

High-entropy alloys: A rich history and a challenge for the future

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A selected history of the high-entropy alloy (HEA) field is provided, emphasizing the first publication in 1788 and the sometimes hidden years in the modern era before the landmark publications in 2004. A summary evaluation of the four core effects shows that a clear consensus has not yet been reached, partly because the field has expanded so much since the core effects were first posed and partly because the core effect wording is imprecise and open to interpretation. A selected list of important HEA milestones achieved since 2004, together with a continually growing number of publications, draws an optimistic picture for the future of the field. However, the HEA field is missing one essential milestone—the first major application of economic and societal impact. We recognize that a new scientific idea has a finite lifetime, and so we introduce a keen sense of urgency to accelerate future HEA studies toward new scientific advances and the first application. Two strategies are suggested to converge more rapidly on these goals: implementing the growing field of autonomous R&D, and focusing on uncontested composition space. Initial steps are already being taken on the first strategy; continued work is encouraged. Four new ideas are introduced to develop the second strategy. First, complex concentrated alloys (CCAs) are already accepted to have as few as three principal elements, and we expand their composition range to allow a maximum concentration of 75 at. percent. Next, we recognize that a small number of extraordinary commercial alloys already satisfy this expanded definition; these are called commercial CCAs. Compositional domains already well-populated by commercial CCAs are deemed to have a relatively low probability of major scientific or practical advances. The third new idea is that there is a hidden HEA penalty—the larger is the number of principal elements, the harder it is to produce extreme properties, especially for properties that follow a rule of mixtures such as stiffness, density, and even melting temperature. This gives a dynamic tension, and results over the past 20 years are hinting that the most attractive properties may often be found in alloys with as few as three principal elements. Finally, future studies are encouraged to supplement the initial approach to define HEA families based on chemically similar elements, as has been done for the 3d transition-metal, refractory metal, noble metal, and lanthanide metal HEAs. Future HEAs are encouraged to more assertively explore novel palettes of elements drawn from two or more of the existing alloy families, and from a more diverse range of elemental behaviors. All three HEA founders questioned unspoken assumptions and artificial boundaries to open new realms of exploration. We learn from these examples and continue to push beyond the new borders of thought that have emerged within the HEA field.

Introduction

This article takes a look at the history of high-entropy alloys (HEAs) for new insights, which are then used to peer into the future. The lookback includes summaries of major HEA milestones and the current status regarding the HEA “core effects.” A great deal of scientific progress has been made; nevertheless, the first major HEA application is still missing, introducing a sense of urgency for future work. Strategies and approaches to accelerate the discovery and development of HEAs with an

improved likelihood of commercial transitions are introduced and discussed at the end of this article.

New concepts are introduced to support these strategies. In particular, we expand the maximum concentrations in complex concentrated alloys (CCAs) beyond 35 at. percent. This broadened allowance illustrates an overlap with a small number of well-known commercial alloys that have extraordinary properties—we label such alloys as commercial CCAs. Extending the range of CCA compositions, and comparing them directly

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with commercial CCAs, has important implications that are also discussed at the end of this article.

A selected history of HEAs

It is commonly believed that the high-entropy field is 20 years old. However, the history stretches back more than 40 years in the modern era, and a surprising 237 years overall. The very early history reveals a work that was revolutionary in scope and execution, and the more recent founders demonstrated the exemplary ideals of innovative thought, courage, and persistence. A brief description of the history supporting the HEA field is given to highlight insights from this unique historical progression.

The first HEA publication—in 1788

We are used to an HEA history starting with the publications of professors B. Cantor and J.-W. Yeh in 2004. Here, we start in the late 1700s with a Prussian scientist, Franz Karl Achard (Figure 1a). He is most famous for establishing a commercial process to extract pure sugar from domestically grown sugar beets.¹ He also studied meteorology, electricity, and gravity, and he acclimatized tobacco to Germany. Franz Karl Achard studied metallurgy only briefly, and in 1788, at the age of 35, he published the book, *Recherches sur les Propriétés des Alliages Métalliques (Research on the Properties of Metallic Alloys)* (Figure 1b–c).² It is a remarkable piece of work. In 314 pages, Achard evaluated 894 distinct, as-cast alloys drawn from a pallet of 11 elements (Cu, Fe, Pb, Sn, Zn, Bi, Sb, As, Ag, Co, Pt). Binary, ternary, and quaternary alloys were evaluated in equimolar compositions, as well as several additional molar fraction ratios. Alloys with five to seven elements were studied only in equimolar ratios. The properties measured for each alloy included density, hardness, strength, ductility, abrasion, and corrosion. As a major finding, Achard concluded that alloy properties were very different from those of their constituent elements, and the properties were unpredictable. To put this work in perspective, Achard studied more alloys in his single study than the entire international HEA community studied in the 13 years following the seminal papers by Cantor and Yeh in 2004.

Achard's landmark study was forgotten for 175 years. It was reintroduced in 1963 by Cyril Stanley Smith as one of the four most important studies in the history of metallurgy, with extraordinary praise, "I know of no other book in the history of metallurgy that can compare with this for systematic study of so many properties of so many alloys."³ The book was then apparently forgotten once again. It was revived a second time, 51 years later, in the first modern book on HEAs.⁴ Now, 237 years after it was first published, Achard's book is freely available to the public as a digital download.² Achard's then futuristic and seminal contribution is recounted here to appropriately honor this early historical work.

Modern HEAs: The hidden foundations

Once again, before starting with the Cantor and Yeh publications in 2004, we go back in time to better appreciate how scientific history is made. Brian Cantor first had the inspiration to explore alloys near the center of multicomponent phase diagrams in the late 1970s.⁵ A Bachelor of Science report followed this insight in 1981 (Figure 2), introducing the concepts of multicomponent alloys, equiatomic alloys, and the vastness of compositional space. For a long time, this held the record for the largest number of principal elements (20). Out of 1,048,555 different permutations of binary, ternary, quaternary ... 20 component systems that could be drawn from his mega-principal element alloy, only a few phases were produced, and the dominant one led to what we now know as the Cantor alloy. Unsuccessful in finding funding for his new idea, and enduring criticism from his peers, Cantor nevertheless persisted in his pursuit. He recruited other students to repeat the work at a level of rigor needed for peer-reviewed publication. The first modern HEA publication in the open literature introduced equiatomic alloy substitution to explore new bulk metallic glasses,⁶ and his landmark paper introducing the Cantor alloy, CoCrFeMnNi, was published two years later.⁷ Cantor pursued a major career change soon afterward, and the main narrative in the HEA history shifts halfway around the world.

