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ROADMAP

2026 Roadmap on compositionally complex and high entropy materials for energy applications

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Abstract

In the study of complex systems, the intricate interdependence among individual components leads to emergent properties that cannot be solely attributed to the properties of the components themselves. This principle is central to compositionally complex materials (CCMs), where interactions between different elements introduced into the structure result in unprecedented material properties. The emergence of high-entropy materials (HEMs) in 2004 further increased complexity by introducing high configurational entropy (S_{config}), which can contribute to stabilizing single-phase solid solutions by counterbalancing enthalpic driving forces for phase separation. HEMs and CCMs represent an emerging family of materials where multiple principal elements occupy equivalent crystallographic sites. This atomic architecture gives rise to extraordinary properties such as tailorable electronic structures, lattice distortion effects, and synergistic interactions, with their vast combinatorial design space enabling the tuning of these effects across a wide range of compositions. Although the field is still in its infancy, early discoveries highlight their disruptive potential, particularly in energy technologies where robustness and durability are critical. Their exceptional

thermal stability, corrosion resistance, and electro-chemo-mechanical durability position HEMs and CCMs as game-changers for applications demanding resilience under harsh operating conditions, such as batteries, fuel cells, and hydrogen storage systems. Beyond performance advantages, CCMs challenge traditional materials discovery frameworks. Their extensive design space makes conventional trial-and-error approaches impractical, creating an ideal platform for deploying AI-driven high-throughput computational screening, multiscale modeling, and autonomous experimental workflows. This convergence of complexity and innovation offers unprecedented opportunities to accelerate the identification of next-generation energy materials. This roadmap compiles insights from leading experts in the field of CCMs across key energy domains, including electrochemical storage, catalysis, thermoelectrics, and turbomachinery. Their contributions critically assess the current state of this material family, highlighting unresolved scientific challenges, technological barriers, and the key advancements needed to move beyond the current state-of-the-art. Special emphasis is placed on combinatorial synthesis and high-throughput approaches and their potential to trigger exponential development of this emerging family of materials. Focused on energy applications, this roadmap provides a comprehensive overview of a time-critical topic, emphasizing the need for material innovation and joint efforts from academia and industry.

Contents

1. Introduction to the roadmap

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The energy paradigm is undergoing a significant transformation, with various conversion technologies gradually replacing the dominant fossil fuels with alternative clean-energy sources. These advanced solutions are unlocking new energy vectors, such as hydrogen and synthetic fuels, while facilitating the direct use of renewables for power generation through wind turbines and fuel cells. Additionally, the storage of renewable electricity is being revolutionized by electrochemical devices like batteries and supercapacitors. Despite their promise, these technologies face substantial materials-related challenges, including limited performance, operational durability, and the finite availability of critical elements. To address these issues and meet the evolving demands of energy systems, continuous innovation in materials science is essential. This ongoing advancement is paving the way toward a sustainable and resilient energy future.

Within this context, a new class of materials designed with a high configurational entropy is emerging with remarkable potential across a broad spectrum of energy applications [1–5]. The so-called high entropy materials (HEMs), defined by their equimolar or near-equimolar composition of five or more principal elements, exhibit a high degree of atomic disorder. This unique structure results in exceptional properties, such as enhanced mechanical strength, superior thermal stability, and remarkable resistance to corrosion and oxidation [5–7]. The synergy of improved thermo-chemo-mechanical stability and high performance positions HEMs as game-changers in energy technologies, where long-term durability and endurance to cycling stresses are cornerstones. The advantages of HEMs are not limited to strictly equimolar compositions. This has broadened scientific interest to include compositionally complex materials (CCMs), i.e. materials composed of multiple principal elements in non-stoichiometric ratios [7, 8]. Indeed, the exploration of CMMs and HEMs represents a natural evolution of traditional research in complex energy materials, such as those used in solid oxide cells (SOCs), batteries, or catalytic systems, which have been optimized for decades to become the state-of-the-art materials, e.g. quaternary oxides like $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-x}$ for solid oxide fuel cells (SOFCs) [9, 10].

The virtually limitless compositional possibilities of HEMs and CMMs present significant challenges in identifying optimal formulations. However, integrating existing physical understanding of the effects of multi-principal elements on the performance of energy materials with advanced tools for materials exploration, such as high-throughput experimentation, modeling, and machine learning (ML), is expected to lead to a significant expansion of the field, with potentially transformative impact on diverse energy technologies [5, 8, 11]. Moreover, the inherent complexity of HEMs and CMMs makes them ideal candidates for the development of specialized ML and Bayesian optimization techniques. These techniques are crucial for efficiently navigating the vast compositional spaces and identifying promising material combinations.

Current synthesis routes for high-entropy materials span solid-state/mechanochemical routes, which are scalable and yield high-crystallinity phases but provide limited control over particle size, morphology, and defect populations; wet-chemical solution processes (co-precipitation, sol–gel, combustion), which enable fine control of homogeneity, nanoscale architecture, and surface area but can suffer from precursor sensitivity and challenges in maintaining single-phase stability at scale; gas-phase and aerosol-assisted techniques support continuous production and controllable porosity but require specialized equipment and may struggle with precise stoichiometric retention. Thin-film deposition methods (e.g. sputtering, pulsed-laser deposition) allow atomic-level tailoring of composition, strain, and interfaces but have high operational costs and are less suited for bulk material output. Collectively, these approaches show that synthesis is a primary lever for directing structure-function relationships, and future progress is expected to come from integrated high-throughput, data-driven, and automated synthesis platforms that accelerate exploration of extensive compositional spaces, enable metastable phase access, and unify design-to-manufacture workflows for increasingly complex materials [12, 13]. Consequently, the study of HEMs and CMMs not only pushes the boundaries of materials science but also serves as a model system for pioneering data-driven approaches in materials discovery. This synergy between advanced computational methods and materials science promises breakthroughs that could redefine the future of energy.

Finally, a notable advantage of CCMs lies in their remarkable compositional flexibility, which opens up opportunities to substitute critical raw materials with less sensitive or more abundant elements. This adaptability not only helps mitigate supply risks but also enables a more diversified use of elemental

resources. However, this multi-element composition also introduces a significant challenge: recycling CCMs becomes considerably more complex. As a result, dedicated research and innovative recycling strategies are essential to effectively manage and recover valuable materials from these advanced compounds.

In this roadmap, the rapidly evolving field of HEMs and CCMs for energy applications is covered, focusing on cases where compositional complexity yields tangible performance advantages. Although HEMs represent a relatively young discipline, their unique properties, arising from the interplay of multiple principal elements, have already led to significant breakthroughs in energy-related technologies [14–16]. This roadmap synthesizes the growing body of impactful results, with particular attention to the performance and long-term stability of CCMs under demanding operational conditions, where their resilience and adaptability offer clear benefits for energy systems.

The document opens with a comprehensive introduction to the use of HEMs and CCMs in energy, establishing the conceptual foundation for the subsequent chapters. It then explores the latest materials discovery methodologies, including computational modeling and high-throughput experimental techniques, which enable the prediction of stability and performance across vast compositional landscapes. These tools accelerate the identification and optimization of promising HEMs prior to synthesis and support the systematic exploration that this class of materials requires. Following this, the roadmap examines the application of CCMs in various energy technologies. It first addresses energy storage, where high-entropy oxides (HEOs) and alloys have demonstrated exceptional performance as electrode materials in lithium-ion, solid-state, post-lithium, and metal–air batteries, offering enhanced capacity, cycling stability, and durability compared to traditional materials. The discussion then turns to the use of HEMs and CCMs in SOCs, highlighting recent advances resulting from cationic engineering of perovskite and fluorite oxide electrodes, which have led to notable improvements in both performance and operational stability. Catalysis is addressed next, with HEMs being explored for several applications such as water splitting and synthetic fuel production. Here, the 'cocktail effect', i.e. the synergistic interaction among diverse elements, creates a rich array of active sites, significantly boosting catalytic efficiency and stability. The roadmap also dedicates sections to hydrogen storage (HS) and thermoelectrics, two pioneering application areas for HEMs. In HS, the multi-elemental nature of HEMs enhances hydrogen absorption and desorption kinetics, increases storage capacity, and ensures robust structural stability, positioning these materials as prime candidates for next-generation HS solutions. In thermoelectrics, HEMs display remarkable thermal stability and electrical conductivity, with their complex compositions allowing for the optimization of the Seebeck coefficient and suppression of thermal conductivity, thereby improving thermoelectric performance. Finally, the roadmap considers the use of HEMs in structural applications. Their often superior mechanical strength and ductility, compared to conventional alloys, can enable the development of highly resilient structural components for demanding environments, such as gas turbine blades and critical infrastructure.

Throughout, the intrinsic features of HEMs, such as lattice distortions, sluggish diffusion, and compositional flexibility, are highlighted as the basis for their exceptional properties and their potential to unlock new directions in energy technologies innovation. By integrating advances in computational discovery, experimental validation, and application-driven research, this roadmap aims to guide researchers and practitioners toward the next generation of sustainable materials for energy technologies.

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2. Overview on compositionally complex and HEMs for energy applications

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Status

CCMs and HEMs have been gaining tremendous attention in energy applications; batteries, fuel cells, supercapacitors, and catalysts, due to their unique element-tunable properties via combinational chemistry, multifunctional nature, availability of different surface-active species, concomitant lattice distortion affecting electronic states, thermal stability, and corrosion resistance (see figure 1). The HEMs are more commonly utilized in energy applications than CCMs because of their single-phase stability and resistance to extreme conditions, such as high temperatures and corrosion [17]. HEMs can be subdivided into high-entropy alloys (HEAs), high-entropy ceramics (HECs), and high entropy polymers (HEPs). Initial reports on multielement materials, particularly HEAs (made up of five or more metallic elements with nearly equal atomic ratios), were published in 2004 [18]. The approach was extended to HECs in 2015 [19]. Following this, a large number of other materials have been introduced to describe this new class of inorganic solids more broadly [20]. In parallel, the concept of high-entropy systems has also been applied to polymers, leading to the introduction of HEPs [21, 22]. Nellaiappan *et al* [23] and Chen *et al* [24] performed the first investigations on the use of HEMs as catalysts for a variety of energy and environmental processes. Since then, a growing variety of HEMs have been reported in various catalytic applications [16, 25]. To date, a large number of HEAs, high entropy (HE) oxides, nitrides, carbides, borides and hydrides are the most widely studied HECs for catalytic applications [20, 26]. However, research on other ceramics, such as phosphides, sulfides, silicides, fluorides, phosphates, oxynitrides, carbonitrides, and borocarbonitrides, is rapidly growing and offers significant potential for various catalytic applications [26]. These materials are expected to provide a fillip for further research on energy using HECs.

HEMs hold promise for a variety of applications because of their unique properties (described above), which can be easily tailored by selecting specific elements and adjusting stoichiometry, altering surface active species, as well as their synergistic effect [16, 27, 28]. Currently, HEMs have received significant attention in the field of catalysis, including electrocatalysis [27, 29, 30], various gas–solid [16, 28] and liquid-solid phase reactions [31], and azo dye degradation reactions [32]. Among various HEMs, HEAs and HEOs have been extensively investigated in catalytic applications. A number of gas–solid reactions have been investigated by researches/scientists that are crucial to environmental applications; CO_2 hydrogenation [33], CO oxidation [16], dry reforming of methane (DRM) [28, 34], and other significant reactions [16, 35]. Recently, supported HEA catalysts have shown excellent activity for the DRM [28]. HEMs have also been explored for energy conversion applications like the oxygen evolution reaction (OER) [16], hydrogen evolution reaction (HER) [1, 36], oxygen reduction reaction (ORR) [1], electrooxidation of Hydrazine [37], and other important electrocatalytic reactions [1, 16]. Water splitting, commonly referred to as the electrolysis of water, has become increasingly recognized as a promising approach for sustainable hydrogen (H_2) production [38–40]. Recently, Zhang *et al* [41] published a review article on HEAs, highlighting their potential as promising catalysts for high-performance water splitting. Water electrolysis is a renewable and environmentally friendly technique that shows great promise for developing a hydrogen-based economy, where hydrogen could replace fossil fuels in various industries, such as energy storage, transportation, and electricity production. Apart from their role in catalysis, HEMs have also been explored in energy storage devices such as batteries [1] and supercapacitors [16, 42].

