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High-entropy materials

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Abstract

Primarily over the last decade, the concept of multiple principal element metallic materials, commonly referred to as high-entropy alloys or more generally high-entropy materials, has taken the field of materials science, particularly structural metallurgy, *by storm*, at least as measured by the plethora of publications that are focused on this topic. In this brief article and the following six short reviews, we attempt to distill what all this is about, with a description of why these materials may be important, why they may differ from traditional materials and how theoretical, computational, and experimental studies can shed light on the science underlying their behavior and potential application.

Introduction

Alloys consisting of multiple elements in which no single element can be considered the principal (or base) element have been the focus of considerable research since 2004 [1, 2]. Various referred to as high-entropy alloys (HEAs), compositionally complex alloys (CCAs), or multi-principal-element alloys (MPEAs), the field has seen explosive growth recently, especially since 2013 judging by the number of published papers and citations in *Web of Science* and is now a “hot topic” in materials science. From 2015 onwards, high-entropy ceramics (HECs) have entered the fray [3] and are garnering increasing interest. In the future, it would not be surprising to witness similar growth in high-entropy polymeric and glassy materials. To be inclusive of all these different classes of materials, we use the term high-entropy materials (HEMs) here. Additionally, for the sake of brevity and to avoid having to introduce various confusing nomenclatures, HEMs in this article include multiphase variants of these materials (*i.e.*, we do not restrict ourselves to solid solutions).

Why all the interest?

One oft-cited reason is the mind bogglingly large number of distinct compositions that can be created by combining multiple different elements together [e.g., 4] only a tiny fraction of which been investigated. This vast unexplored compositional space is assumed to contain rich veins of useful materials just waiting to be mined. However, as one of us has on occasion pointed out [e.g., 5], the so-called Anna Karenina principle — all happy families are alike; each unhappy family is unhappy in its own way, popularized by Jared Diamond in his book *Guns, Germs and Steel* [6] — should give us pause, or at least temper some of the prevailing unbridled optimism. Briefly, the principle states that when success can be undermined by deficiencies in any one of multiple governing factors, the likelihood of failure increases dramatically. Diamond uses domestication of animals to demonstrate that, despite the large number of candidate

animals, only a small fraction has been successfully domesticated. Take zebras and horses, for instance: they appear to be identical — except obviously for the stripes; yet only one has been domesticated (for a variety of reasons). Not unlike happy families, therefore, successful domestication is foiled if even a single relevant factor is lacking.

Similar constraints bedevil the discovery, development, and deployment of new and useful materials. First, it is rare for just one property to be a critical requirement; rather, an optimal balance of several different properties is usually needed. For example, in structural applications, strength, toughness, ductility, density, corrosion resistance and cost are just some of the factors that may have to be simultaneously optimal. An alloy that has sufficient strength but is too expensive is unlikely to be practical in most cases. Second, even if we suppose that only a single property is a critical requirement, that property may nevertheless be affected by multiple factors, none of which can exhibit deficiencies, or failure will occur. Consider the case of fracture which is a “weakest-link” phenomenon. Any one of multiple factors can result in brittleness, including complex crystal structure, insufficient number of slip systems, low mobile-dislocation density, high stress to move dislocations, weak grain boundaries, harmful segregants, lack of sustained strain hardening (or worse, strain softening), porosity, cracks, etc. It is not sufficient to eliminate one or two of these weak links because all it takes is one of them to cause brittle fracture. Therefore, every single one of them must be eliminated or overcome by alloying and processing. Additionally, a material that initially has sufficient ductility may become embrittled when its microstructure undergoes certain changes during service or after it has been exposed to certain environments. Given all this, only a small subset of the available compositions (the proverbial needle in a haystack) may well be suitable for a given application.