More than 10 years after Cantor's first inspiration, Jien-Wei Yeh had an independent vision for a new class of alloys. His motivation was different; he believed that increased configurational entropy could play an essential role in stabilizing single-phase solid-solution alloys. This led him to the same domain explored by Cantor—the dark and unexplored inner regions of multicomponent phase diagrams. He found a similar reception to his idea—disdain from colleagues and lack of financial support—and he had the same reaction, to trust his curiosity and to explore this new domain nevertheless. The first publication was a Master of Science thesis in 1996 (Figure 2), and in 2004, he released a remarkable volley of five papers introducing configurational entropy as a property worth considering in alloy design.^{9–13}

A discussion of the influential papers that form the initial foundation of the HEA field must also include the evocative thought piece by S. Ranganathan, "Alloyed pleasures: Multimetallurgical cocktails."¹⁴ Three distinctive materials are described in this paper: bulk metallic glasses, superelastic and superplastic "gum" metals, and nanocrystalline HEAs. In this paper, HEAs are hailed as "a new frontier in metallurgy."

Together, these papers constitute the modern foundation of the HEA field. While the six papers from Cantor and Yeh in 2004 certainly mark a beginning, we should not lose sight of the fact that they are also the culmination of a long period of insight, courage, and persistence. These are rare and valuable traits in the scientific enterprise and are applauded here. Much more detail on the founding of the HEA field, including brief introductions to the main contributors, is provided elsewhere.^{5,15}

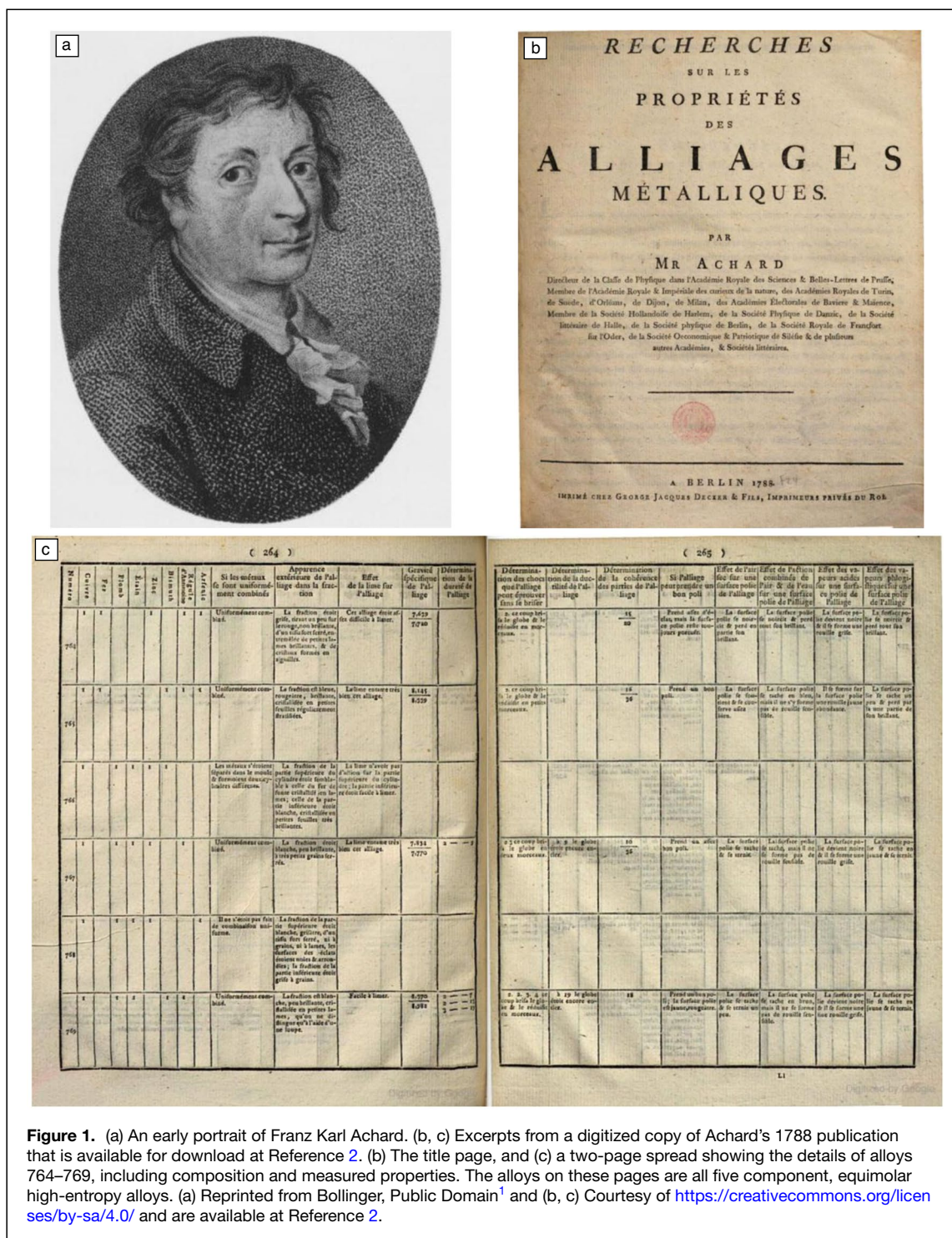


Figure 1. (a) An early portrait of Franz Karl Achard. (b, c) Excerpts from a digitized copy of Achard's 1788 publication that is available for download at Reference 2. (b) The title page, and (c) a two-page spread showing the details of alloys 764–769, including composition and measured properties. The alloys on these pages are all five component, equimolar high-entropy alloys. (a) Reprinted from Bollinger, Public Domain¹ and (b, c) Courtesy of <https://creativecommons.org/licenses/by-sa/4.0/> and are available at Reference 2.