Current and future challenges

The current and future challenges for compositionally complex and HEMs in energy applications are deeply intertwined with their chemical complexity, structural stability (single vs multiphase), synthesis scalability, as well as sustainability. However, many of these obstacles will probably be surmounted by developments in computational methods, including artificial intelligence (AI) and ML-aided design, devising novel processing strategies, and material design, opening the door for the incorporation of HEMs into a variety of energy devices, including batteries, superconductors, and catalysts. The most important challenge is perceived in taking the lab-based research to industrial-scale application, realizing the long-cherished dream of putting HEMs into applications. The wider use of these innovative materials in the energy sector will depend on striking a balance between cost savings, durability, scaling up, and

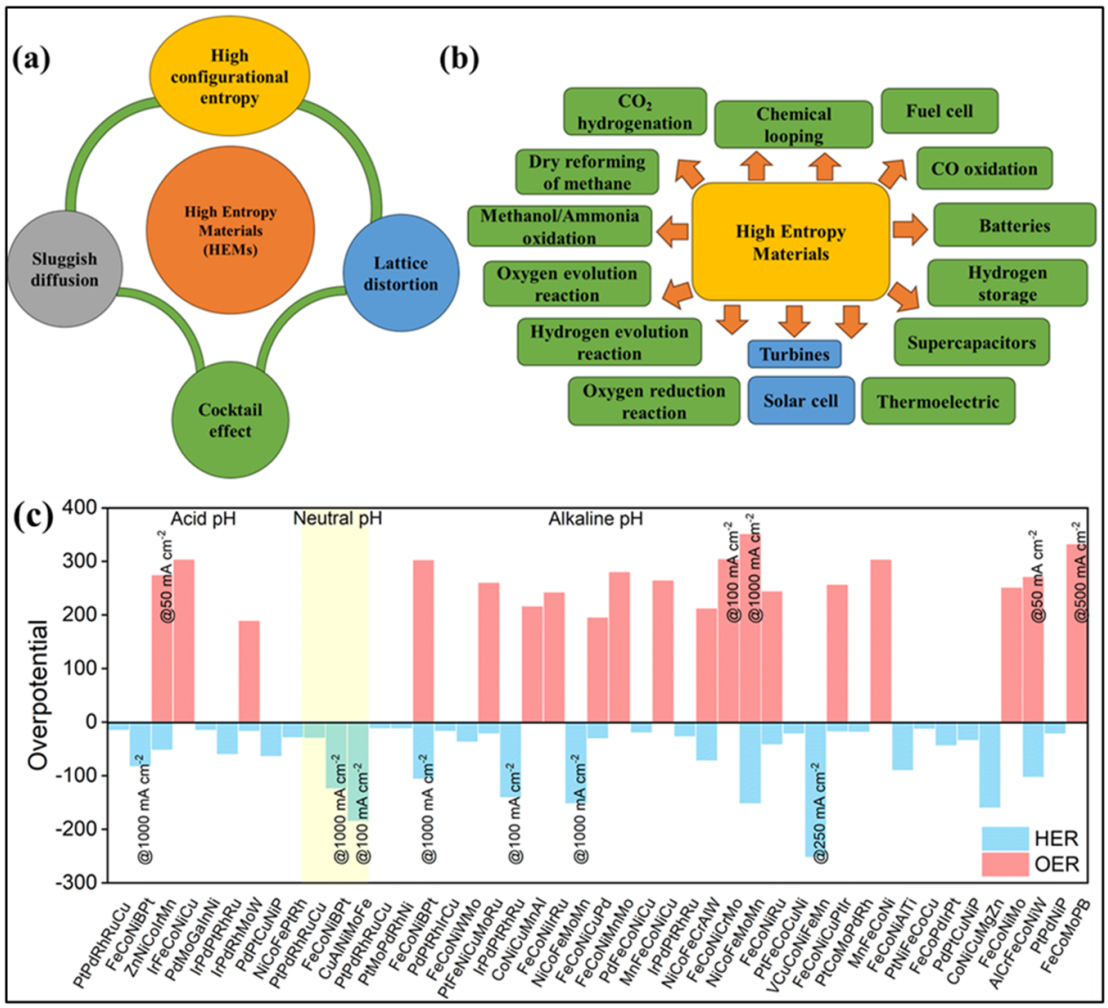

Figure 1. (a) Four core effects of HEMs. (b) Some important applications of HEMs for energy applications. (c) Comprehensive plots of catalytic activity of HEA catalysts, Reproduced from [41] with permission from the Royal Society of Chemistry, showing a comparison of activity (overpotential) in acid (0.5 M H_2SO_4), neutral (1 M PBS), and alkaline (1 M KOH) electrolytes.

environmental impact while maintaining performance improvements, which can be achieved by material scientists working with scientists and engineers from other fields.

Advances in science and technology to meet challenges

High-entropy materials have significantly contributed to addressing crucial challenges in energy applications by enhancing stability, durability, and sustainability. Computational tools, such as ML and density functional theory (DFT), speed up the design of HEMs with reasonable characteristics, including high conductivity and thermal stability, minimizing the requirement for extensive experiments. Advanced manufacturing techniques, such as 3D printing (additive manufacturing) and nanostructuring, ensure precise control over microstructures, while cost-effective synthesis processes reduce production costs. High-resolution characterization methods, such as high-resolution transmission electron microscopy, x-ray photoelectron spectroscopy (XPS), and synchrotron x-ray diffraction, can provide detailed insights into atomic structures, allowing fast design and development and further enhancing HEMs' performance. Sustainability efforts need to emphasize the use of earth-abundant, relatively cheap, and eco-friendly materials and recycling methods to minimize environmental impact. Thermodynamic modeling and electrochemical optimization improve the durability of HEMs in applications such as batteries and fuel cells. Their performance is further boosted by improvements in electrolytes and coatings. This multidisciplinary approach is transforming HEMs into efficient, resilient, and sustainable materials, driving advancements in energy technologies. The realization of industrial-scale applications, translating the lab-based research to a technology readiness level of 7–9, in various energy sectors, will hold the key to the success of HEMs. In order to achieve this goal, scientists and engineers from various domains need to work together to use HEMs in machines.

Concluding remarks

In conclusion, HEMs and CCMs exhibit unique properties, making them valuable for energy applications such as batteries, supercapacitors, fuel cells, and catalysts. HEMs, particularly HEAs and HECs, are more widely used due to their single-phase stability and their durability in harsh environments. These materials show promise in catalytic and energy conversion applications, including water splitting, CO_2 reduction, CO_2 conversion into fuels, HS, and electrocatalysis reactions, owing to their customizable compositions and synergistic effects, enhancing performance in energy storage and environmental processes. In the future, many more materials are expected to enter the realm of high-entropy materials due to the increasing understanding of their unique properties and the development of advanced computational and experimental approaches. The applications of HEMs in energy domains will provide a significant boost to this field, making them the real 'game changer'.

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3. Modeling HE and complex materials

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Status

Accurate modeling of HE single-phase and compositionally complex multi-phase materials requires the use of atomistic methods. These are broadly categorized into two classes (supercell and spectral), which utilize representative states—ordered configurations of atoms—for modeling disordered systems. For each class, the representations' properties (thermodynamic, vibrational, elastic, etc) are first calculated using DFT, and then an appropriate averaging scheme (taking into account any degeneracy of the representations) is used to compute the overall property of the disordered system. Supercell methods use few representations with a large number of atoms. They include Monte Carlo simulations, special quasi-random structures (SQSs) [43] and the similar local atomic environment method [44]. Conversely, spectral methods use many representations with a small number of atoms. Common approaches are cluster expansion, generalized quasi-chemical approximation [45], the Lederer-Toher-Vecchio-Curtarolo (LTVC) method [46], and the partial occupation (POCC) method [47].

Presently, for supercell methods, SQS is the dominant technique. Here, representations are generated such that their atomic pair correlation function is approximately equal to the correlation function of a perfectly random (infinite-temperature limit) alloy. While SQS has been used to predict electronic and mechanical properties of HEMs [48, 49], the lack of finite-temperature information means that no phase transitions may be extracted from this approach.

On the other hand, a variety of spectral methods see practical use. When computational resources are not a limitation, then the cluster expansion, which expands the system's energy in terms of interacting clusters of atoms, is an accurate way to explore the phase diagram. However, for high-throughput, the LTVC method (see figure 2), where cluster interactions are ignored, is more appropriate, still able to predict the miscibility gap by defining an order parameter [46]. Note that when exploring novel compositions, one must consider that combination and decomposition of precursors can generate invariant points, often occurring, but not necessarily, through shifting stoichiometry and critical temperatures of loci already present in the binary/ternary precursors phase diagrams. A typical example is the appearance of unexpected eutectic transformations—melting of the mixture—at temperatures below the expected solid-solution temperature, or the impossibility of removing all oxygen impurities in high-entropy metal carbides by carbothermal reduction [50]. More insidious are invariant points of the whole mixture not inherited from the precursors that can be found *a posteriori*.

If only the properties of a disordered material are of interest, then the POCC method is the ideal choice. This technique, as implemented in AFLOW [51], models a disordered system as an ensemble of small periodic cells (tiles), generated by enumerating all the unique decorations of the system's lattice, up to a cell-size cut-off, for a given species concentration. A property of the disordered system is evaluated by computing that property for each tile and then performing a temperature-dependent ensemble average over the tiles. Multiple methods, from both supercell and spectral, can be combined, supplementing each other, to extract the necessary information relevant to the application.

A few common methods do not fall into either classification. First, the virtual crystal approximation [52] which often gives non-optimal results for materials other than simple metal alloys. Second, the Korringa–Kohn–Rostoker coherent potential approximation, a Green's function approach to calculate properties of disordered systems [53]. While the accuracy of this method can become exact with a large enough simulation cell [54], its usage has been limited to materials with metallic bonding where the local atomic environment is uniform.

Current and future challenges

While many basic properties of high-entropy and complex materials can be reliably computed, there are still a number of challenges that need to be overcome to computationally screen these materials for energy applications. The most pertinent difficulty is determining whether a given composition will

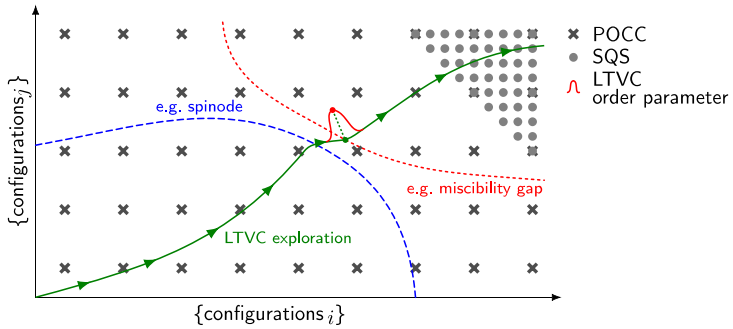

Figure 2. The standard SQS method samples the space of configurations only in regions associated with infinite temperature (the system is completely isomorphous or it has already mixed). To characterize phase transitions, it is needed to (1) sample all configurations (like in the cluster expansion or in POCC) and (2) seek for changes in free energies (Monte Carlo methods to determine phase boundaries) or for features in order parameters evolution (like in LTVC). To characterize microstructure, one needs to go even further by studying convexity of the locus of minimum free energy (like in AFLOW-Spinodal [50]).

synthesize as a high-entropy or CMM. This outcome determines the emergence of local structure in complex materials, enhancing mechanical properties and affects many thermophysical properties, such as thermal conductivity, thermal expansion, specific heat and melting temperature.

Recently, it has been shown that HEMs can exhibit improved electro-, photo-, and thermo-catalytic activities, necessary features; e.g. for water splitting, CO_2 reduction, and hydrogenation applications. Identifying promising candidates requires accurate electronic density of states and relative enthalpy calculations, specifically for highly ionic oxidic systems [50]. The latter are notoriously difficult to get from standard high-throughput compatible DFT due to the chemically distinct nature of different oxides and with respect to their elemental references [55, 56]. The optimization of the catalytic reactions requires the growth/fabrication of surfaces, nanostructures and nanoparticles with controlled sizes, structures, and morphologies. The outstanding radiation resistance of high-entropy materials (in particular carbides) makes them attractive for applications within nuclear fusion reactors [57]. This requires the microscopic understanding of the radiation-matter interaction, and the characterization of the electromagnetic response (e.g. absorption, excitation, dissipation) of those complex materials. It has also been proposed that these materials can serve as important thermoelectric generators used to harvest green energy from waste heat [58]. The exceptional thermal stability of HEMs provides a unique advantage in the proper realization of these high temperature devices. Here, part of the enhancement in the figure of merit is due to the atomic mass and ionic radius mismatch, which influence the heat-carrying phonons. Evidently, screening such materials necessitates an efficient methodology to approximate phonon contributions.

Another recent revelation is using high-entropy materials as photothermal devices for power generation in solar power plants [59]. Lattice defects play a significant part in photothermal efficiency and simulating the experimental defect concentration involves enormous supercells. Therefore, such modeling needs to be done using interatomic potentials [60]. The recent successes of machine-learned interatomic potentials enable first-principles-level accuracy at a fraction of the computational cost, allowing simulations of millions of atoms and detailed exploration of chemical order, lattice distortion, and defect effects in high-entropy materials. However, a key challenge remains: generating sufficiently large and representative training datasets. Overcoming this is crucial for handling the vast compositional and configurational complexity of high-entropy materials, as well as ensuring the transferability and uncertainty quantification of machine-learned potentials across diverse chemical and structural conditions [61].

Advances in science and technology to meet challenges

Recently, the monumental task of classifying whether a solid solution will form a HE single-phase or compositionally complex multi-phase has been addressed by the disordered enthalpy–entropy descriptor (DEED) [50]. DEED captures the balance between the enthalpy cost and the entropy gain of synthesizability through the thermodynamic density of states (see figure 3(a)). Calculating the thermodynamic density of states from spectral methods is usually effortless because the energy distribution of the representations is already known (see cPOCC in figure 3(b) when this is not the case). It can still be calculated from supercell methods, using sampling, such as the Wang–Landau algorithm, although this is much more computationally expensive.