Admittedly, structural materials set a high bar since their relevant properties are notoriously difficult to balance. Not all materials of interest may be as severely constrained; therefore, some may be easier to optimize than others. Additionally, as pointed out by Curtin et al. in this issue: “from among the millions of possible alloys, we only need to discover a handful that can satisfy multiple requirements” [...for this endeavor to be successful]. The relative youth of the HEM field may explain why few new materials with dramatically superior combinations of properties have been discovered so far. In time, we will be in a better position to judge the true potential of this new class of materials. Meanwhile, how do we search for the hidden gems?

Combinatorial and high-throughput approaches

Given the vastness of the compositional space that needs to be explored, high-throughput experimental and theoretical techniques have been proposed for rapid screening. Traditionally, thin-film deposition by evaporation or sputtering has been the usual way to obtain gradients in chemical composition that span large areas of multicomponent space in a single specimen [e.g., 7, 8]. Spatially resolved characterization of these films then allows correlations to be drawn among composition, structure, and certain properties. Large alloy libraries can be rapidly synthesized and, depending on the characterization technique used, local structure and properties can also be measured relatively quickly. Examples of fast characterization techniques include structure determination using high-intensity synchrotron X-rays and hardness/modulus measurements using nanoindentation [e.g., 9, 10]. If bulk specimens are needed, say for tensile testing, rapid alloy prototyping by conventional casting [e.g., 11] or additive manufacturing [e.g., 12] is preferred over thin-film deposition. Both chemically homogeneous and compositionally graded specimens can be produced in relatively bulk forms [13, 14]. Once regions of interest with close-to-desired microstructures, properties, or both, have been identified by high-throughput screening, bulk alloys in that vicinity can be synthesized with tighter control of composition for detailed characterization and optimization.

Knowledge of the phases present in multicomponent systems as a function of external variables such as temperature is key to controlling microstructure and, hence, properties. As Zhang and Yang point out in this issue, thermodynamic calculations based on the CALPHAD approach, despite their limitations, are currently the only viable way to accomplish this. The accuracy of the calculated results depends on the reliability of the underlying thermodynamic databases which are typically extrapolations from unary, binary, and some ternary systems. Comparison with experimentally observed microstructures helps refine the databases and improve their accuracy. When coupled with high-throughput calculations of phase stability [15], CALPHAD can be a powerful and complementary technique to combinatorial experiments.

High-throughput first principles calculations can also rapidly predict phase stability in multicomponent space [16, 17], albeit with some limitations. The approach considers all possible binary pairs among the constituent elements of any given HEA and assumes that if there are any that show a strong tendency for clustering (positive mixing enthalpy) or a strong tendency for compound formation (negative formation enthalpy), then a mixture containing these elements is unlikely to form a solid solution. By comparing with alloys that are known (experimentally) to form solid solutions, limit values are then set for the calculated enthalpies between which solid solutions are expected to be stable. An advantage of this approach is that many more alloys can be treated computationally than experimentally allowing for the possibly faster discovery of new compositions with potentially interesting combinations of elements. A disadvantage is that the enthalpy limits bounding the solid solution window are somewhat arbitrary and thus are not reliably predictive. Nevertheless, like the experimental combinatorial approach, high-throughput computational approaches can serve as useful screening tools to identify interesting regions in multicomponent space for further investigation, keeping in mind the following caveat. Since the mechanical properties of many metallic alloys are a function of their (often metastable) microstructures, Pickering and Jones [18] recently noted that “One can envisage that rapid approaches might dismiss most steels (had they not yet been discovered), due to their polymorphism and tendency to form brittle martensite when cooled quickly.”