Modern HEAs: Major milestones and events

A selected set of important events in the progression of the HEA field is illustrated in Figure 2. In the exuberance following the 2004 publications, the field focused on HEAs that emphasized the five elements of the Cantor alloy (Co–Cr–Fe–Mn–Ni), along with other 3d transition-metal elements, such as Cu, Ti, and V, plus the compound-forming

element, Al. In “a new frontier of metallurgy” that offered hundreds of billions of new alloy possibilities, it was a matter of time before distinctively new and different alloy families were conceived and studied. The first such expedition introduced HEAs based on refractory elements in 2010.¹⁶ Similar to the experiences of Cantor and Yeh, this idea was conceived several years earlier; it was rejected for funding

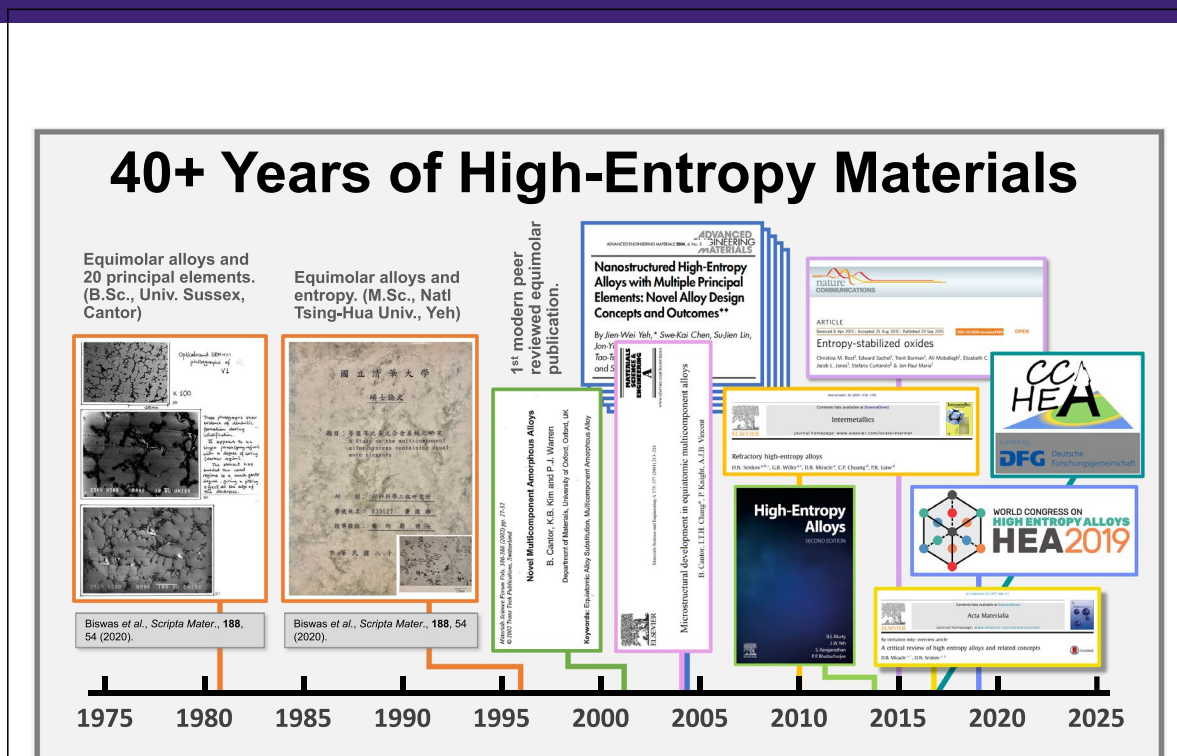
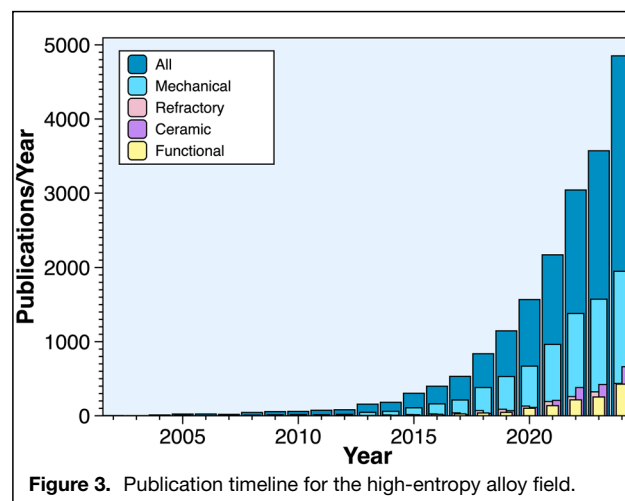


Figure 2. Selected milestones in the modern history of the high-entropy alloy (HEA) field. Images from the early BSc and MSc theses are reprinted under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>) and are available in Reference 8.

twice, and the work proceeded through belief in the concept and a small amount of seed funding (pp. 124–128 in Reference 5). This was quickly followed by a second paper reporting exceptional strength at temperatures as high as 1600°C,¹⁷ firmly launching refractory HEAs (RHEAs) as a major new direction in HEAs (Figure 3). Inspired by this successful example, new HEA families were subsequently introduced based on light elements,^{18–20} lanthanide elements to produce hexagonal close-packed (hcp) solid-solution alloys,²¹ high-entropy brasses and bronzes,²² and noble metals.²³ Although ceramic phases were included in some of the initial HEA publications,^{9,10} monolithic ceramic high-entropy materials were first introduced as a dedicated area in 2015.²⁴ Similar to RHEAs, ceramic high-entropy materials are now a distinct and sizable category of HEAs (Figure 3). A recent review of ceramic high-entropy materials boasts more than 750 citations.²⁵

The first major book was published in 2014⁴ with a 2nd edition in 2019.¹⁵ HEA overview articles began to appear at the same time as the books.^{26–29} The 2017 critical analysis included in Figure 2²⁹ was published late enough to capture the major expansion driven by the new HEA families previously mentioned, and it was published early enough to still give a detailed and comprehensive evaluation of the entire field. Just under 1500 papers were published by the time this paper was published, by 2019 the number had reached nearly 4000, and more than 19,000 papers were available by the end of 2024, making a current review of the entire field a daunting task!



Another major milestone heralding the growing interest in a new technology is the funding of major, national initiatives. Such efforts have been established in Taiwan,³⁰ Germany,³¹ Japan,³² South Korea,³³ and the United States.^{34,35} Begun in 2017, the German Research Foundation Priority Programme, Compositionally Complex Alloys—High-Entropy Alloys, is shown in Figure 2 as one of the early, high-profile national initiatives. Symposia and workshops exploring recent advances in HEAs have proliferated. As a notable milestone, the 2019 World Congress on HEAs was the first recurring, standalone conference dedicated to HEAs and organized by a major, international professional society (The Minerals, Metals & Materials Society).

The publication history for HEAs and CCAs is shown in Figure 3. After a decade of incubation, significant annual growth has occurred. A clear bias is shown for mechanical properties, with refractory, ceramic, and functional materials each representing major areas of research.