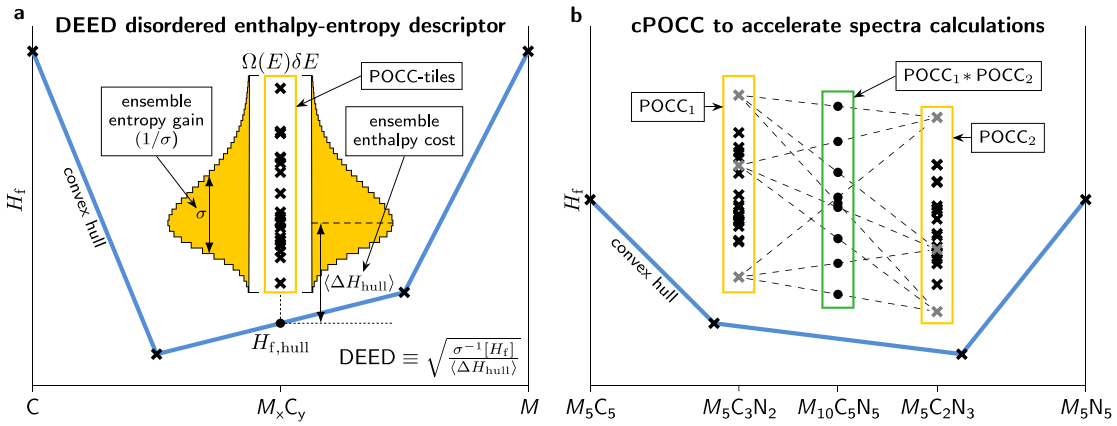

Figure 3. (a) DEED workflow. Calculations of the phase diagram convex hull (blue line) and POCC-tiles (black crosses) give enthalpy spectral momenta. During synthesis, the enthalpy ensemble average is associated to the enthalpy loss of order, while the variance gives the entropic disorder gain. DEED is the balance of the two. (b) cPOCC workflow. The POCC expansion of a $M_{10}C_5N_5$ (M = metal species) high-entropy ceramic (green box) with five unique metal species would require 17.5 million 20-atom tiles. This is overcome by a partition into $M_5C_2N_3$ and $M_5C_3N_2$ subsystems (yellow boxes), each giving 490 ten-atom tiles (black crosses) and the subsequent energy convolutions (black circles). Reproduced from [50]. CC BY 4.0.

Physically motivated empirical corrections can compute accurate enthalpies for ionic HE ceramics. To go beyond the thermodynamic limitations of corrections depending only on compositions, the coordination corrected enthalpies (CCE) method leverages bonding topology and oxidation numbers [56]. This can properly renormalize—even for defect systems—the enthalpy landscape over the whole composition range as demanded for HEMs discovery [50]. The AFLOW-CCE module presents an economic workflow requiring only structural inputs to obtain the corrections.

Plasmonic resonance, the coupling of light to charge density, has recently been discovered in HEMs [62]. This phenomenon can provide an alternative venue to design exotic photothermal devices [63], as well as to realize nanostructures [64] and nanoparticles for photo/electro catalytic reactions [65], where extended and/or localized plasmon–polariton resonances are used to enhance, propagate or dissipate electromagnetic radiation [66].

Designing state-of-the-art deep neural network multi-component interatomic potentials has been greatly simplified due to DeePMD-kit [67], which also integrates with common molecular dynamics packages, lowering the entry barrier for researchers to create such potentials for high-entropy materials.

Concluding remarks

Disordered materials exhibit remarkable properties and are primarily modeled by two methods: supercell or spectral. Supercell methods represent the disordered system by large ordered configurations of atoms, but no finite-temperature information about the disordered material can be extracted; i.e. phase transitions. On the other hand, since spectral methods use a Boltzmann average of small ordered configurations, they are more suitable to investigate the phase diagram at arbitrary temperatures. Using the thermodynamic density of states obtained from a spectral method, we were recently able to take a crucial first step for property characterization. We created a disordered enthalpy–entropy descriptor that captures the balance between the enthalpy cost and the entropy gain, predicting functional synthesizability of single-phase or compositionally complex multi-phase in disordered materials. Still, many important phenomena such as thermal conductivity, specific heat, work function, spinodal decomposition, and plasmonic resonance have yet to be systematically explored in these materials. Furthermore, phase boundaries, microstructural features, precipitates, existence/position of eutectoids, and miscibility gaps will define their applications. Thus, there is a need for novel high-throughput methodologies to tackle these tasks.

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4. High-throughput exploration of CMMs

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Status

A very important scientific and technological question is how to discover, optimize and deploy urgently needed materials with unprecedented, disruptive properties enabling new technologies or making existing technologies sustainable, e.g. replace scarce elements in existing engineering materials by sustainable ones. A promising search space for this are CCMs. The exploration and exploitation of CCM has a long, non-high-throughput history but needs a transformation towards accelerated discovery, optimization and upscaling to deliver urgently needed materials, e.g. for future energy systems, to mitigate climate change. Examples of existing CCM are steels, superalloys, glasses, amorphous magnets and more recently 'high entropy alloys'. The latter are defined as materials with five or more constituents [68], frequently in equiatomic composition, however this is not a criterion for optimized functional properties, i.e. all possible compositions in CCM need to be explored to identify optimum composition(s) for desired properties. Whereas entropy stabilization may play a role in some cases, e.g. against oxidation [69], most functional properties in HEM emerge from the combination of five or more different atoms in one single-phase alloy or compound [8]. The immense search space of elements from the periodic table and their combination into CCM—remaining largely unexplored up to now—makes it likely that hitherto undiscovered materials with desirable properties can be identified.

Current and future challenges

However, this opportunity space is a challenge as the possibilities for quinary combinations from about 50 elements (non-toxic, non-radioactive) yields more than two million combination possibilities. Even when reducing the number of starting elements by focusing on sustainable elements, the combinatorial challenge remains. For each selected CCM systems (A–B–C–D–E) the content of each element needs to be optimized for the desired applications and existence ranges ($A_{a1\text{-}a2}B_{b1\text{-}b2}$–$C_{c1\text{-}c2}$–$D_{d1\text{-}d2}$–$E_{e1\text{-}e2}$) for single-phase or multiphase materials in these systems need to be identified. Additional to composition and crystal structure (which stable or metastable phase(s) exist) the microstructure (e.g. grain size), mainly controlled by processing, needs to be considered in order to optimize intrinsic and extrinsic materials properties. It is a technological challenge to set up the necessary high-throughput tools and infrastructure to enable data-guided discovery in a multidimensional search space combined with multi-criteria optimization to reconcile conflicting requirements, e.g. between sustainability and functionality of new materials. A further challenge is how to bring new materials from discovery into technology. Materials on their own are usually not useful but need to be integrated into systems, which highlights also the importance of interfaces between materials in systems. This means that high-throughput experimentation needs to be extended from compositional and structure screening to system-level screening, i.e. proofing quickly if the discovered materials properties persist if they are integrated into a system. For all of the above, it is a challenge to set-up and use a comprehensive data science infrastructure (research data management systems based on FAIR principles) and then based on this, a suite of AI tools which will help to master the complexity of CCM.

Advances in science and technology to meet challenges

A means to address the 'combinatorial explosion' challenge is to perform high-throughput investigations [8]: best by combining high-throughput simulations, e.g. based on DFT and ML with high-throughput experimentation, e.g. combinatorial synthesis of thin-film materials libraries and the automated characterization of properties. It is essential that the right screening parameters are identified, and that all high-throughput characterization is of high quality; accuracy should be preferred over speed. This can provide consistent and complete multidimensional datasets on CCM systems, which are the basis for multifunctional existence diagrams, comprising correlations between composition, processing, structure and properties, which can be used for the design of future materials. An example is the high-throughput exploration of compositionally complex solid solutions for electrocatalysts [70, 71]: synthesis and characterization of thin-film libraries is tightly coupled with high-throughput calculations. From the comparison of large computational and experimental data knowledge is inferred which is then used to design optimal catalysts. Novel microlibraries combined with scanning electrochemical microscopy is a new approach to accelerate the exploration of the multidimensional search spaces of multinary

electrocatalysts [72]. Also, atomic-scale understanding of surface reactions in electrochemistry can be accelerated by new methods such as the combinatorial processing platform approach [73].

A necessary advancement is to establish accelerated data-driven experiments aiming to substantially reduce the number of necessary experiments combined with increased throughput by (semi-)autonomous experimentation ('self-driving labs' [42, 74–77] 'closed-loop' or with 'human in the loop' [78]. Whereas fully autonomous experiments might be desirable in specific cases, (semi-)autonomous experimentation appears easier to be implemented in existing high-throughput labs and offers advantages of synergistic collaboration between human and AI and capabilities. Acceleration of high-throughput experimentation can be achieved by implementing active learning (Bayesian approach) in the high-throughput workflow: instead of measuring all measurement areas on a materials library only a few but the most important ones are measured, which can yield reliable results with less effort. Whereas this approach works nicely for search spaces with a continuous variation of properties (e.g. [79]), it still has to be proven if it works also, if a hit region is only very small in a composition space. Further advancements would be to achieve the fusion (seamless integration) of simulation and experiments as well as to realize electronic workflows to guide researchers through the complex search space of CCM, using (interactive) visualization software to navigate multi-dimensional data. Adapting AI tools for high-throughput experimentation provides many opportunities: machine-learning accelerated simulations (e.g. [80]) or generative AI (e.g. [81]) could propose materials with desired properties following an inverse design approach (from desired properties as input to the materials showing these properties as output) which then can be investigated by accelerated high-throughput experiments; fast, automated but also interactive analysis and visualization of large datasets, identification of hidden correlations in datasets, learning from comparison between computational and experimental datasets, acceleration of experiments by active learning, inference of synthesis-composition-property relationships from datasets, and (semi)-autonomous high-throughput experimentation. The realization of AI companion agents, which support human researchers in the scientific inference process is a promising approach. An example is a companion agent for x-ray diffraction analysis as an essential tool in materials discovery [82].

Concluding remarks

In conclusion the exploration and exploitation of CCM by combined computational and experimental high-throughput methods supported by AI agents is a promising approach and it is hard to imagine that without it the complexity of CCM could be mastered. However, at the same time (i) the atomic-scale characterization of identified new materials to provide understanding of the discoveries and (ii) the screening of system properties also needs acceleration. This holistic understanding will then accelerate the creation of materials innovations.

Acknowledgments

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5. Highly complex materials for Li-ion and all-solid-state batteries (ASSBs)

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Status

The steadily increasing demand for higher power and more energy dense (rechargeable) batteries is putting pressure on the development of electrode and electrolyte materials with improved properties [83]. This has led to major advancements in electrochemical energy storage, especially in the context of Li-ion batteries (LIBs), also including quasi- and all-SSBs, with more sustainable, ethical, and cost-effective technologies (e.g. Na-ion batteries, SIBs) currently attracting great attention in academic and industrial research [83–85]. Yet, battery applications in the automotive industry are driving the need for more advanced materials and technologies, calling for new concepts and chemistries. In this regard, the introduction of compositional/occupational disorder into established host materials can be seen as a new paradigm offering unprecedented design possibilities.

Among others, oxides, sulfides, and halides have been shown to be synthetically accessible as high-entropy (compositionally complex) electrodes and electrolytes. Various crystal structures relevant to battery applications can accommodate disorder, and indeed, the number of reports on the synthesis and characterization of high-entropy anode, cathode, and electrolyte materials is increasing rapidly [86–89]. Typical examples of beneficial effects arising from high-entropy methodologies encompass increased longevity due to structural stabilization (mitigating strain accumulation, e.g. by suppressing adverse phase transitions) and enhanced ion mobility due to favorable energy landscapes, to name a few (see figure 4). Nevertheless, clear correlations between compositional/occupational disorder or, in other words, configurational entropy and performance have yet to be established [90]. In general, the key differences between low- and high-entropy materials lie in their structural and compositional characteristics (i.e. cocktail effects resulting from intricate interactions between the constituent elements and lattice distortions), which may strongly influence the physical and chemical properties. However, more in-depth investigations are necessary if the fundamental mechanisms are to be fully understood.

A certain degree of stabilization during electrochemical cycling has been observed in different kinds of high-entropy LIB materials, such as intercalation/insertion-type layered and disordered rock-salt oxides [91, 92], as well as sulfides for conversion reactions [93]. Even compositionally complex doping of layered cathodes [94] has been shown beneficial to their cyclability. Apart from that, it has been demonstrated that increasing entropy in solid electrolytes can directly affect their ionic conductivity (and stability), resulting in $\sigma_{\mathrm{ion,rt}} \gg 10$ mS cm^{-1} [95–97], and similar strategies are also being applied to liquid electrolytes in recent years. While different materials have already been tested as electrodes and electrolytes, the effect that compositional and structural tailoring has on the electrochemical properties has not yet been explored in detail and requires further study. That's why it remains largely unclear whether the concept of compositional complexity offers clear advantages in terms of key performance indicators, although improvements over medium- and low-entropy counterparts have been demonstrated in various cases. In layered oxide and polyanionic cathode active materials (CAMs), for example, the introduction of high configurational entropy led to increased specific discharge capacity and capacity retention (see table 1).