Mechanistic approaches

A more fundamental approach relies on in-depth studies of select model alloys from which mechanisms governing basic phenomena and properties can be identified. If the underlying factors behind those mechanisms are well understood, they can be generalized to a wider class of alloys with like characteristics to trigger the same mechanisms. Here, mechanical properties are offered as an example to illustrate this approach, which can, in principle be applied to other properties also. The equiatomic CrMnFeCoNi alloy (the so-called Cantor alloy) is one of the most studied HEAs. Above 800 °C, it is thermodynamically stable as a single-phase solid solution having the face-centered cubic (FCC) structure, but it decomposes into a few different metallic and intermetallic phases at lower temperatures [19, 20]. The decomposition occurs at a slow enough pace in the case of micron-size or larger grains that cold working followed by annealing retains the solid solution state [21, 22]. Its strength and ductility simultaneously increase with decreasing temperature down to the cryogenic range [23]; consistent with this, it exhibits very high fracture toughness down to liquid nitrogen temperature [24]. In relatively ductile metals, tensile ductility is usually limited by the onset of necking as given by the Considère criterion. Postponement of necking by the introduction of mechanisms that provide strong, sustained, work hardening leads to greater uniform elongations. Early on, nanotwinning was identified as a potent work-hardening mechanism [25] that can result in simultaneously high strength and ductility. Since twinning in FCC metals can occur by movement of Shockley partials on {111} planes, and these partials also produce trailing stacking faults, one would expect nanotwinning to become easier if the stacking fault energy (SFE) is decreased. Consistent with this notion, a related medium-entropy alloy (equiatomic CrCoNi),

with a somewhat lower SFE, twins more readily than CrMnFeCoNi [26] resulting in a higher strength-ductility combination [27] and fracture toughness [28] than those of CrMnFeCoNi.

Phase boundaries introduced during straining can have a similar beneficial effect on strength and ductility as the nanotwinning described above. The earliest study demonstrating this effect utilized an off-equiatom variant of the Cantor alloy [29]. Its composition was such that FCC was the stable structure at elevated temperatures but hexagonal close packed (HCP) at lower temperatures. Consequently, the alloy underwent a partial FCC to HCP (martensitic) transformation upon cooling after thermomechanical treatment. Subsequent tensile straining led to deformation-induced transformation of the remnant (metastable) FCC to HCP which resulted in increased strength and ductility compared to alloys that showed little or no such martensitic transformation. Since low SFE energy promotes twinning as well as FCC to HCP transformation, its value can be tuned by tweaking the alloy composition to activate one or the other, or both, mechanisms.

It is worth noting that high strength-ductility combinations require high values of work hardening that can be sustained over large strains. Obstacles such as deformation-induced twin boundaries and phase boundaries have the advantage that their density is not static but increases with strain. Under the right conditions, therefore, they can overcome the dynamic recovery processes that tend to naturally decrease the hardening rate. In this respect, static obstacles such as grain boundaries and precipitates are at a disadvantage because their density is fixed at the start of deformation and does not change with strain; therefore, they can have only second-order effects on obstacle density (by increasing the dislocation density). Nevertheless, under the right circumstances, it has been shown that a high volume-fraction of multi-element intermetallic precipitates results in significant and simultaneous increases in strength and ductility [30]. Dense dislocation walls and microbands of plasticity were observed in strained specimens, which presumably allow for increased dislocation storage and, in turn, sustained work hardening.

Recently, the two mechanisms of precipitation hardening and deformation-induced martensitic phase transformation have been employed synergistically to achieve simultaneously high strength and ductility in a medium-entropy alloy (MEA) [31]. Two alloys were compared, one a single-phase solid solution and the other a two-phase alloy with nanoscale precipitates uniformly distributed in a matrix having the same composition as the solid solution alloy. The composition of the single-phase alloy was such that it underwent a martensitic transformation from FCC to body-centered cubic (BCC) upon quenching from elevated temperatures. Although the matrix of the two-phase alloy had the same composition, its transformation to BCC was suppressed by the constraint of the surrounding precipitates. Consequently, its matrix after quenching remained in a metastable FCC state. Upon tensile straining, the matrix progressively transformed from FCC to BCC which, along with the normal precipitate strengthening mechanism (cutting or bowing depending on precipitate size and spacing), resulted in significantly higher strength and ductility compared to the solid solution alloy. This dual functionality of the precipitates was confirmed by altering precipitate size and spacing while keeping their volume fraction constant. Contrary to what would happen if conventional precipitate hardening was the only operative mechanism, finer and more closely spaced precipitates resulted in lower strength than when the precipitates were coarser and spaced further apart. In other words, when the spatial confinement due to precipitates becomes too high, it completely suppresses the formation of deformation-induced martensite, thereby eliminating that mechanism during straining and weakening the alloy. Only when the two mechanisms (conventional precipitate hardening and deformation-induced phase transformation) act in concert can the highest strength be obtained.