Other major indicators may exist as milestones along the path to maturity and impact of a new technology. Those shown in Figure 2 are by no means exclusive, and the reader may add their personal favorite milestones as well. One essential accomplishment is missing in this timeline—the first major HEA application. This will be discussed further.

What have we learned?

What makes HEAs remarkable and worth continued effort, and how can this 20-year perspective be used to guide future research? Here, we attempt a brief summary of the current understanding of the defining characteristics of this field.

The four core effects: Early HEA postulates

In 2006, Yeh introduced four “core effects,”³⁶ which have had a seminal influence on studies around the world. The core effects are an early attempt to describe common HEA features by qualitative observations made on alloys in the first two years. The wording is not sufficiently precise for them to be testable hypotheses, and the core effects may rather be considered as postulates—statements that are suggested to be true to stimulate further research and discussion. This is the spirit with which the core effects are considered next, where we reevaluate their validity in the light of the many studies that have been published since 2006. The original wordings of the core effects are reproduced below from Reference 36 as a reminder of the initial meanings.

The entropy effect states that, “HEAs composed of chemically compatible elements are composed of only a few solid solution phases or even one single phase, which is attributed to their high mixing entropies.” The phrase, “chemically compatible elements” is an essential qualifier that is often overlooked. Consistent with classical thermodynamics, configurational entropy dominates only when other contributions to the Gibbs free energy, especially the formation enthalpies of intermetallic phases, are sufficiently small. In this sense, HEAs could equivalently be considered as low enthalpy alloys.²⁹ It seems to be an unstated assumption in the HEA field that configurational entropy can be independently adjusted by controlling alloy composition. However, alloy composition (including the number of principal elements, N) also has a direct influence on enthalpy. In the entropy–enthalpy tug-of-war, a large N statistically favors enthalpy, since the maximum possible (ideal) configurational entropy increases rather slowly, as $\ln(N)$, while the probability that at least one pair of principal elements has a large interaction enthalpy increases with a much faster, combinatorial dependence. A CALPHAD evaluation of more than 111,000 equimolar alloys with $3 \leq N \leq 6$ supports this view, predicting that 16% of the ternary alloys are solid solutions at 600°C, while less than 2% of the $N=6$ alloys are solid solutions at 600°C.³⁷

Nevertheless, configurational entropy dominates in several extensive, multicomponent, single-phase, solid-solution phase fields consisting of “chemically compatible elements.” The disordered, face-centered cubic (fcc), single-phase field produced by the 3d transition-metal elements, exemplified by the Cantor alloy, is the first and best known, followed by the disordered body-centered-cubic (bcc) phase field bounded by the refractory elements, and home to the two well-known Senkov alloys, NbMoTaVW and HfNbTaTiZr. There are other extensive, multi-principal element, single-phase, solid-solution phase fields as well. These are not restricted to disordered alloys alone, but are also produced by intermetallic phases, such as the B2 structure ($cP2$, CsCl prototype), and ceramic phases, such as the B1 rock-salt structure ($cF8$, NaCl prototype). These phase fields all show that a product phase with a single-crystal structure can be created by combining several different crystal structure inputs. Stated differently, a parent phase structure does not have to match the intended product crystal structure. This was known before, for example, austenitic stainless steels have principal elements with both fcc and bcc crystal structures. However, the last 20 years have shown that this effect extends much farther than previously thought. We can apply a structure in–structure out analysis to predict the probability of forming a particular crystal structure in a large number of similar alloys (e.g., disordered fcc solid solutions) based on the frequency with which that same crystal structure appears in the input phases.²⁹ However, we still do not have a simple, robust model to predict the product phase from the input phases for individual alloys and materials. There is still work to be done to understand this core effect.

The lattice distortion core effect says, “... a multi-element lattice is highly distorted because ... atomic sizes are all different from one another. The distortion ... influences the ... behavior of materials, by the so called ‘lattice distortion effects’.” After 20 years, data are still sparse and are generally limited to a handful of fcc and bcc single-phase, solid-solution alloys. Nevertheless, there is a growing consensus that lattice distortions in fcc and bcc solid-solution alloys are important and can influence the phase stability.^{29,38,39} Atomic size effects are also clearly important for strength. A volume misfit term is included in physics-based equations for fcc ($\Delta \bar{V}_n$ in Reference 40) and bcc (ΔV_i in Reference 41) metals, and a direct relationship has been shown between measured mean-squared atomic displacement and strength.⁴² However, the influence of local distortions on other properties is not yet firmly established. Further, the exact meaning of “highly distorted” is uncertain. Measurable differences are seen within a given family of alloys, and larger differences are seen between different alloy families. Data are very limited and primarily explore 3d transition-metal HEAs and CCAs, with a small number of refractory HEAs and metallic glasses. More importantly, distortions in commercial alloys have not been measured for comparison. Such a comparison is recommended to better distinguish between alloys having lightly, moderately or highly distorted structures. Additional

details on atomic distortions in alloys are provided in References 5, 43, and 44.

The third core effect addresses diffusion: "... the cooperative diffusion of elements ..., in combination with the lattice distortion which hinders atomic movement, will limit the effective diffusion rate in HEAs." Analytical solutions of diffusion equations for concentrated alloys with ≥ 3 diffusing elements are vastly more complex than for dilute, binary alloys.^{5,45,46} Practical solution of these equations from measured concentration gradients requires a number of assumptions, approximations, and simplifications, so the results of comparisons lose precision. Two additional considerations complicate a clear evaluation of this core effect. The experimental uncertainties associated with diffusion measurements in general, and for HEAs in particular, are not usually reported, but earlier work suggests that a standard deviation between one-half and one order of magnitude is common for metallic elements and binary alloys.⁴⁷ Modern measurements may give more precise results, but the propagation of errors through the complex thread of assumptions and analyses required for HEAs work against this trend. As a second complication, the uncertain meaning of the term "sluggish" that is often used to describe this core effect may contribute to some of the variability in interpretation. With this background, more than two dozen experimental and computational studies fail to generate a convincing consensus, with some claiming support and others opposing the sluggish diffusion postulate. The diffusion rate is often comparable to simpler alloys when evaluated at equivalent absolute temperature, and diffusion is typically one-half to one order of magnitude lower than simpler alloys when the comparison is made at an equivalent homologous temperature. To use more precise wording, diffusion is often slower than average when compared against simpler alloys with similar elements, and primarily only when normalizing by melting temperature. It is not slower than any other alloy with the same crystal structure, and it is not unprecedented, anomalous or exceptional.⁴⁸ More detailed analyses are given in References 5 and 49–51.