Current and future challenges

Even though the field of 'high-entropy batteries' is relatively nascent, rapid progress has led to the development of materials that show promise, on the lab scale, for use as negative or positive electrodes and as electrolytes. In all cases, compositional complexity has been added to structurally pre-defined materials through cation and/or anion mixing (i.e. substitution or doping). Yet, only for some of the materials reported so far, introducing compositional/occupational disorder has led to considerably enhanced properties. For this reason, it is still unclear if high-entropy concepts will help advance the development of next-generation battery technologies. Particularly, it remains to be proven that CMMs are indeed capable of outperforming state-of-the-art counterparts. Regardless, the virtually infinite chemical space available for exploration could pave the way for many discoveries in the future. However, currently, mostly trial-and-error approaches are adopted in the literature to test for new chemistries. With regards to that, high-throughput and automated experimentation, as well as computational materials science based on AI and ML, is becoming increasingly important and has the potential to help make discoveries much more rapidly. Nevertheless, the vast compositional design space poses major challenges for identifying

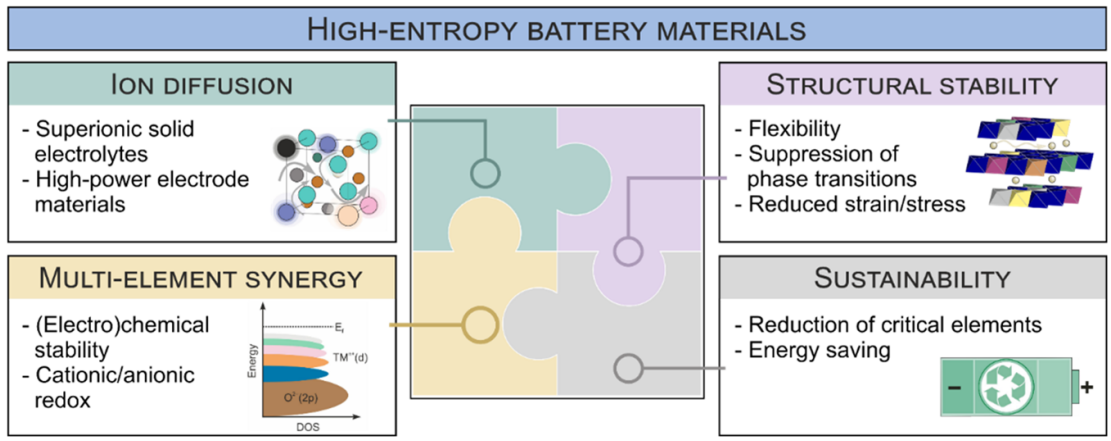

Figure 4. Important attributes of battery materials, including anodes, cathodes, and electrolytes, and overarching benefits arising from high-entropy methodologies.

Table 1. Comparison of key performance indicators of different cathode active materials and their high-entropy counterparts.

Material	Specific discharge capacity (25 °C) (mAh g^{-1})	Capacity retention (%)	References
$Li_{1.15}Na_{0.05}Ni_{0.19}Mn_{0.56}Fe_{0.02}Mg_{0.02}Al_{0.02}O_{1.97}F_{0.03}$	221 (0.5C)	77% (after 200 cycles)	[98]
$Li_{1.2}Ni_{0.2}Mn_{0.6}O_2$	232 (0.5C)	36% (after 200 cycles)	
$Na_{0.7}Mn_{0.53}Ni_{0.26}Fe_{0.15}Mg_{0.01}V_{0.01}Co_{0.01}Cu_{0.01}Zn_{0.01}Sn_{0.01}O_2$	127 (20 mA g^{-1})	~83% (after 100 cycles)	[99]
$Na_{0.67}Mn_{0.57}Ni_{0.28}Fe_{0.15}O_2$	130 (20 mA g^{-1})	~72% (after 100 cycles)	
$Na_{3.5}Fe_{0.5}V_{0.5}Cr_{0.5}Ti_{0.5}(PO_4)_3$	173 (0.1C)	70% (after 5000 cycles at 5C)	[100]
$Na_3V_2(PO_4)_3$	148 (0.1C)	21% (after 5000 cycles at 5C)	

promising battery materials and, more importantly, for determining trends. Likewise, sophisticated characterization techniques are needed to gain insight into structure-property relationships. Because of the complexity of high-entropy materials across length scales, ranging from atomistic to macroscopic, developing a kind of holistic picture is difficult and requires combining complementary experimental and theoretical approaches.

Aside from the more general attributes of electrode and electrolyte materials, other intriguing characteristics are often neither examined nor reported but may be vital to understand the material's properties and behavior. This includes specifically the role of defects. For example, antisite (point) defects in high-capacity, layered oxide CAMs have been intensively studied and recognized to strongly affect their cycling performance and rate capability [101]. Unfortunately, little is known about if and how crystallographic defect chemistry of CMMs can be used for electrochemical energy storage. Of note, their susceptibility to local element clustering and phase separation during synthesis and/or cycling operating is relatively high.

Collectively, the current and future challenges in the development and use of 'high-entropy batteries' are primarily related to the material's synthesis and characterization, with the exploration of structure-composition-property relationships being complex but key to success.

Advances in science and technology to meet challenges

The vast space of compositions and structures makes it impossible to prepare, characterize, and test all materials following a classical laboratory approach. For that reason, combinatorial synthesis and automated characterizations are necessary to guide, in an efficient manner, through the labyrinth of potential compositions, aiming at systematically screening a multidimensional space. High-throughput synthesis and characterization techniques offer one of the best opportunities to achieve this goal. However, even with high-throughput methodologies, the enormous number of possible element combinations cannot be fully covered. Hence, knowledge from experience and insights gained from experiments must be fed back into the next rounds of synthesis. Ideally, this occurs through an iterative combination of AI-driven preparation in combination with ML methods in a closed loop. Such an approach may not only pave the

way toward the development of tailored energy materials [102], but also help identify cross-connections between experimental results and compositions that may elude human observers.

Equally important with regards to application of high-entropy materials in rechargeable batteries is to evaluate their techno-economic potential. First and foremost, this relates to the production itself, which relies largely on (laborious) high-temperature (HT) solid-state synthesis with limited control over particle morphology and primary/secondary particle size (specific surface area). Note that the latter attributes are known to have a profound effect on slurry rheology in general, as well as on the cycling performance of layered cathodes. Therefore, a closer look should be taken at the preparation of high-entropy precursor electrode materials, especially so-called pCAMs. Likewise, the synthesis of (quasi) single-crystal electrodes, which are currently in the spotlight of next-generation electrochemical energy storage, e.g. for SSBs, should be explored. The same holds for the stabilization of anion (oxygen) redox in high-entropy CAMs to boost energy density. However, due to the early stage of research, the compatibility of high-entropy materials with established industrial processes cannot be reliably assessed at this time.

Overall, it can be stated that advances made in science and technology strongly drive the field and will help assess the true merits of high-entropy methodologies. Regardless, research on compositionally complex battery materials will clearly benefit from high-throughput approaches and screening of key properties, such as electrochemical stability, (cationic/anionic) redox activity, and charge transport.

Concluding remarks

So far, there are only a few examples of high-entropy electrode and electrolyte materials that are seemingly capable of outperforming their compositionally less complex counterparts. In general, there is clearly a lack of research on 'high-entropy batteries' given the infancy of the field, which makes it somewhat difficult to predict if this new paradigm will represent an advance in electrochemical energy storage in the near future. This is also due, in part, to major difficulties in property-targeted compositional design of energy materials. Nevertheless, the exploration of compositionally complex anodes, cathodes, and electrolytes for application in LIBs, SIBs, or SSBs holds promise for improving established materials or even finding new material classes. However, the true benefits of high-entropy methodologies remain largely unclear at present, making additional research efforts imperative. In this regard, using high-throughput/automated experimentation, along with computational materials science and advanced characterization techniques, is believed to be key to accelerate research and development processes. Moreover, it should be noted that several aspects, such as synthesis optimization (including tailoring of particle size, shape, faceting, etc.) and upscaling, effects of different kinds of defects on properties, recycling, and so forth, have been either completely or partially disregarded so far, but will become important for real-world applications.

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6. Complex materials for post-lithium batteries

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Status

Rapidly growing markets for portable electronics, electromobility, renewable energy and decentralized energy production, such as home solar cells, have led to an explosion in the necessity for LIBs. In recent years, the demand especially for Li has skyrocketed, leading to supply constraints also due to geopolitical and environmental factors. The current supply and the increase in demand has led to concerns about whether the supply can keep pace, especially as the transition to renewable energy and electric mobility accelerates.

In order to overcome the limitations of resource scarcity and environmental impact of the current Li-ion technology, but also to reduce safety concerns like thermal runaway, dendrite formation, overcharging or leakage that can lead to heat generation, and in extreme cases, explosions, alternative technologies must be developed for lithium-based and post-lithium technologies [103, 104]. Post-lithium technologies represent a current frontier in energy storage, aiming to solve the abovementioned issues and to maintain the development of our society and to enable the critical transition to a renewable energy future. A promising class of materials are complex multicomponent and HEMs [18, 99, 105]. Their application spans over a wide range of different Li- and post-Li technologies, typically owing to their possibilities to precisely tailor the electrochemical performance, to offer very specialized properties and due to synergistic effects appearing by the multitude of integrated components [14, 106].

HE ceramics [19, 20] and HE metal–organic frameworks (MOFs) [107] are particularly in focus for the development of new active materials for electrodes and electrolytes. Oxide-based HE ceramics, as well as sulfides, fluorides, and many other types of ceramics, are being developed as electrode or electrolyte materials. It has been found that many compounds exhibit complex interactions among the constituent elements, leading to unpredictable advantageous properties of the materials like increased cycle life, improved capacities, elevated working potentials, just to name a few [108, 109]. For instance, some HE oxides demonstrate significantly increased electrical conductivity, formed by a metallic 3D network due to the alloying of certain constituent elements, which substantially enhances the kinetic properties of the material and, consequently, its cycling stability [110]. HE MOFs have also shown significantly improved cycling stabilities in sodium insertion materials by inhibiting a phase change that would otherwise occur during the charge/discharge process.

Beyond empirical observations, several complementary mechanistic concepts have emerged to explain the enhanced structural stability of high-entropy and multicomponent materials in post-lithium chemistries. While configurational entropy may contribute to the stabilization of certain solid solutions, growing evidence suggests that structural, electronic, and kinetic effects often dominate the observed improvements in cycling stability. A central factor is the introduction of local lattice distortions and elastic strain fields arising from the coexistence of cations with different ionic radii, valence states, and bonding preferences. These distortions disrupt long-range ordering and suppress cooperative phase transitions such as P2–O2 transformations or Jahn–Teller distortions in sodium- and potassium-based layered oxides, thereby stabilizing host structures during repeated ion insertion and extraction. In addition, electronic landscape engineering plays an important role. The presence of multiple redox-active species creates a distribution of local electronic environments that enables charge compensation across several elements rather than concentrating redox stress on a single site. This mitigates over-reduction, suppresses irreversible phase evolution, and stabilizes electrochemically active frameworks over prolonged cycling. On a kinetic level, chemical disorder leads to a broadening of ion migration pathways and distributed activation energies, which reduces the likelihood of transport bottlenecks under high current densities. In several HEOs and sulfides, this effect has been correlated with enhanced effective ionic mobility and reduced degradation during fast cycling. At the electrode–electrolyte interface, compositional complexity can furthermore promote the formation of thinner and more uniform solid electrolyte interphases, likely as a result of spatially distributed surface reactivity and a reduced tendency toward localized decomposition. Together, these effects illustrate that the stabilization mechanisms of high-entropy materials in batteries arise from an interplay of structural disorder, electronic flexibility, and kinetic smoothing rather than from entropy alone.

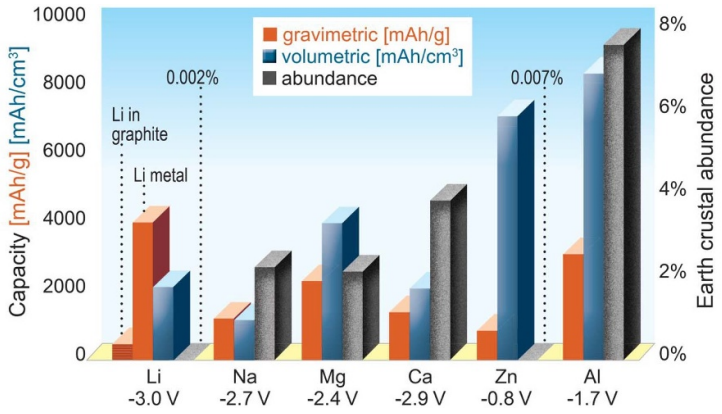

Figure 5. Earth crustal abundance, gravimetric and volumetric capacities of Li and post-Li anodes.

Current and future challenges

Post-Li technologies offer advantages over Li-ion but face significant challenges before market introduction, as illustrated in figure 5. Key scientific issues include lower practical energy density and reduced lifespan and cycle stability of several post-Li chemistries. Multicomponent and HEMs provide a promising solution [111].

