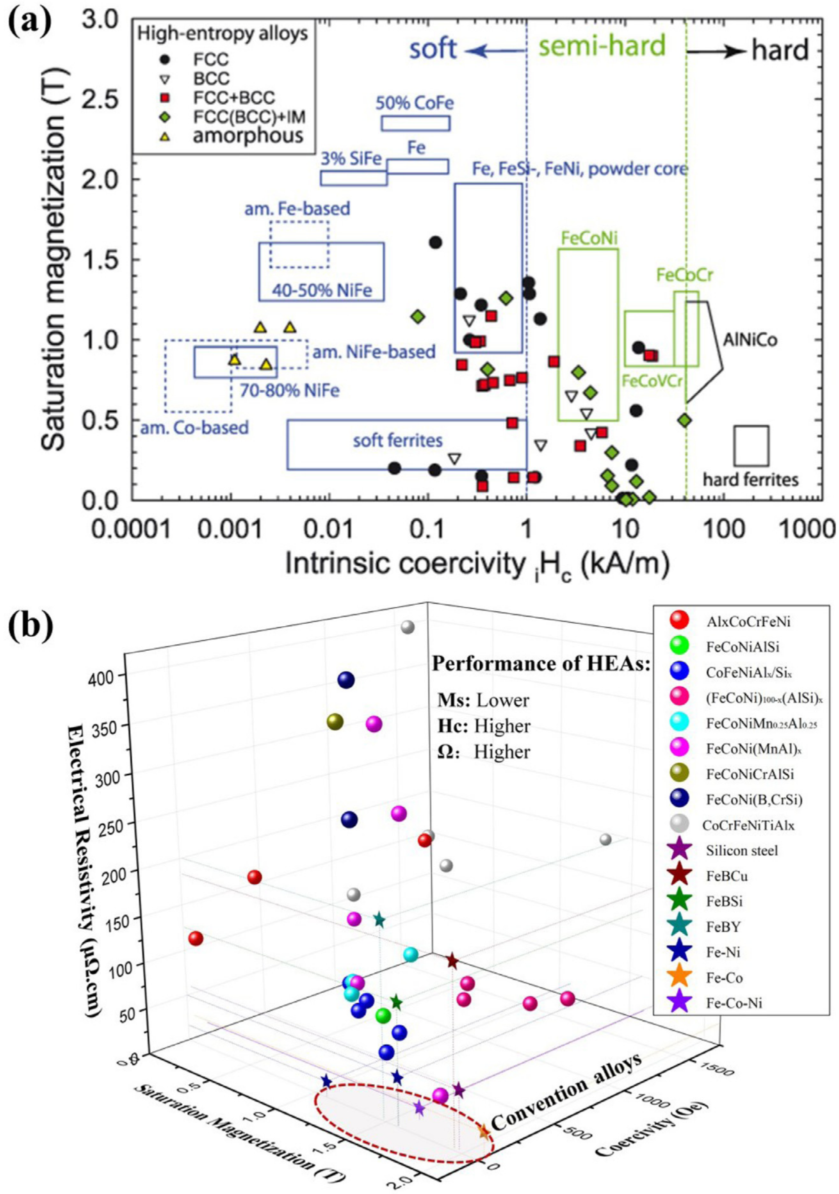

Fig. 3. (a) Saturation magnetization (T) versus coercivity (Hc) of HEAs compared with major conventional soft and semi-hard magnetic materials (Cited from Ref. [41]). (b) The electrical resistivity (Ω), T, and Hc of HEAs compared with those of conventional alloys (stars for the performances of conventional alloys, balls for the performances of HEAs) [4,35-41].

selectivity and activity of the catalyst. In fact, the outstanding performance is not only caused by simple element blending, but more likely from the synergistic effect of multi-element composition and solid solution structure. Briefly, multi-elemental composition in an alloy structure plays a positive role in high-performance catalysts.

Functional high-entropy films

The multi-component films developed on the basis of similar concept of HEAs, also referred as high-entropy films (HEFs), have generated great excitement as a new class of films [55]. The attractive performances, such as thermal stability, wear resistance, corrosion resistance and high strength, make them promising functional materials. The functional performances and mechanisms of HEFs are shown in Table 1. Here, the potential applications of HEFs as solar absorber coating and diffusion barrier film in integrated circuits are mainly discussed.

Solar absorber coating

The solar absorber coating is a key factor in determining photothermal conversion efficiency. In addition to the high absorption efficiency, the excellent corrosion resistance and mechanical properties at elevated temperature are also required for application in harsh environments. Recently, the unique combination of functional and mechanical properties of HEFs attracts interest in solar absorber fields. Moreover, Ye et al. [61] have found that the uneven atomic plane caused by severed lattice distortion can facilitate the phonon scattering, thereby increasing the energy absorption.