The "cocktail effect" is the last core effect. Based on the paper, "Alloyed pleasures: Multimetallurgical cocktails,"¹⁴ it is often thought to suggest synergistic and unexpected properties from a mixture of many elements. The actual wording is much simpler: "HEAs can be viewed as an atomic-scale composite. For example, if more light elements are used, the overall density will be reduced." This is essentially a "rule-of-mixtures" effect. This has been solidly demonstrated for some properties (density, elasticity) but is clearly inaccurate for others, especially structure-sensitive properties such as ductility and strength. There is an unintended and often overlooked consequence of averaging properties from a fixed palette of elements—the range in available properties shrinks as the number of elements increases.^{52,53} Stated differently, extreme properties are best achieved in alloys with fewer principal elements when the rule of mixtures applies. In the limit, the most extreme values of density, modulus, T_m , or cost will usually

be found for elements. Sustainability can also become more challenging as the palette of elements grows. This places a dynamic tension on the number of elements when optimizing multiple properties.

In summary, after hundreds of studies, none of the core effects have been disproven, and neither have they been solidly and consistently validated. All have both supporting and detracting results. More details are offered in References 5, 29, 43, 44, and 50. Some uncertainty may come from the dramatic expansion of the HEA field since the core effects were first proposed in 2006. Stated simply, given the cosmic extent of complex compositions, it may be unreasonable to expect all HEAs to behave in the same way regarding the core effects. We further propose that a major part of this inconsistency results from the imprecise wording used to describe some of these effects—phrases such as "chemically compatible elements," "highly distorted," and "sluggish diffusion" are hard to evaluate quantitatively. For example, what is sluggish in one frame of reference may be swift in another, and what could seem to be highly distorted in one small data set may be only slightly or moderately distorted when considering a broader range of materials. Quantifying these distinctions may provide a more certain foundation for consensus. We conclude that the four core effects represent an early intuition into important phenomena. Like asking a good question, these postulates have focused research and generated much new knowledge on the unique blend of properties offered by HEAs. However, future studies addressing the topics in the core effects are strongly urged to introduce more quantitative definitions to the vague descriptors. Quantitative comparisons with the characteristics of commercial alloys are also essential to build a scientific consensus on the truly unique features offered by HEAs and CCAs. This idea will be developed later in this article.

The next 20 years—A sense of urgency

We emphasize the motivation of Franz Karl Achard in his seminal study—"it was 'incontestable utility' that prompted him to undertake 'this very laborious research.'"³ After 20 years of intense study by the entire international scientific community, there is still no compelling first application for HEAs. Even more troubling, there is not yet consensus on the most promising class of applications or the most promising family of CCAs to focus future efforts. An idea loses the ability to motivate and inspire the best scientists and engineers when it persistently fails to produce a major advance of scientific, societal or economic impact. As a recent case in point, the bulk metallic glass (BMG) field began in 1988 by the publication of practical metallic glasses with a wide supercooled liquid regime⁵⁴ and was firmly launched in 1993 with the first open publications of practical metallic glasses (not based on Pd) that could be produced in thicknesses ≥ 1 mm.^{55,56} The number of BMG publications grew rapidly, reaching a peak of >2000 papers in 2009 alone (more than the HEA field had achieved after 19 years; see Figure 3). However, a sharp drop

occurred in the following two years, and the publication rate has remained stable ever since. While BMG applications such as golf club heads were well-publicized for a limited time, it is difficult to independently verify a major, lasting commercial application of BMGs. We propose that the lack of an application with significant economic or societal impact is a major contributor to the decline after 2009, along with competition for talent and resources from the growing HEA field. We conclude that an idea will not last forever. Further, major societal issues such as climate change continue to advance, adding to the urgency for future HEA efforts.

If the cosmic breadth of alloy compositions is a strength of the HEA field, it can also be a liability. Efforts can become sufficiently diverse and unfocused to dilute opportunities to develop compelling new materials with major applications. A few focused efforts that include national initiatives in Taiwan,³⁰ Germany,³¹ Japan,³² South Korea,³³ and the United States^{34,35} have sought to develop HEAs for targeted applications, with no major successes. In spite of these failures, we nevertheless firmly believe that materials with an unprecedented balance of properties are still achievable in CCAs and HEAs. However, we also firmly believe that a new perspective is needed to discover and develop compelling new HEAs and CCAs before time runs out.

With this preamble, we ask the question, “what is the future of HEAs?” We develop two answers next, *accelerate* and *focus*.

Accelerate!—Autonomous R&D

It has long been recognized that the development of new materials is a tedious and lengthy process. This has inspired approaches such as design of experiments that minimize the number of tests required to generate needed data, and combinatorial, high-throughput evaluations that accelerate material characterization by parallelizing material synthesis and experimentation. The Materials Genome Initiative sought to double the speed of materials development by adding computational tools and digital data to these earlier capabilities. In the last decade, the convergence of automation and artificial intelligence (AI) has spawned an emergent capability in autonomous R&D. This approach uses AI agents to analyze data, update models, revise hypotheses, and then define and perform subsequent experimentation (both computations and physical measurements) by directing robotic, automated processes for synthesis and characterization.

The fields of biology, chemistry, and pharmaceuticals have been early adopters of this new approach, where an emphasis on solution or vapor chemistry lowers implementation barriers and has enabled “self-driving laboratories.” Inorganic materials—metals and ceramics—offer more substantial challenges, especially when mechanical properties are a primary objective. HEA studies show a strong bias toward mechanical properties (Figure 3), and so these additional, more challenging issues must be addressed if autonomous R&D methods are to accelerate HEA studies. An early overview of autonomous

R&D describes initial methods and exemplar approaches,⁵⁷ and a more recent paper describes the additional, unique challenges offered by inorganic, structural materials.⁵⁸ Two major themes that are developed in the more recent manuscript are briefly summarized here: AI for materials; and rapid, on-demand synthesis. More details on the current state-of-the-art and remaining challenges in the rapid development of bulk, inorganic materials such as HEAs can be found elsewhere, including a detailed description of high-throughput measurements.^{58,59}

AI is already widely used to make suggestions that are supported by vast data sets, but the field of materials science is characterized by relatively sparse data sets. On the other hand, decisions in materials science can be strengthened by physical models. New AI tools are needed that can provide high-quality recommendations by integrating sparse data sets with established physical models. Further, current approaches applying AI to materials selection typically optimize a single objective function,⁵⁷ but materials applications generally require balancing many properties simultaneously. AI methodologies that can rapidly navigate multi-objective optimization are thus also needed. Other areas where AI can accelerate the exploration of materials (including HEAs) are in microstructure classification, data analysis (such as indexing diffraction results), definition of deformation processing parameters, and predictive planning and decision-making in the materials development workflow.