While lithium-ion batteries currently dominate in gravimetric and volumetric energy density, post-lithium high-entropy systems may surpass lithium-based technologies under specific conditions. In stationary energy storage, where material abundance, cost stability, safety, and lifetime are more critical than weight or volume, sodium- and potassium-based high-entropy cathode and electrolyte systems could become competitive owing to their superior thermal stability and reduced fire risk. In addition, the compositional flexibility of high-entropy materials enables tuning of redox potentials, lattice rigidity, and interfacial chemistry simultaneously, capabilities that are increasingly difficult to realize in heavily optimized lithium systems. This makes post-Li HE materials particularly attractive for applications requiring long-term cyclability, tolerance against overcharge or deep discharge, and operation under harsh conditions (e.g. high temperatures or fluctuating current loads). Beyond Na and K systems, magnesium- and aluminum-based batteries offer multi-electron redox and thus intrinsically higher volumetric capacities, provided that suitable high-entropy host lattices enable fast ion transport and reversible phase behavior. In such scenarios, HE design strategies could unlock performance regimes inaccessible for conventional lithium-ion materials. Consequently, post-lithium high-entropy materials are not envisioned as direct replacements for LIBs in all consumer applications, but rather as enabling technologies for infrastructure-level use cases where sustainability, safety, and durability dominate over maximal energy density.

Developing novel multicomponent materials is complex and time-consuming, though highly rewarding. While multicomponent materials are also utilized in Li-ion technology, the evolution from the first Li-ion cathode material, $LiCoO_2$, in 1991 to the multicomponent $LiNi_xCo_yMn_zO_2$ ('NCM') in the early 2000s, and the continuous optimization of NCMs over the past 30 years, underscores the complexity of developing such systems, even with 'only' three incorporated elements. Complex multicomponent and HE systems often involve five or more different metals, leading to an almost limitless number of possible compositions and stoichiometries. This combinatorial explosion makes the targeted development of desired materials an exceedingly time-consuming task, nearly impossible to achieve with an Edisonian approach, following traditional synthesis and development techniques.

Furthermore, not all possible element combinations yield competitive material properties. Often, it requires extensive development and optimization of the structure, choice of elements, and stoichiometry before meaningful progress can be achieved. Therefore, it is critical to have the capability to select the most promising materials from an almost infinite chemical parameter space and identify suitable synthesis pathways. However, the complexity of these materials also complicates this selection process. Computational approaches, such as DFT calculations, which could provide predictive insights for a specific material class, are often too computationally expensive or impractical due to the high complexity of the HE materials, which result in too many variables or overly large unit cells for precise calculations.

Additionally, the selection of elements in complex materials must be done with careful consideration, which significantly reduces the range of possible elements. Ethical, biological, and geopolitical factors may further limit the selection; for instance, toxic, critical, or unethically sourced elements should be avoided. This reduction in available elements limits the possibilities for creating complex materials from the most optimal components. Moreover, the disposal and recycling of materials containing multiple ingredients can become a highly challenging issue.

The sustainability of complex multicomponent materials presents both opportunities and challenges. On the one hand, high-entropy systems offer the possibility to incorporate abundant and non-critical elements (such as Fe, Mn, Mg, Al, or Na), thereby reducing dependence on cobalt, nickel, or lithium. On the other hand, the integration of many elements complicates established recycling strategies, which are optimized for relatively simple material streams. Future battery design must therefore adopt '*recyclability-by-design*' principles, including element grouping by chemical affinity, phase-selective leaching strategies, and compositionally tolerant recycling routes. Research is urgently needed on selective separation techniques for multicomponent oxides and sulfides, as well as life-cycle analyses that quantify ecological costs per stored kilowatt-hour. In addition, reuse concepts (e.g. remanufacturing of cathode powders) could become particularly attractive for stable high-entropy systems that retain structural integrity even after long cycling periods.

Another point that must be considered is the highly reactive surface of HE materials. Due to the heterogeneous surface, highly reactive regions with atomically small catalytic centers can emerge, which can promote side reactions, such as increased electrolyte decomposition and the associated gas generation and solid electrolyte interphase formation, during the complex processes of charging and discharging. Additionally, the formation of single-phase materials is not always straightforward; different synthesis conditions can easily lead to phase separation phenomena that significantly alter the elemental interactions. For these reasons, it is crucial to precisely determine and control the choice of elements as well as the synthesis conditions. Moreover, a dedicated coating strategy can prevent these effects from having a negative impact.

Advances in science and technology to meet challenges

The complexity of developing new materials, coupled with the vast array of possible element combinations and a lack of powerful modeling strategies, highlights the importance of experimental high-throughput methods. These techniques significantly accelerate the synthesis and characterization of numerous samples, currently achieving about 500 samples per week. However, this throughput appears modest against the backdrop of potential combinations; for instance, selecting 5 elements from 15 choices with 100 different stoichiometries results in ca. 300 000 combinations. Thus, while high-throughput methods are invaluable, additional strategies involving data handling, ML, and design of experiments are essential to effectively deal with this vast number of possibilities in a reasonable timeframe [112, 113].

In fact, the rise of AI and self-learning neural networks may offer a promising approach to address these challenges. Automated synthesis, battery assembly, and testing stations can efficiently manage and analyze large datasets, enabling predictive modeling of elemental combinations and stoichiometries through iterative experimental cycles. For example, Bayesian optimization tools can assist AI in narrowing down possibilities and making informed suggestions for experiments, accelerating the development of complex and HE materials for various post-Li battery types [114]. However, a critical issue remains the synthesis itself. For a well working host material it is not sufficient 'just' to mix atoms randomly, as it is the case in multicomponent sputtering-based methods. Rather, it is a particular phase with a particular structure that must be formed during the process.

Several post-Li techniques have been proposed, each with distinct performance parameters. These include batteries that substitute lithium with metals like sodium, magnesium, zinc, calcium, potassium, and aluminum, as well as those using organic or MOF-based cathodes (e.g. Prussian blue or porphyrins), batteries utilizing anions as charge carriers (e.g. chloride or fluoride) [115], redox flow batteries, and SSBs. For instance, typical sodium-ion CAMs include $Na_x(Co,Mn)O_2$, $Na_x(Fe,Mn)PO_4$, $Na_x(Ni,Mn)O_2$, $Na_xV_2O_5$, and $Na_x(Ni,Co,Mn)O_2$, which incorporate two or three transition metals, akin to those in existing lithium-ion batteries. The selection of elements hinges on their individual properties, with synergistic effects optimizing capacity, stability, and cost. Recently, such sodium-ion batteries have entered the HE domain by incorporating an even broader range of elements, enhancing their interactions. Up to seven different cations (Mn, Ni, Co, Ti, Mg, Al, Fe) have been integrated into a P2-layered structure, demonstrating superior electrochemical performance compared to materials with fewer elements. HE P2-layered materials have shown promise in mitigating phase transitions, which typically lead to degradation and capacity loss [116, 117]. Similarly, HE Prussian blue analogues (PBAs) can insert

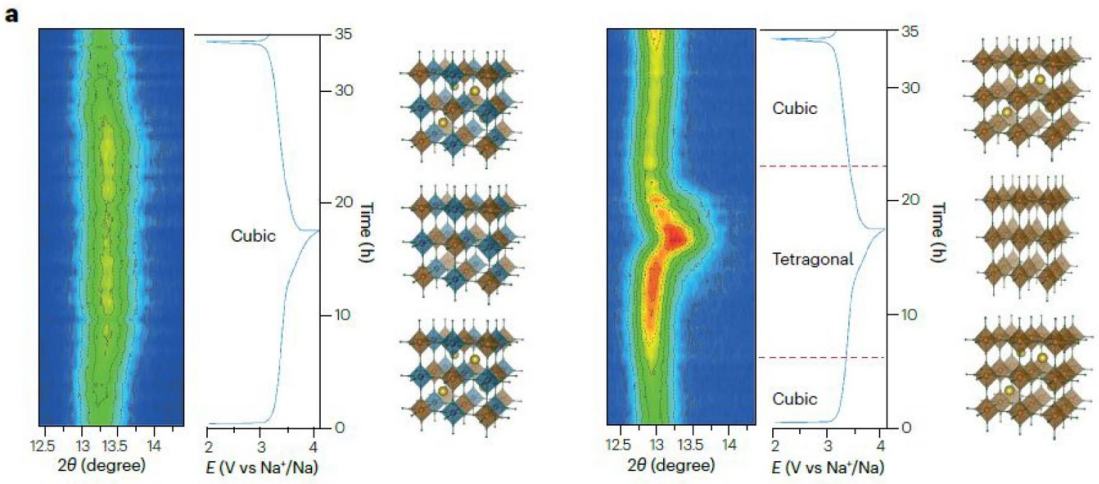

Figure 6. Example for entropy mediated structural stabilization. The high entropy Na-PBA (a) remains in a cubic structure over the whole charge/discharge process, while the normal PBA (b) undergoes a phase transition between a cubic and a tetragonal structure, which finally leads to material decomposition and reduced cycle life. Reproduced from [14], with permission from Springer Nature.

sodium ions and act as CAMs, demonstrating stabilization of their cubic structure during cycling (see figure 6) [107]. These complex materials enhance properties and cycle life in conventional post-Li batteries, showing promise despite mechanisms still under investigation.

Concluding remarks

The use of complex multicomponent and HEMs holds great promise in future batteries. This is due to the flexibility in composition, which allows tailoring of properties, the thermodynamic stabilization due to the introduced configurational entropy, and the synergy effects resulting from the interactions of the incorporated elements. This synergy, commonly known as the 'cocktail effect', has been demonstrated in many different classes of materials. Examples include the reduction of toxic elements and the stabilization of certain crystal structures, leading to improved cycle life. Additionally, lattice distortions can impact mechanical properties.

With the implementation of AI-supported self-driving labs, it is expected that a plethora of novel functional materials for post-Li technologies can be developed in the near future. This development could critically strengthen the research field, paving the way for a significant market introduction to address and support the major energy challenges of today

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7. HEMs for rechargeable aqueous metal–air batteries

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Status

Metal-air batteries have a long history, dating back to the 19th century (figure 7) [118]. They operate through an electrochemical reaction between a metal anode and atmospheric oxygen, mediated by an ion-conducting yet electronically insulating electrolyte. During discharge, the metal anode oxidizes, releasing electrons to an external circuit and metal ions into the electrolyte. In aqueous electrolytes, these metal ions react with hydroxide ions produced at the cathode through the reduction of oxygen from the air. In organic electrolytes, the metal ions diffuse to the cathode and react with oxygen to form metal oxides. The process is reversed during charging. One of the key advantages of metal–air batteries is that the cathode material is sourced directly from the air, rather than being stored in the battery, allowing for significantly enhanced energy densities [119, 120]. Furthermore, aqueous metal–air batteries are particularly promising due to their potential for cost-effectiveness, safety, and sustainability. Among metals, zinc stands out in aqueous electrolytes for its excellent electrochemical stability, low cost, and ease of assembly in ambient air. While primary metal–air batteries are already commercially available—offering substantial energy in a compact, lightweight form that extends device lifespan and reduces battery replacements—their rechargeable counterparts face several challenges that must be addressed before they can be widely adopted.

The primary challenge in developing aqueous secondary metal–air batteries lies in the performance of the cathode, which is highly dependent on the activity and durability of catalysts capable of accelerating the OER and ORR. These reactions involve sluggish, multi-electron proton-coupled kinetics, resulting in slow reaction rates and high overpotentials that limit the charging rate, output power, and energy efficiency of aqueous rechargeable metal–air batteries. While numerous mechanisms and catalyst design strategies have been explored, achieving efficient bifunctional activity—where both OER and ORR proceed with low overpotential and high reversibility—remains a major obstacle. Current state-of-the-art catalysts, such as Pt/C (for ORR) and Ir/C (for OER), deliver high activity but suffer from poor stability in aqueous electrolytes and high costs, limiting large-scale application. Consequently,to address these limitations, the development of noble-metal-free, bifunctional oxygen catalysts is being actively pursued as a promising alternative.

One of the key challenges in developing bifunctional oxygen catalysts lies in the varying binding strengths of the intermediates involved in both the OER and the ORR. Because intermediates in the two reactions require different adsorption energies, catalysts optimized for one often underperform for the other. In this context, high-entropy materials (HEMs) are highly promising as bifunctional OER/ORR catalysts due to their compositional flexibility, which enables precise tailoring of surface properties for optimal catalytic performance [130, 132, 133]. Additionally, their broad range of surface sites offers a nearly continuous distribution of adsorption energies. This quasi-continuous spectrum of binding energies can significantly lower overpotentials in electrocatalytic processes, while simultaneously boosting catalytic activity in multistep and even reversible reactions, such as OER and ORR [134, 135]. While high-entropy aqueous electrolytes in zinc-air batteries [136] and several HEM-based solid-state electrolytes have been also proposed [135], our focus here will be on the challenges and opportunities of using HEMs as oxygen catalysts at the air cathode of metal–air batteries, where they show particular promise. A benchmark comparison between noble-metal catalysts and high-entropy materials using in state-of-the-art air cathodes for ZAB showing in table 2.

The electrocatalytic performance of representative HEM-based catalysts—including ORR half-wave potential ($E_{1/2}$), OER potential at 10 mA cm^{-2}, and corresponding ZAB metrics such as power density, specific capacity, and durability—is comprehensively summarized in table 3.

Current and future challenges

Applying HEMs as air cathode in metal–air batteries presents significant challenges on different fronts, from the design and engineering of the materials to the characterization of their properties, particularly under relevant reaction conditions, and the understanding of the key reaction sites and mechanisms behind their performance, which hamper their optimization.