The above examples show that it is possible to activate individual or multiple deformation mechanisms, sequentially or in parallel, to obtain the best possible combinations of strength and ductility. This mechanism-based approach is a “bottom-up” approach and requires fundamental knowledge of the various factors, both at the chemical/atomic scale and the microstructural scale, that affect the relevant mechanisms. An advantage of HEAs and MEAs is the presence of multiple compositional knobs that can, in principle, be individually tuned to affect each mechanism separately. For example, a certain element might lower the SFE, which also affects twinning propensity and transformability from FCC to HCP. However, another element might affect the propensity for transformation from FCC to BCC. In the latter case, one can imagine an alloy that initially deforms by ordinary dislocation plasticity, followed by twinning and eventually by phase transformation to achieve an extended region of sustained work hardening. Furthermore, prior to the onset of plasticity and work hardening, it may be possible to achieve high yield strengths by tuning yet another element in the alloy that hinders the initial activation/movement of dislocations without affecting the other aspects above. Such tailored control over individual mechanisms is difficult in conventional alloys containing only one or two principal elements. In the long run, this ability to control individual mechanisms, precisely when needed, may well prove to be the single biggest advantage of HEMs.

In this issue

In addition to this introductory paper, this issue comprises five articles on metallic materials and one on ceramics, reflecting the relative intensities of current and past research activities in these two classes of materials. Research on high-entropy ceramics has been gathering pace of late and will likely constitute a larger fraction in the future.

The paper by Curtin et al. focuses on modeling the yield strength and phase behavior of HEAs by utilizing broadly applicable theoretical frameworks for understanding defect structures, their energetics and kinetics. Solute interactions with dislocations are presented as illustrative examples of the broader problem of interactions of solutes with other kinds of defects such as grain boundaries and crack tips. Their optimistic conclusion is that the current state of theory and modeling is such that they can provide sufficiently accurate guidance to experimentalists for down-selection from the vast number of possible compositions to those that might be interesting from a scientific or technological viewpoint.

The paper by Zhang and Yang reviews CALPHAD type thermodynamic simulations and their coupling with mobility databases to predict phase stability and kinetics of HEAs. Deficiencies are identified, mainly in the databases of the constituent ternaries, that currently limit the accuracy of phase stability predictions in quaternary, quinary and higher order systems. While experimental studies of phase stability and diffusion are needed to provide additional data to shore up current databases, they also discuss the value of machine learning models that combine CALPHAD-generated features with additional material descriptors to improve the fidelity of phase predictions in multicomponent systems.

The paper by Inui et al. reviews uniaxial mechanical properties of single- and multi-phase HEAs with a focus on yield strength, work hardening behavior and ductility. Fundamental aspects of deformation behavior are discussed by analyzing single-crystal properties. In the case of FCC HEAs, except for the fact that they exhibit very high levels of solid solution strengthening, the basic features are similar to those of conventional FCC metals. In the case of multiphase alloys, a wide range of behaviors have been reported depending on the operative deformation mechanisms, again not unlike those seen in conventional alloys.

How these uniaxial mechanical properties of HEAs play out in terms of their fracture resistance and damage-tolerance is discussed by Gludovatz and Ritchie, who note the limited studies on this vital aspect for these materials. As described above, the single-phase FCC HEAs, specifically those based on the Cr-Co-Ni systems, display remarkable strength with ductility which translates into exceptional fracture toughness properties that are comparable to those of the toughest materials known. Moreover, in certain alloys these properties are further enhanced at cryogenic temperatures, largely because of a synergy of deformation mechanisms, primarily dislocation slip, stacking fault formation, nanotwinning and phase transformation [32, 33], which can act in sequence to prolong strain hardening and delay the necking instability. Conversely, BCC HEAs, particularly those involving refractory elements which are generally characterized by ultrahigh elevated-temperature strength, appear to display very low tensile ductility and toughness, a problem that must be solved by alloy design and microstructure control if these materials are ever to see extensive industrial use.