Perhaps the most significant barrier to the vastly accelerated evaluation of HEAs is primary and secondary synthesis. In addition to composition, mechanical properties depend very sensitively on both length scale and microstructure. Test samples for mechanical properties generally require a minimum dimension of about 10 times the grain diameter, or roughly 100–500 μm for commercially produced products. This eliminates thin films as a synthesis method. Laboratory-scale primary synthesis of experimental alloys, exemplified by the arc-melting process, is almost entirely manual and has changed very little in the past century. Additive manufacturing (AM) offers some promise to accelerate primary synthesis, but procuring powder and establishing deposition parameters that can produce defect-free materials remain two substantial bottlenecks. Radically new approaches to dramatically accelerate primary synthesis may incorporate automation, advanced robotics, sensors, and new designs for power sources and synthesis workflow. Once the constituent elements are homogeneously combined in the primary synthesis step, thermomechanical (also called secondary) processing is needed to control the microstructure. While concepts exist to significantly accelerate primary synthesis, the ability to automate and accelerate deformation processing for targeted microstructural control of metallic materials remains a daunting challenge that requires major new ideas and tools.

The acceleration offered by autonomous R&D is essential, but by itself is not enough. Early estimates suggest a 2–3 orders of magnitude acceleration from autonomous research,

but the composition space offered by HEAs and CCAs is many orders of magnitude larger still. Something more is needed. An additional dramatic increase in efficiency can be achieved by limiting the scope of the vast composition space studied to those domains that are most likely to uncover new scientific phenomena and to produce compelling materials with an exceptional balance of properties. Next, we propose two strategies to focus on compositional domains where CCAs and HEAs face little competition from current alloys.

Focus on sparsely populated alloy spaces and unsatisfied applications

With the benefit of 20 years' experience, we offer some thoughts regarding past attempts to identify HEAs with significantly improved properties. This analysis inspires two strategies to concentrate future studies in directions that are most likely to commercialize HEAs and CCAs. First, we recommend exploring composition spaces that are not already densely populated by compositionally similar alloys. Second, we encourage HEA/CCA efforts to emphasize important new applications where the needed materials are not currently available.

Before we can discuss opportunities to explore sparsely populated composition spaces, we must first show that some HEAs compete in already occupied compositional domains. Stated simply, many HEAs, especially those based on the 3d transition-metal elements, compete with well-known, compositionally similar commercial alloys drawn from the same palette of principal elements. Twenty years of effort, including the concerted HEA national initiatives cited earlier, have been unsuccessful in displacing these current materials. We propose that the potential for significantly better materials in alloy spaces already densely populated by compositionally similar commercial alloys is greatly diminished, and that new scientific discoveries and major applications are far more likely to come by focusing on uncontested composition spaces.

To emphasize this important point, we reevaluate the essential, defining compositional characteristics of CCAs. Terms such as multi-principal element alloys (MPEAs) and CCAs are already used to broaden the range of compositions and microstructures beyond those in the standard HEA definition (≥ 5 principal elements with concentrations between 5 and 35 at. percent). Consistent with earlier work,^{29,48} we allow CCAs to have as few as three principal elements, and we further follow accepted convention by defining a principal element as one with a concentration ≥ 5 at. percent. However, we reconsider the maximum allowable concentration in CCAs. Rather than enforcing an upper bound to maintain proximity to the equimolar composition, we look for common maximum compositions that produce an exceptional balance of material properties. For inspiration and guidance, we evaluate the compositions of outstanding commercial alloys with an extraordinary balance of properties. These alloys often have three or more principal elements and differ from the conventional CCA definition only because one element exceeds the 35 at.% threshold. These

remarkable commercial alloys have a maximum concentration that is usually between 40 and 70 at.%, and is sometimes above or below this range. From this analysis, we redefine the maximum acceptable concentration in a CCA to be as high as 75 at. percent. This may surprise some readers. Nevertheless, this is supported by the observation that commercial alloys with ≥ 3 principal elements, and with a maximum concentration between 35 and 75 at.%, always have a balance of properties that are superior to the more recent HEAs and CCAs. We label these alloys as commercial CCAs. They were introduced earlier (see Sect. 5.6 and Sect. 6.2.1.1 in Reference 29) and are described in more detail next.

Several distinct classes of commercial CCAs are already established within the Co–Cr–Fe–Ni quaternary space. Six such CCA families are shown in **Table 1**: austenitic stainless steels (Fe–Cr–Ni); solid-solution Ni–Cr–Fe alloys; solid-solution and age-hardenable Ni–Co–Cr alloys; age-hardened superalloys (Ni–Cr–Co–Al, Ti, Nb); Fe–Co–Cr hard magnetic materials; and the Alnico family of hard magnets (Fe–Ni–Co–Al). Stainless steels and superalloys are well-known commercial CCAs with exceptional properties. Ni–Co–Cr commercial CCAs also deserve special mention, because they represent the so-called “cryogenic steels” that have recently drawn much attention in the HEA community due to the outstanding properties of the ternary equimolar CoCrNi alloy.^{60–62}

The balance of properties achieved by the HEA community in the 3d transition-metal family of alloys does not compete with these established commercial CCAs. Stainless steels equal or exceed the mechanical properties (see Figure 19 in Reference 29) and corrosion resistance (see Figure 16 in Reference 64) of more recent HEAs, and commercial CCAs are more affordable and sustainable by removing Co as a principal element. Commercial Ni–Cr–Fe solid-solution CCAs such as Inconel 600 and Incoloy 800 are significantly stronger than the Cantor alloy (see Figure 19 in Reference 29). Equimolar CoCrNi has strength and fracture toughness that are no better than commercial Ni–Co–Cr CCAs (represented as “cryogenic steels” in Figure 4 of Reference 62), and the commercial alloys are age-hardenable and have better corrosion resistance due to the addition of Mo.^{65–67} Although two national HEA initiatives have sought to replace age-hardened commercial superalloys, reports of success are not apparent in the literature. The story is the same for magnetic materials; both Fe–Cr–Co and Fe–Ni–Co–Al commercial CCAs remain superior to more recent HEAs.^{68,69} Beyond first-tier characteristics, commercial CCAs have also been optimized for many other features that are essential for commercial success, including manufacturability at scale, joinability, repair, cost, and sustainability.