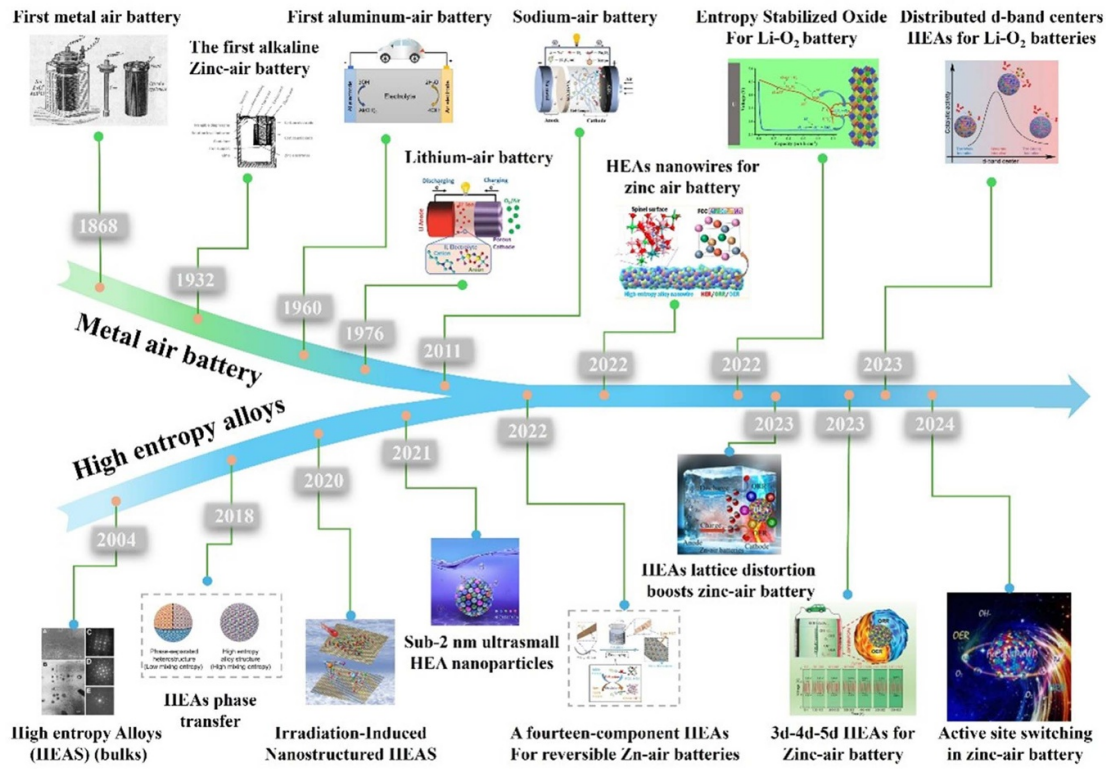

Figure 7. Progress of metal–air batteries and parallel advancements in the use of HEMs as air cathodes. Reproduced from [121] with permission from the Royal Society of Chemistry. [122] John Wiley & Sons. © 2023 Wiley-VCH GmbH. Reproduced from [123]. CC BY 4.0. [18] John Wiley & Sons. Copyright © 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. From [124]. Reprinted with permission from AAAS. [125] John Wiley & Sons. © 2020 Wiley-VCH GmbH. Reprinted with permission from [126]. Copyright (2021) American Chemical Society. Reproduced from [127] with permission from the Royal Society of Chemistry. Reprinted with permission from [128]. Copyright (2020) American Chemical Society. [129] John Wiley & Sons. © 2022 Wiley-VCH GmbH. Reprinted from [130], Copyright (2023), with permission from Elsevier. [131] John Wiley & Sons. © 2023 Wiley-VCH GmbH. [132] John Wiley & Sons. © 2023 Wiley-VCH GmbH. Reproduced from [133] with permission from the Royal Society of Chemistry.

HEMs offer vast possibilities by combining five or more distinct elements, enabling the use of abundant, non-toxic materials for high-performance oxygen cathodes. However, this diversity also creates the primary challenge: exploring the vast range of possible compositions requires high-throughput computational techniques to predict stable compositions, structures, and properties. Establishing reliable structure-composition-performance correlations remains an ongoing challenge.

Another challenge lies in producing HEMs in cluster or nanoparticle form. One approach uses thermodynamics to drive HEM formation, leveraging entropy while limiting particle growth at high temperatures. Alternatively, HEM formation can be kinetically driven by promoting rapid reactions of homogeneous reactant mixtures or using highly diluted precursor dissolutions. Experimental evidence suggests enthalpy also plays a role, with nanoscale surface energy costs stabilizing the HEM phase [148]. Scaling up production while maintaining quality and performance for applications like electric vehicles and energy storage remains another hurdle. Environmental impact and economic viability are also crucial considerations. Efficient synthesis processes and the use of abundant, non-toxic materials are essential for sustainability. However, recycling HEMs is particularly difficult due to the integration of multiple metals, complicating the separation and recovery of individual elements.

Durability and corrosion resistance under aqueous ZAB conditions represent another critical challenge and evaluation criterion for HEM-based catalysts. In strongly alkaline or near-neutral electrolytes, repeated oxygen evolution and reduction cycling can cause dissolution of metal cations, surface reconstruction, or phase segregation, leading to performance degradation. Nevertheless, the multi-element synergy and high configurational entropy of HEMs mitigate these effects by suppressing selective leaching of individual components and enhancing overall structural stability. The 'sluggish diffusion effect' inherent to high-entropy systems reduces atomic mobility, thus impeding corrosion propagation. Recent studies have demonstrated that properly engineered HEMs exhibit remarkable durability in aqueous conditions, due to thermodynamic contribution. For example, the septenary HEA PtAuPdFeCoNiCu exhibits excellent ORR stability, with a positive half-wave potential drift of 7.4 mV after 20 000 cycles of

Table 2. A benchmark comparison of the ZAB performances obtained using state-of-the-art air cathodes between noble-metal catalysts and high-entropy materials.

Catalysts	Open circuit potential (V)	Peak Power density (mW cm^{-2})	Specific capacity (mAh/gZn) @ (mA cm^{-2})	Cycling condition and stability (h)	References
AlNiCoRuMo	1.48	146.5	—	2 mA cm^{-2}, 10 min/cycle for 120 h	[137]
AlFeCoNiCr	—	125	800@20	2 mA cm^{-2}, 10 min/cycle for 60 h	[138]
Pt/C + IrO_2	1.41	87	732@20	2 mA cm^{-2}, 10 min/cycle for 28 h	[138]
Pt/C + RuO_2	1.56	112.44	896@8	8 mA cm^{-2}, 20 min/cycle for 100 h	[132]
FeCoNiMoW	1.59	116.90	1040@8	8 mA cm^{-2}, 20 min/cycle for 660 h	[132]
FeCoNiPdWP	1.60	123	886@8	8 mA cm^{-2}, 20 min/cycle for 700 h	[133]
Pt/C + RuO_2	1.56	111	793@8	8 mA cm^{-2}, 20 min/cycle for 120 h	[133]
PtFeCoNiMnGa	1.52	130.6	811.4@10	5 mA cm^{-2}, 20 min/cycle for 120 h	[139]
Pt/C + RuO_2	1.44	107.3	758.1@10	5 mA cm^{-2}, 20 min/cycle for 40 h	[139]
CoCuFeAgRu	1.44	136.53	987.9@10	10 mA cm^{-2}, 10 min/cycle for 40 h	[140]
$FeCoNi_2F_4(OH)_4$	1.44	131	873@10	5 mA cm^{-2}, 20 min/cycle for 370 h	[141]
CoNiFeCuMoOOH	—	150	—	10 mA cm^{-2}, 20 min/cycle for 160 h	[142]
Pt/C + RuO_2	—	118	—	5 mA cm^{-2}, 20 min/cycle for25 h	[142]
PtFeCoNiMn	1.46	545	—	50 mA cm^{-2}, 20 min/cycle for 165 h	[143]

Table 3. Comparison of electrocatalytic ORR and OER activity and ZABs performance obtained using HEMs-based electrocatalysts.

Catalysts	OER@ 10 mA cm^{-2}	ORR@$E_{1/2}$	Power density (mW cm^{-2})	Specific capacity (mAh/gZn@ mA cm^{-2})	Stability (Cycles@ mA cm^{-2})	References
AlNiCoRuMo	—	0.88	146.5	—	360@2	[137]
FeNiCoMnRu	1.64	0.78	81	735.5@10	—	[144]
CrMnFeCoNi	1.51	0.78	116.5	836@8	720@8	[130]
FeCoNiMoW	1.46	0.71	116.90	1040@8	1980@8	[132]
FeCoNiPdWP	1.46	0.81	123	886@8	2100@8	[133]
MnNiCuCoZnF	1.54	0.85	219	768@10	600@10	[145]
PtFeCoNiMnGa	1.47	0.87	130.6	811.4@10	360@5	[139]
SrFeCoNiMoO	1.53	0.64	96.6	799@10	150@10	[146]
CoCuFeAgRu	1.51	0.84	136.53	987.9@10	240@10	[140]
$FeCoNi_2F_4(OH)$	1.52	—	131	873@10	1110@5	[141]
CoFeCuNiZn	1.50	0.88	202	811@10	72@2	[147]
PtFeCoNiMn	1.47	0.94	545.5	—	495@50	[143]

cyclic voltammetry in alkaline media [149]. Similarly, FeCrCoNiAl based HEO films have been shown to exhibit minimal metal leaching and sustained OER performance under alkaline conditions [150]. Although the superior stability of HEAs is often attributed to the hypothesis of thermodynamic contributions. Korma´nyos *et al* proposed a dissolution mechanism primarily governed by kinetics [151]. They synthesized a compositional series of alloy thin films, ranging from a single-element system (Pt) to binary (PtRu), ternary (PtRuIr), quaternary (PtRuIrRh), and quinary (PtRuIrRhPd) counterparts. Under alkaline conditions, the dissolution of certain metals was found to decrease with increasing compositional complexity. However, this improvement was relatively modest and was more plausibly attributed to the dilution of each constituent element within multicomponent alloys rather than to any cooperative thermodynamic stabilization effect. Controlling the composition and distribution of elements,

especially at the surface of HEM nanoparticles, requires precise atomic-scale characterization to obtain accurate feedback to tailor these materials. Elemental arrangement is often not random; short-range ordering, revealed through both experimental and theoretical studies, may play a critical role in defining OER/ORR performance [152–154]. However, accurately characterizing this ordering is challenging, complicating the establishment of reliable structure-composition-property relationships. Besides, the surface composition of HEM nanoparticles often varies significantly from the bulk material due to differences in chemical environments, which can greatly influence their performance in applications that involve interaction with external media [155]. An important objective in this field is to pinpoint the most efficient atomic configurations and develop methods to increase their prevalence for improved performance. Reaching this goal involves not only identifying the precise composition and atomic structure but also understanding how these characteristics change under real-world operating conditions.

Advances in science and technology to meet challenges

Recent scientific and technological progress has significantly addressed challenges associated with using HEMs in metal–air batteries. Improved synthesis methods, such as solution-phase synthesis, now provide better control over HEM composition and microstructure, while also lowering fabrication costs. These innovations facilitate the scalable production of HEMs without compromising electrochemical performance, bringing them closer to practical application. However, further progress in the tailored synthesis of these materials requires identifying and controlling the dominant synthesis mechanism, which requires accurate calculations of enthalpy and entropy, along with advanced computational models that can simulate dynamic processes during synthesis. This improved mechanistic understanding is crucial for reliably producing HEM nanoparticles with consistently optimized properties in a more sustainable, efficient, and cost-effective manner.

In the need to explore the wide range of possible compositions HEMs offer and establish reliable structure-performance correlations, HEMs serve as an ideal platform for advancing material screening strategies, including *ab initio* molecular dynamics combined with DFT, combinatorial and automated high-throughput synthesis, and ML tools for guiding HEM design and OER/ORR performance optimization [72, 156–158]. These computational–experimental frameworks accelerate discovery and help identify stable, active surface configurations under realistic voltages and electrolytes.

However, optimizing the performance of HEM-based air cathodes hinges on precisely identifying the active reaction sites and their chemical environment under operational conditions. This requires atomic-scale characterization, particularly of the surface chemistry of HEMs within battery environments. Conventional techniques like HAADF-scanning transmission electron microscopy (STEM)-EDS often lack the sensitivity to detect subtle short-range ordering, necessitating the development of alternative *ex situ* and *in situ/operando* methods, along with advanced data processing techniques, to gain deeper insights into the atomic distribution within HEM nanoparticles and how they evolve during operation. These experimental findings should be integrated with more sophisticated theoretical models and simulations to accurately predict HEM performance and behavior under operational voltage and within the battery electrolyte.

Concluding remarks

Although still in the early stages of development, HEMs offer exciting opportunities to enhance the performance of metal–air batteries, particularly by addressing the limitations of air cathodes in accelerating the sluggish OER and ORR without relying on scarce or toxic elements. The vast range of available HEM compositions, combined with their surface site versatility, provides an ideal platform for developing materials with superior catalytic activity, stability, and durability.