The $NbTiAlSiN_x$ and $NbTiAlSiWN_x$ films have been systematically investigated for solar absorber coating [62,63]. Results demonstrate that the NbTiAlSi-based HEFs exhibit excellent thermal stability after annealing at 700 °C for 24 h. The $NbTiAlSiN_x$ films show a relatively high solar absorptance of 0.079 [62]. To further obtain the higher absorptivity through enhancing extinction interference, the multiple layers have been designed as shown in Fig. 4. For example, the multi-layer MoNbHfZrTiN/MoNbHfZrTiON

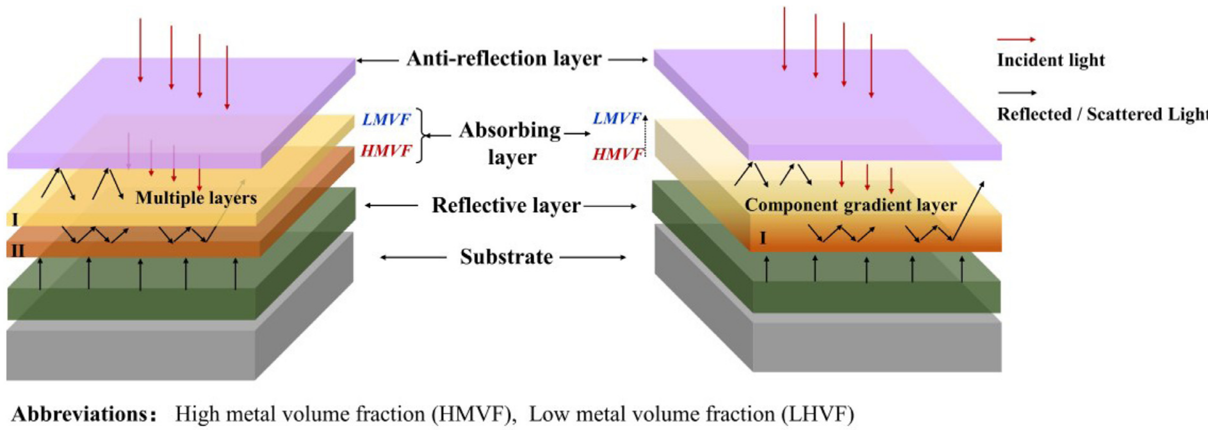

Fig. 4. Schematic diagram for the solar absorber coatings including multiple absorbing lays and component gradient absorbing layers.

Table 1
The preliminary evaluation of functional performances of high-entropy films.

Performances	Evaluation	Mechanism	Ref.
Thermal stability	Exceptional	The slow co-diffusion effect slows down the phase transition rate, hinder grain growth, and improve creep resistance.	[22]
Anti-oxidation	Good	Synergistic effect of low oxygen solubility, slow diffusivity rate, and stable oxide scales formed by sufficient amount of the elements.	[56,57]
Thermal conductivity	Lower	The chemical disorder reduces the electron mean free path, and further reduces the thermal conductivity.	[58]
Corrosion resistance	Exceptional	Synergistic effect of slow diffusion, amorphous and nanocrystalline structure, and strong passivation layers formed by doping elements.	[59]
Wear resistance	Good	Severe lattice distortion and solid solution strengthening facilitate to obtain promising wear resistance.	[60]

HEFs have been developed for the solar absorber coating [12]. It shows extremely excellent performances with the absorptance and emittance value of 0.935 and 0.07, respectively. Moreover, it could be speculated that designing a composition gradient film with a graded metal content is also an effective way to improve the solar absorptance, as shown in Fig. 4. In addition, the method of co-sputtering deposition was applied to the combinatorial fabrication and screening of potential photothermal conversion materials [13]. The phase structure and optical performance of FeCrVTaW alloy films were systematically tested, and the solar absorptance reached ~0.08.

Diffusion barrier films

In order to inhibit rapid interdiffusion between Si substrate and Cu metallization in integrated circuits, an effective diffusion barrier layer is strongly demanded. The outstanding thermal stability and diffusion resistance of HEFs make it possible to be used as diffusion barrier films, especially for maintaining the robust diffusion barrier ability under the elevated temperature and lower thickness. The study of Jiang et al. indicates that no Cu-Si intermetallic compounds generated in the interconnect structures after annealing at 900 °C when AlCrTaTiZr/AlCrTaTiZrN films are used as diffusion barrier films [64]. Moreover, the (AlCrRuTaTiZr)Nx film with a total thickness of only 4 nm also shows a good diffusion barrier ability [15]. Under annealing at 800 °C, the interdiffusion between Cu and Si through the multilayer structure is effectively prevented.

Prospects and future opportunities

In the short term, several specific issues remain to be solved to promote the innovative applications of HEAs. First, exploring the interaction between entropy and enthalpy of HEAs is a key issue to obtain optimal performance. Second, internal structure of HEAs is more complex, and the effect of local chemical order on the functional performances of HEA remains an open issue, which is generally confirmed by indirect calculation methods rather than experiment. Third, the concept of “high-entropy ceramics” has proposed in the fields of engineering and energy, such as high-entropy nitrides, carbides, and oxides. In contrast to the HEAs, the solid-solution unit transforms from the metal atom into binary compound, making the traditional phase formation rules no longer applicable to high-entropy ceramics. Therefore, the development of phase formation rules for high-entropy ceramics is also a meaningful work.

In summary, the unique characteristics of HEAs in dynamics, thermodynamics, and structure have aroused great interest in functionally oriented materials. The HEAs may open additional pathways for designing promising functional materials with unprecedented combinations of functional and mechanical performances. Designing and developing potential innovative applications to meet the emerging requirements, rather than simply copying existing performance, is a promising future pursuit of HEAs.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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