The superior properties of commercial CCAs in the Co–Cr–Fe–Ni realm are hardly surprising after considering the amount of time invested to develop them. The history of stainless steels goes back 200 years, to the early 1800s, when Cr was first added to Fe to resist acid attack.⁷⁰ What is now known as AISI Type 304 austenitic stainless steel was patented in 1912 after almost a century of continuous studies. There are

now five distinct classes of stainless steels with many dozens of standard alloys that have been developed and optimized in the 100+ years since then. Even the relatively new Ni–Co–Cr family of commercial CCAs has a long history. Proprietary work began on this class of alloys in the 1960s, leading to a patent in 1967⁷¹ and the first open publications in 1968 and 1970.^{65,72} This represents more than 60 years of focused effort to refine the composition, microstructure, processing, and properties.

The preceding discussion comes from a privileged perspective afforded by 20 years of HEA research. These perspectives were not apparent at the start of the HEA field for several reasons. First, almost none of the exceptional commercial CCAs meet the composition-based definition of an HEA. As illustrated in Table I, most of these alloys

have only three or four principal elements, and the very few with five concentrated solutes usually have one element that exceeds the conventional 35 at.% limit. Only one commercial HEA is shown in Table I; there are others, but they are rare. Second, the insight that exceptional properties could be achieved in quaternary and even ternary concentrated alloys was not shown in the HEA literature until barely 10 years ago.⁶¹ Since that seminal work, it has become increasingly accepted that ternary and quaternary CCAs may have most or all of the practical benefits offered by HEAs.^{29,48,73} Here, we develop that insight further by expanding the accepted compositions of CCAs and introducing long-established commercial CCAs. Finally, much of the work over the past 200 years that led to many well-known commercial alloys is not available in the open literature. This knowledge is

Table I. Commercial alloys drawn from the Co–Cr–Fe–Ni alloy space.

Alloy	N ^a	Principal Element Concentration (at.%)							Sum of Dilute Element Concentrations ^b
		Fe	Ni	Cr	Co	Mo	Al	Ti	
Fe–Cr–Ni (austenitic stainless steel) ^c									
304	3	67.4	8.4	20.0					4.2 (Mn, Si, C)
316	3	64.8	11.3	18.1					5.8 (Mn, Si, Mo, C)
Ni–Cr–Fe and Fe–Ni–Cr ^{c,d}									
Inconel 600	3	8.1	72.3	16.9					2.7 (Mn, Si, C)
Inconel 625	4	5.3	58.1	24.3		5.5			6.8 (Nb, Si, Co, Al, Mn, Ti, C)
Incoloy 800	3	42.3	30.4	22.2					5.2 (Si, Mn, Al, Ti, C)
Inconel 718	3	19.2	52.2	21.1					7.5 (Nb, Mo, Ti, Al, C)
Ni–Co–Cr (including cryo steels) ^{c,d}									
MP35-N	4		35.4	22.8	33.3	6.2			2.3 (Ti, Fe)
AEREX 350	3		45.5	19.5	25.3				9.7 (Ti, Al, Mo, Ta, Nb, W)
Inconel 617	4		55.2	24.7	12.4	5.5			2.2 (Al)
René 41	4		54.6	21.2	10.8	6.0			7.4 (Ti, Al, C)
Ni–Cr–Co–(Al,Ti) (age-hardened superalloys) ^c									
Nimonic 115	4		51.7	15.7	13.8		10.1		8.7 (Ti, Mo, Fe, C, Zr)
Inconel 100	5		55.5	10.4	13.8		11.0	5.7	3.6 (Mo, V, C)
PWA 1480	4		64.5	11.7	5.2		11.3		7.3 (Ta, Ti, W)
MAR-M247	4		65.9	9.1	9.1		11.9		4.0 (W, Si, C, Mn)
Fe–Cr–Co (permanent magnetic materials) ^c									
Fe–Co–Cr	3	47.1		29.5	21.4				2.0 (Si)
Fe–Co–Cr	3	59.5		28.8	9.4				2.3 (Ti, Mo)
Fe–Ni–Co–Al (permanent magnetic materials) ^c									
Alnico 2	4	50.7	16.6		11.3		19.0		2.4 (Cu)
Alnico 7	5	38.4	16.0		21.3		15.5	5.5	3.3 (Cu)
Alnico 12 ^e	5	31.4	16.3		31.6		11.8	8.9	

Concentrated elements are listed individually, and the concentration sum for dilute elements is given with a list of the included elements.

The most concentrated element is shown in bold text. Compositions are taken from Reference 62.

^aN is the number of principal elements, defined by a concentration ≥ 5 at.% and ≤ 75 at.%.

^bDilute elements, defined by concentrations ≤ 5 at.%, are listed in order of decreasing concentrations.

^cThe principal elements are listed in order of decreasing concentrations.

^dSome of these are solid-solution alloys; others have sufficient concentrations of (Al, Ti, Nb) for age hardening.

^eThis alloy satisfies the composition-based definition of an HEA.

usually hidden in proprietary reports and embedded within industrial processes. As a case in point, the present author has been able to find only a small number of articles published in the past 55 years in the open literature on exemplar alloys for the so-called Ni–Co–Cr “cryo steels.”^{65–67,72,74}

While incremental improvements relative to these commercial CCAs may still be found, it may be like searching the tailings of a spent mine to find an overlooked nugget. We suggest that the most substantial opportunities of new scientific findings and major practical advances will be found in uncontested alloy spaces that have not already been scoured by two centuries of focused alloy development and technological advances.

With this background, we are now ready to introduce opportunities in uncontested CCA composition space. Refractory CCAs (RCCAs) are one such family of alloys. RCCAs have shown promise for significantly improved strength, specific strength, and environmental resistance relative to commercial refractory alloys.⁷⁵ The commercial refractory alloys that RCCAs contend with for applications are often dilute. TZM (Mo–1.0 Ti–0.1 Zr–0.2 C at.%) is arguably the most widely used commercial refractory alloy and has only dilute solutes. Commercial refractory alloys with only one concentrated solute are also very common, but commercial refractory alloys with ≥ 2 concentrated solutes are uncommon.⁷⁶ As a result, significant opportunities exist to explore RCCAs without undue competition from existing alloys.