However, HEMs also present significant challenges, including cost-effective synthesis, reliable and precise control over composition, particularly on the surface of small particles, long-term stability, accurate identification of catalytic sites and reaction mechanisms, and the optimization of their surface chemistry based on the gained insights. Additionally, scaling up production for real-world applications while maintaining consistent performance remains a critical hurdle.

As research progresses, establishing clear structure-property relationships and exploring how high configurational entropy and elemental diversity affect HEM stability and OER/ORR performance will be crucial. Advances in innovative synthesis methods, surface engineering, atomic scale characterization tools, and computational modeling will be essential to overcoming these challenges and unlocking the full potential of HEMs as a transformative solution to current metal–air battery limitations, offering more efficient, higher density, and durable energy storage solutions for the transportation sector and the integration of renewables.

Acknowledgments

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8. Complex materials for solid oxide fuel and electrolysis cells applications

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Status

The SOFCs and solid oxide electrolysis cells (SOECs) are technologies of great prominence for the next-generation power industry. Offering unmatched efficiency, low level of pollution, and ability to both produce (Fuel-to-power mode) and store (Power-to-fuel mode) energy [9, 159], they are tailor-made for the renewable sources-dependent power grids, enabling seamless coverage for the inherent fluctuations in their energy production levels. Compared to other technologies, such as LIBs, SOFCs and SOECs offer a number of advantages, including a much lower cost of energy storage [159], a much lower dependence on critical raw materials [159], and the possibility of utilizing the already existing gas infrastructure [160]. However, despite the advantages listed, the widespread application of SOCs is still relatively limited, mainly due to their excessive operating temperatures, exceeding 850 °C in legacy HT-SOFCs [9]. Such temperatures have a profound impact on the longevity of the stacks, increase their start-up time, impose the need for extensive thermal insulation, which decreases the mobility of the whole technology, and last but not least, significantly increase the cost of stack production, which by itself is a major limitation [161]. The introduction of intermediate-temperature SOFCs (IT-SOFCs, 650 °C–850 °C) and the subsequent development of low-temperature SOFCs (LT-SOFCs, <650 °C) [9], while partially addressed these issues, at the same time created new ones. Although the change from ceramic to metallic interconnects that occurred during the transition from HT- to IT-SOFC [9], allowed for a significant reduction in production costs, it also introduced new mechanisms of cell degradation, such as Cr-poisoning, resulting from the interaction between new interconnects and cathode materials [162]. Furthermore, while in HT-SOFCs electrolyte performance was to a large extent the limiting factor, the decrease in operating temperatures combined with the insufficient catalytic activity of the state-of-the-art air electrodes resulted in the latter becoming the main obstacle in further development of SOC technology [9]. This proved to be a major problem, as in most commonly utilized perovskite-structured electrode materials, increasing catalytic activity usually comes at the cost of decreased functionality: reduced structural stability, increased thermal expansion, and increased susceptibility to Cr- and CO_2-induced degradation [162]. Therefore, out-of-the-box solutions have constantly been pursued to overcome this performance-functionality trade-off, among which the utilization of the HEOs appears to be particularly promising because of their unconventional features stemming from the synergistic effects.

Current and future challenges

The introduction of HEOs created a number of new opportunities in the design of almost all components of SOCs, particularly electrodes and electrolyte layers [163, 164]. However, this vastness of the available compositional space can also be considered one of the biggest challenges associated with the HEOs application.

In the context of electrolyte materials, the HEO design principles to a large extent collide with the established guidelines of electrolyte design. Most state-of-the-art oxygen and/or hydrogen ionic conductors display a sharp correlation between the content of acceptor dopants and the ionic conductivity, greatly limiting their useful content [165, 166]. At the same time, the elements responsible for the creation of the main oxide structure are also not easily exchangeable, often leading to inferior performance for their combinations [167]; see figure 8(a). As a consequence, the near-equimolar approach to element ratios typically associated with HEOs appears to be of limited use. Nevertheless, compositionally-complex oxides that do not strictly follow the definition of HEOs have already shown good promise, either by implementing multicomponent combinations of dopants in a moderate concentrations [164], or with regard to protonic conductors [168]. The recent developments of novel types of ionic conductors [169] also create further opportunities for utilization of multicomponent systems.

In terms of HEOs electrodes, so far the main focus has been on the air-electrode materials, which is to be expected considering the already adequate performance of state-of-the-art fuel-electrodes [9]. It is important to realize that as of today, the main strength of the HEOs air-electrodes appears to lie not in their raw electrochemical performance, which is mostly comparable to that of the conventional analogs, figure 8(b), but rather in the possibility of bringing together the conflicting requirements of performance and functionality, either by improving the former in otherwise inferior but highly functional materials, such as alkali-free compositions [171, 172], or by enhancing the stability and degradation resistance of materials characterized by already superior catalytic performance [164, 173, 174], or by

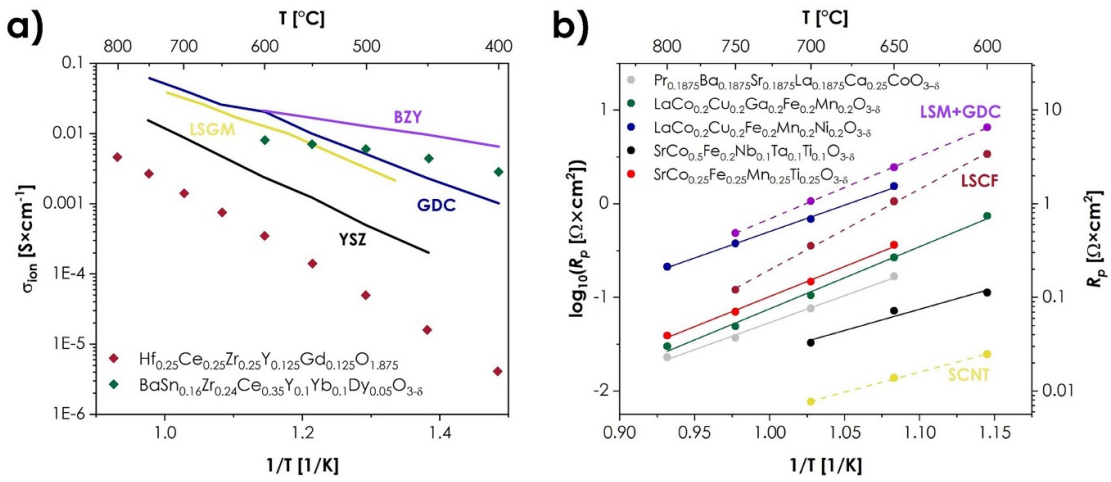

Figure 8. (a) Ionic conductivity of representative compositionally-complex oxides: oxygen ion-conducting, fluorite-structured $Hf_{0.25}Ce_{0.25}Zr_{0.25}Y_{0.125}Gd_{0.125}O_{1.875}$ [167] and proton-conducting $BaSn_{0.16}Zr_{0.24}Ce_{0.35}Y_{0.1}Yb_{0.1}Dy_{0.1}O_{3-\delta}$ (total conductivity value) [168], compared to the ionic conductivities of state-of-the-art solid electrolytes [170]; (b) Cathodic polarization resistance of the selected compositionally-complex perovskite cathodes [171–175]. Values for typical state-of-the-art materials: SCNT ($SrCo_{0.8}Nb_{0.1}Ta_{0.1}O_{3-\delta}$) [176], LSCF ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$) [177], and LSM ($La_{0.8}Sr_{0.2}MnO_3$) [178] are provided for comparison.

exploiting groups of materials which previously have not been considered of interest, such as spinels [179]. Furthermore, what seems to be a common denominator for nearly all HEOs compositions studied is their exceptional stability under operating conditions [163, 164]. As a result, it can be argued that compositionally-complex oxides are an attractive and, above all else, feasible alternative to the materials currently used, for both conventional SOFC and Proton Ceramic Fuel Cells (PCFCs). However, we are still lacking basic understanding of the origins of the aforementioned effects, and therefore means to effectively design HEO electrodes without the extensive trial-and-error screening procedures.

Advances in science and technology to meet challenges

As mentioned, the number of possible combinations and the presence of highly unpredictable synergistic effects make the design of HEOs a particularly challenging process, as well as a time- and resource-consuming one. In the conventional approach, the design of functional materials for energy applications can be successfully aided by *ab initio* modeling, in recent years combined with the use of ML. However, as of today, the use of *ab initio* methods is still somewhat limited with regard to high-entropy materials, because their inherent chemical complexity results in an excessive number of atoms in the calculated unit cells and their possible arrangements, leading to prohibitively high computational demands. The development of new approaches and acceptable simplifications should be considered a priority in this context. Furthermore, the more widespread utilization of first-principle descriptors [180] combined with their further optimization, would also contribute greatly to the effectiveness of the theoretical procedures. The next problem is our general lack of understanding of the exact nature of synergies occurring within the HEOs systems, as in most cases the exclusive focus of studies is placed on the 5- and higher component systems, which are already too complex for a basic analysis. The possible solution for these issues could be a more extensive application of ML. This would require increased availability of the experimental data for simpler low- and medium-entropy subsystems which, together with the *ab initio* results for such compositions, would provide feasible learning sets for the ML algorithms. This in turn would enable much more effective screening of the possible elemental combinations, especially with regard to non-equimolar compositions, which are omitted in most of the experimental reports, despite their likely superior, non-compromised performance.

Experimental developments should closely correlate with the theoretical advancements, providing the necessary data for verification of the theoretical models. It can be expected that a wider application of the atomic-level probes, including the synchrotron radiation methods, would be required to provide valid reference points in this regard. Lastly, it should be noted that currently most of the available data concern the laboratory scale, short-term measurements. As the main advantages of HEOs appear to be in the area of long-term stability under operating conditions of the cells, long-term studies in the environment of the actual stacks, rather than simple button-type cells, appear to be the most logical step going forward.

Concluding remarks

Despite the relatively short timeframe of their development, it can be said that the HEOs have already proved to be a viable option for replacing the state-of-the art SOC materials, often offering the combination of performance and functional features unavailable to their more conventional counterparts. Their further evolution should focus on the development of fast screening procedures and protocols, enabled through an in-depth understanding of their structure-composition-properties relationships. In this context, it should be expected that the role of non-equimolar combinations, as well as simpler, medium-entropy compositions, will constantly increase over time, since the ultimate goal of the compositionally-complex SOC materials should be to preserve and maximize the apparent benefits of their multicomponent character (and the arising synergies) at minimum cost, rather than creating compositions heavily compromised by strict adhering to the general, and to a large degree arbitrary guidelines of HEOs design.

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9. HEMs for electrocatalysis, catalysis and photocatalysis

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Status

HEMs such as oxides, carbides, borides, sulfides, nitrides, silicides, and alloys are composed of multiple principal elements in near-equiatomic or varied proportions and exhibit the well-recognized high-entropy, lattice-distortion, sluggish-diffusion, and cocktail effects [181], making them suitable for a wide range of catalytic applications. These properties are particularly advantageous for electrocatalysis, thermocatalysis, and photocatalysis, where multi-step reactions and harsh operating conditions demand robust and tunable materials (figure 9). The concept of HEMs emerged in the early 2000s, initially in the field of alloys and metallurgy. However, their application in catalysis and energy conversion processes has rapidly gained attention due to their potential to overcome limitations of traditional catalysts, which often suffer from stability issues, low efficiency, or limited scalability. As researchers have progressively uncovered the intricate relationship between entropy, atomic structure, and performance, HEMs have become pivotal in addressing these challenges. The continued importance of HEMs in electrocatalysis, catalysis, and photocatalysis is largely attributed to their multi-elemental composition, which allows for the fine-tuning of material properties [182]. In this regard, distributed surface chemistries create quasi-continuous adsorption-energy spectra that better accommodate multistep mechanisms than single-component catalysts. Additionally, entropy-stabilized frameworks and sluggish diffusion help suppress phase segregation and dissolution under operating conditions, offering unique advantages for long-term stability [181, 182]. These properties can overcome some of the limitations of conventional catalysts, such as limited stability, low activity, and susceptibility to poisoning.

In electrocatalysis, HEMs offer exceptional properties including tailorable electronic structures, synergistic effects from multiple elements, and inherent stability under harsh electrochemical environments. In addition, HEMs can overcome some of the limitations of conventional catalysts, such as limited stability, low activity, and susceptibility to poisoning. Furthermore, the high versatility of HEMs allows for the design of catalysts that can be optimized for specific reactions, thus making catalytic processes more efficient and cost-effective. HEMs and CCMs have been explored for oxygen evolution and reduction (OER/ORR) in water electrolyzers, fuel cells, and metal–air batteries; for CO_2 reduction (CO2RR) toward CO, formate, and C_2^+ products; and for hydrogen evolution/oxidation (HER/HOR) in alkaline cells. Their multi-site synergy and composition-tunable adsorption energies underpin both activity and durability [181–183]. In thermocatalysis, HEMs and CCMs, such as HEAs supported on oxides and halite/spinel/perovskite HEOs, have demonstrated improved resistance to coking and sintering and stable turnover under redox transients in DRM, CO oxidation, and selective hydrogenations, leveraging oxygen-lattice buffering and robust ensemble effects [16]. In photocatalysis and photo-electrocatalysis, by designing HEMs with tailored compositions, researchers can fine-tune the material properties to optimize photocatalytic activity, such as absorption of light across a broad spectrum, efficient charge separation, and increased chemical stability [184, 185]. Entropy-engineered chalcogenides and oxides provided band-edge tunability and defect-tolerant transport, while HEAs acted as co-catalysts that accelerate interfacial charge transfer for solar water splitting and CO_2 conversion [183].