One intriguing aspect of HEAs is whether these non-dilute solid solutions with multiple principal elements can induce unconventional atomic structures or elemental distributions, such as local chemical ordering or clustering, which could affect defect behavior and enhance mechanical properties, a topic which is discussed by Zhang et al. The existence of such compositional heterogeneity, e.g., in terms of the pairing of like vs. unlike atoms in both the FCC and BCC alloys, has been predicted in atomistic simulations using density functional theory-based Monte Carlo methods and molecular dynamics. [e.g., 34, 35], and detected experimentally, e.g., with extended X-ray absorption fine structure analysis [36] and using transmission electron microscopy in CrCoNi [37-39]. Although such studies have indicated that local chemical ordering can affect critical deformation properties as the stacking-fault energy, dislocation motion and slip mode, the actual effect of such compositional heterogeneity on bulk mechanical properties has been difficult to quantify and is still open to question. Suffice to say, if such local order can be mechanistically understood, controlled, and found to have a definitive influence on properties such as strength, ductility and strain hardening, the prospect of “tuning disorder with order” represents a further fascinating means to tailor these materials to achieve desired properties.

Last but certainly not least, the paper by Toher et al. examines non-metallic materials and summarizes progress on disorder-enhanced properties of HECs. They discuss how configurational entropy can stabilize multi-component ceramics, more so than metallic alloys. Examples are highlighted where disorder allows the attainment of properties generally viewed as mutually exclusive, for example, high stiffness with low thermal conductivity, or high hardness with good fracture toughness. The authors argue that high-entropy ceramics will play an important role in the future in batteries, catalysts, thermoelectrics, magnets, ferroelectrics, and coatings for harsh environments.

Outlook

The notion of designing materials with multiple principal elements, which essentially started by looking for potential metallic alloys at the center of phase diagrams rather than at the edges or corners and (at least in theory) by avoiding the presence of undesirable intermetallics by promoting configurational entropy relative to the enthalpy of phase formation, has developed into a far larger quest of discovery of new compositionally complex materials with improved properties or desired combinations of properties. Although most studies to date have focused on the structural behavior of metallic alloys, the field has now branched out to properties other than mechanical properties, such as magnetism and catalysis [40, 41], and to other classes of materials, especially ceramics. The exponential increase in the literature on this general topic has been remarkable and has certainly led to several success stories - the exceptional cryogenic mechanical properties and fracture toughness of specific CrCoNi-based HEAs being one notable example - but akin to the “explosion” in research interest in intermetallics in the 1970s to 1990s,

to date few industrially relevant materials have been developed and are used in service. This by no means is meant to downplay the new discoveries, interesting properties, conceptual advances, and scientific understanding of intermetallics that came out of that intense period of research activity, including anomalous strength increase with increase in temperature due to cross-slip pinning in Ni₃Al and thermal vacancies in FeAl, and moisture-induced embrittlement of what should otherwise have been ductile FeAl and Ni₃Al, to cite but a few prominent examples. Rather, it highlights the lag time between material discovery and actual adoption in industry, which is generally measured in decades rather than years, certainly for structural materials. This should not dampen the enthusiasm that high-entropy materials have created throughout the metallurgical and materials sciences communities, not simply in the discovery of new alloys and ceramics but also in the identification of new physics associated with the mechanical properties of concentrated solid solutions, local ordering in disordered structures, processing science and microstructure design for the creation of desired properties and combinations of properties. We trust with the following six brief review articles that the reader will be able to appreciate the depth and breadth of this exciting new field of research.

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Ethics declaration

Conflicts of interest

The authors declare that they have no conflict of interest.

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