Copper alloys are another opportunity to explore an uncrowded CCA composition space. Most commercial bronzes are dilute Cu alloys or have just one concentrated solute (Sn, Al, Ni, Zn or Pb). All wrought commercial brasses have only one concentrated solute (Zn), as do Cu–Ni alloys. Only a very small number of Cu alloys have two concentrated solutes, including some aluminum bronzes (Al+Fe or Al+Mn), brazing alloys (P+Ag), and nickel-silvers (Zn+Ni). High-entropy brasses and bronzes (HEBBs) are already showing significant potential to give a commercially important new balance of properties.^{22,77} This includes a high-strength replacement for cartridge brass^{78,79} and a machinable, lead-free alloy for drinking water systems.⁸⁰ HEBBs are perhaps the farthest along the path to commercialization—this may give a fitting historical parallel, since bronze was the first alloy in human history.

Other examples of sparsely populated commercial CCA space may include noble metal CCAs and complex, concentrated functional materials. Especially for catalytic applications, noble metal CCAs compete with commercial materials that are largely elements and dilute alloys. A common alloying approach for many functional materials (iso-structure or iso-electronic substitution) is to alloy with elements that are chemically and structurally similar to the host atom, and to perform such substitutions one sublattice at a time.²⁹ Inspired by 20 years of HEA studies, a more aggressive approach may include alloying elements with different crystal structures than the host, and substitution on more than one sublattice at a time.

The second strategy to focus HEA/CCA development is to target compelling new applications where an enabling material is not currently available. Selected examples include revolutionary energy (fusion), revolutionary transportation (rotating detonation engines), energy harvesting (thermoelectric materials), and energy production (catalysis for hydrogen generation). Each particular application requires careful thought in selecting the palette of candidate elements. For example, materials for fusion reactors must emphasize the need for high-temperature properties, but must exclude elements that can be “activated” (made radioactive) by exposure to the fusion environment. This strategy naturally emphasizes applications with no viable competing current alloys, lowering the barrier to commercialization.

Building future bridges

We foresee two future bridges in the HEA field. The first was introduced in the previous section between “traditional” HEAs and commercial alloys. Moving from HEAs to CCAs by reducing the number of principal elements was the first step. Here, we take a bold, second step by noting that exceptional mechanisms and properties can still be achieved even when expanding the allowed composition range. This forms a bridge between the HEA field, which starts in the middle of multidimensional composition space, and conventional alloys, which start at elemental compositions. Over time, these two different approaches have progressively nudged toward each other. Here, we show that a select number of bridges can be made between these two very different alloying strategies, acknowledging that some of the most stunning commercial alloys are already CCAs. These bridges seem to meet at a point of dynamic tension between competing forces—a larger number of principal elements favor vast diversity but it drives toward an average response, while a smaller number (and nonequimolar ratios) still give adequate complexity while allowing materials to retain extreme properties. This is especially true for properties that follow a rule of mixtures. Making this connection allows us to learn from extensive commercial knowledge in a selected set of exceptional materials. It also focuses new attention on an often ignored composition space that falls between traditional alloys and conventional HEAs, but has already demonstrated the ability to produce unusual scientific phenomena that have been translated to commercial alloys with exceptional properties. These bridges acknowledge the creativity, insight, and capability represented by decades or centuries of conventional alloy development and inspire future HEA/CCA studies.

The second bridge spans different CCA families. Only a small number of HEA families have been explored to date, and most seem to follow an unconscious and unspoken bias toward chemically compatible elements. Consider HEAs based on 3d transition metals, refractory elements, precious metals, and the lanthanide elements. In each case, the constituent elements are sufficiently similar to be recognized as members of the same elemental type. The light metal CCA family is an exception,

where the palette includes such chemically incongruous elements as Li, Be, Sc, Mg, Al, and Ca.

In the future, we project that HEAs and CCAs will assertively combine elements that may represent two or more of these early “chemically compatible” alloy families. Some examples currently exist where elements are chosen from more than one HEA family, such as CoCrFeNiPd and AlCo-CrCuFeMoNi. However, these crossover alloys are rare, and they seem rather cautious, often adding only one out-of-place element per alloy. The present paper calls for a more systematic and bold approach to blending elements from different HEA families. Obvious first bridges may be made between refractory, 3d transition-metal, and noble metal elements. Farther in the future, as computational methods and autonomous R&D capabilities become established, HEAs and CCAs may be devised from more adventurous groupings of elements. This new approach is proposed to supplement, not replace, a robust exploration of alloy families built on elements with common features. These bold expeditions into truly vast and unknown composition spaces, using unexpected elemental combinations, will allow us to finally realize the full potential offered by the ideas of Achard, Cantor, and Yeh.

Summary remarks

HEAs have an exceptionally rich history that extends over 200 years. The field continues to show impressive growth—the number of publications continues to increase year by year, and many major milestones have been achieved. After 20 years, the four core effects have neither been universally verified, nor have they been disproven, and they continue to motivate basic studies probing HEA stability and kinetics. As a counterpoint to these positive trends, there is still no clear prospect for a first application of major economic or societal impact. We introduce an urgency to achieve this essential milestone, and we propose several strategies to accelerate progress toward this goal. Autonomous R&D of HEAs and CCAs is the first recommendation, enabling an acceleration of up to three orders of magnitude compared to conventional alloy exploration and development methods. As a second strategy, we advocate focusing on a limited subset of vast, multidimensional composition space that is most likely to yield new scientific discoveries and commercially relevant alloys. We suggest that important advances are far less likely in compositional regimes that are already densely populated by longstanding commercial CCAs such as stainless steels, cryogenic steels, age-hardenable superalloys, and hard magnetic materials. Instead, several established HEA/CCA families are identified that do not compete with a significant number of compositionally similar commercial alloys and that may provide a better chance of achieving a new balance of exceptional properties. As a second approach to emphasize unexplored composition spaces, we also recommend targeting HEA/CCA development to achieve properties needed for breakthrough applications for which adequate materials do not yet exist. Finally, we foresee that the future will break beyond the present unspoken barrier

of chemically compatible elements that seem to be the basis for most current HEA and CCA families. Bold expeditions into untouched compositional spaces will draw on the exemplary scientific and technical standards set by Achard, Cantor, and Yeh, allowing the full potential of the HEA concept to be developed. The possibility for new scientific advances and for developing commercially important HEAs and CCAs still remains strong.

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Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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