As research in this field progresses, further advances in HEMs catalysis are expected to unlock new opportunities in energy conversion, environmental remediation, and the production of chemicals from renewable feedstocks. By leveraging the unique configurational entropy and multi-element synergies of HEMs, future investigations aim to pioneer the development of highly efficient, durable, and economically viable catalysts tailored to meet the demands of emerging industrial and environmental challenges.

Current and future challenges

While the potential of HEMs in electrocatalysis, catalysis, and photocatalysis is undeniable, major issues must be addressed to fully realize their practical applications. These challenges span across various aspects of HEMs research, from their synthesis and characterization to their performance and scalability. A key obstacle is establishing a clear link between composition, active motifs, and catalytic function due to typical HEM short-range order, surface segregation, and dynamic reconstruction under operando conditions [181, 182]. Static scaling analyses on idealized sites are insufficient to capture the complex ensemble effects inherent to multi-component systems. Furthermore, achieving uniformity in HEM properties remains difficult due to the stochastic distribution of elements during synthesis. Current methods often lack the precision required to control composition and structure in CMMs [186] Beyond

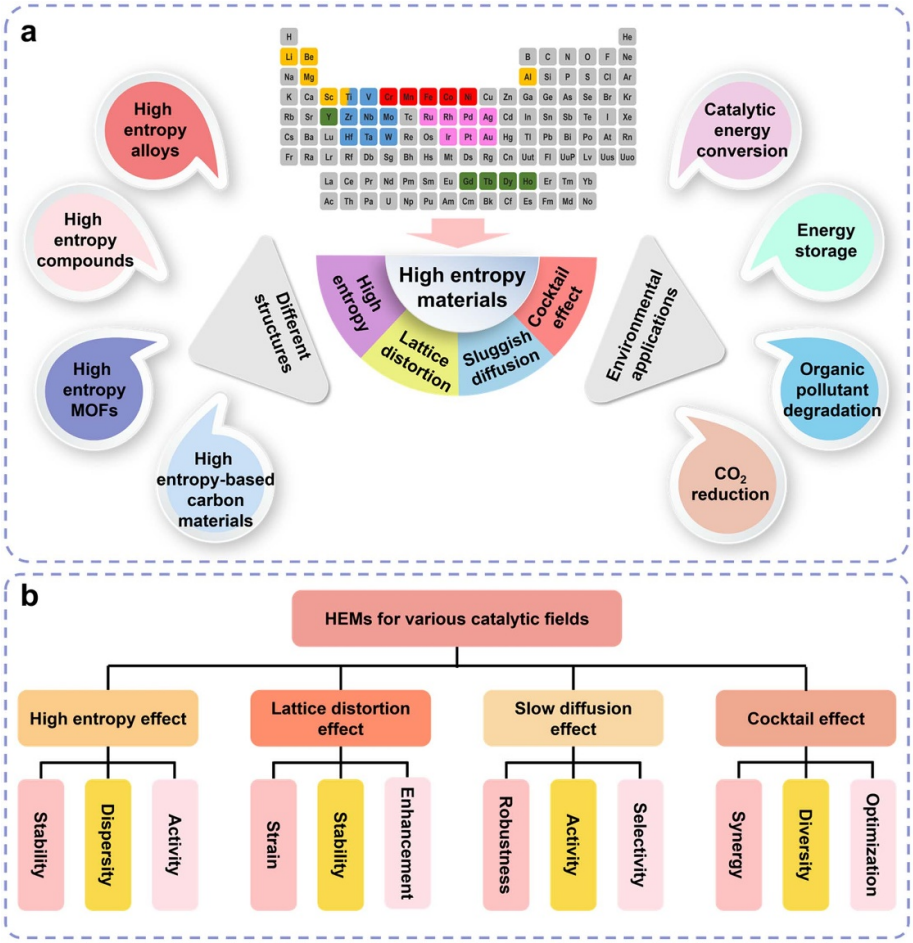

Figure 9. (a) The diverse applications of HEMs, highlighting their unique properties, with a particular focus on their environmental applications. (b) Factors of HEMs affecting the catalytic performance in various catalytic fields.

architectural constraints, traditional synthesis routes are generally unsuitable for large-scale production, underscoring the need for advanced techniques that ensure both consistency and reproducibility.

Another major goal of the community is understanding the synergistic effects between the different elements in HEMs. The HE in these materials leads to complex atomic interactions and predicting how these interactions affect the material's properties is not straightforward. While computational methods such as DFT and ML were used to predict the stability and behavior of HEMs, they fail to capture the full complexity of these systems [187]. Further research is needed to develop more accurate models that can predict the functional properties of HEMs. This will require a deeper understanding of the atomic and electronic structures of these materials, as well as the role of defects, grain boundaries, and other structural features that may influence their performance.

For HEMs to be viable in real-world applications, they must demonstrate long-term stability and durability, especially under harsh operating conditions [186]. Although HEMs offer promising thermodynamic stability, their performance can deteriorate over time due to corrosion, sintering, or the loss of active sites. Ensuring durability in realistic environments, such as alkaline or near-neutral electrolytes for OER/ORR/CO2RR and HT redox cycling for DRM, requires quantifying selective leaching, coarsening, and support interactions, even though multi-element dilution and sluggish diffusion often mitigate these effects [182]. Benchmarking efforts are further complicated by inconsistent testing protocols (electrolyte composition, loading, IR compensation, gas purity), which obscure genuine entropy-enabled advantages. Establishing minimum reporting standards and implementing agreed accelerated durability tests are essential to enable fair comparisons [181, 182]. Ultimately, identifying and mitigating degradation mechanisms will be essential for the successful commercialization of HEM-based catalysts.

The environmental impact of HEMs, particularly in terms of the materials used in their synthesis, is another concern. Many HEMs rely on rare or expensive elements, which could limit their widespread adoption. The extraction and use of these elements may also raise sustainability issues. Research into finding alternative, more abundant elements for HEMs is crucial, as is the development of methods for

recycling and reusing these materials once they have been deployed in industrial applications. Scalability and sustainability also demand earth-abundant element sets, ink-to-electrode transferability for high-loading electrodes, and recyclability-by-design strategies to avoid end-of-life lock-in. Looking forward, future research in HEMs will address these issues by developing more robust and scalable synthesis techniques, optimizing material performance through computational and experimental studies, and ensuring the sustainability of these materials for large-scale industrial applications.

Finally, despite the interest in accelerated discovery of new compositions, this is constrained by data incompleteness and the fact that negative results and process metadata are rarely reported, limiting the effectiveness of ML approaches for navigating high-dimensional compositional spaces.

Advances in science and technology to meet challenges

To address the challenges facing HEMs, significant advancements in both science and technology is essential. Advanced characterization techniques, such as x-ray diffraction, scanning electron microscopy, and synchrotron-based techniques, remain critical to better understand the atomic-level structure and stability of HEMs. These tools provide insights into the phase transitions, elemental distribution, and catalytic activity of HEMs, which are essential for designing more efficient and durable catalysts [16]. Complementing these efforts, operando multimodal characterization, including XAS, XPS, Raman under bias, environmental TEM, atom probe tomography, and ToF-SIMS, now enables real-time observation of transient reconstruction, segregation, and near-surface stoichiometry that govern catalytic turnover and degradation [182].

Computational models and simulations are poised to play a crucial role in guiding the design of HEMs (figure 10). By leveraging DFT, ML, and high-throughput screening, researchers can predict promising elemental combinations and optimize properties for specific catalytic applications. Descriptor learning is evolving beyond single-site scaling, incorporating adsorption-energy distributions, strain and ensemble effects, and generative models aligned with the multi-site nature of HEMs [181, 182]. Closed-loop, active-learning workflows prioritize informative experiments and converge with far fewer trials while maintaining physical interpretability [71, 76]. These approaches, combined with combinatorial and high-throughput platforms such as thin-film microlibraries and micro-electrochemical scanning, have matured to map composition–activity–stability landscapes reproducibly and at scale, enabling data-guided selection of robust optima [70, 72].

Advances in synthesis techniques are equally critical to improve scalability and reproducibility of HEMs. Emerging methods such as additive manufacturing, atomic layer deposition, and liquid-phase processing could offer new pathways to fabricate HE catalysts with precise control over composition and structure. Hybrid strategies that integrate HEMs with nanomaterials or tailored supports can further enhance catalytic performance and stability. Support and interface engineering help stabilizing dispersion, modulating local reaction micro-environments, and improving charge transfer, bridging the gap between discovery catalysts and high-loading electrodes or structured reactors. In electrocatalysis, increasing active site density and ensuring efficient charge transfer remain crucial, achievable through doping, surface functionalization, and electronic structure modulation [188]. For photocatalysis, advances in controlling charge separation, carrier dynamics, and light absorption are essential; techniques such as femtosecond laser spectroscopy and time-resolved spectroscopy provide valuable insights into charge dynamics, enabling rational design of HEMs with optimized properties. Integrating HEMs with advanced materials such as quantum dots or carbon-based nanostructures, offers additional opportunities to boost photocatalytic efficiency.

Finally, sustainability considerations must be embedded in these technological advances. Developing cost-effective and environmentally responsible synthesis methods for large-scale production, along with recyclability-by-design strategies, will be key to widespread adoption. In parallel, adoption of community protocols for OER/ORR/CO2RR and thermocatalysis durability will enable honest benchmarking and accelerate down-selection of scalable formulations [181, 182]. By combining advanced synthesis techniques, enhanced characterization, and data-driven design, the potential of HEMs in catalysis and energy applications can be fully realized.

Concluding remarks

High-entropy materials offer immense potential in the fields of electrocatalysis, catalysis, and photocatalysis, with the ability to address some of the most pressing challenges in energy storage, conversion, and environmental sustainability. While there are still significant challenges to overcome in terms of synthesis, scalability, and understanding their complex behaviors, advances in computational techniques, high-throughput screening, and *in-situ* characterization hold great promise for unlocking their full potential.

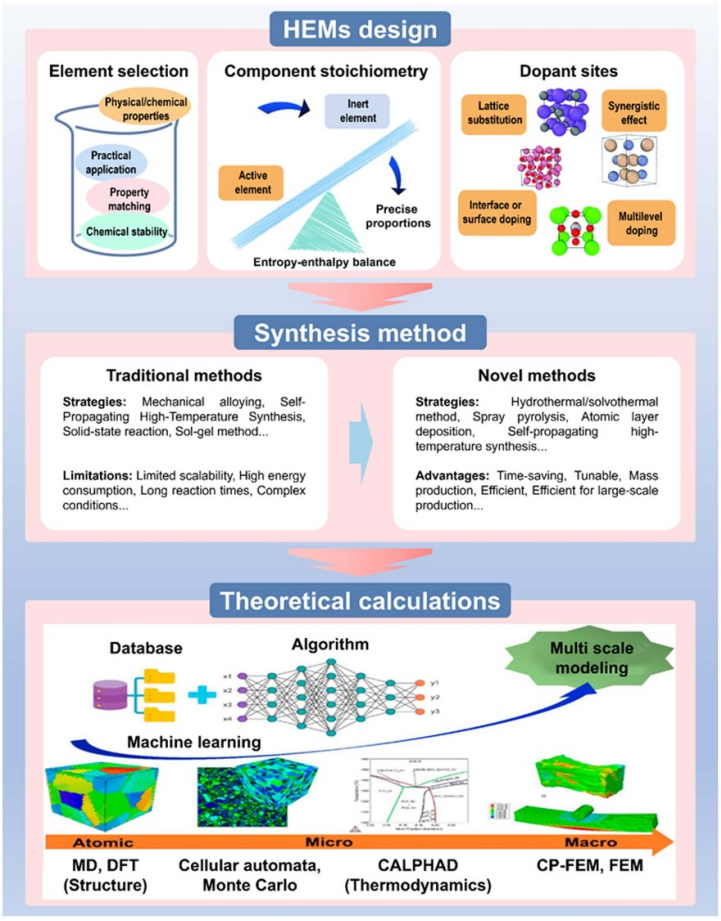

Figure 10. Overview of general approaches to the study of HEMs.

HEMs have evolved from a conceptual framework to application-driven screening across key reaction families, including OER, ORR, HER, HOR, CO_2 reduction, dry methane reforming, CO oxidation, selective hydrogenations, and solar-driven conversions. In the near term, establishing minimum reporting and durability standards, while prioritizing earth-abundant and recyclable chemistries, is essential. Looking further ahead, the focus will shift toward designing advanced compositions validated under operando conditions and tailored to device-relevant loadings and architectures, enabling systems that surpass current lifetime-efficiency benchmarks while ensuring recovery and recycling.

As research in HEMs continues to evolve, these materials are poised to play a crucial role in the development of next-generation technologies that can help solve global energy and environmental challenges.

Acknowledgments

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10. HEA for Hydrogen Storage

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Status

Hydrogen, as a clean and sustainable energy carrier, has the potential to enable the energy transition towards net-zero emissions. Along the production-storage-transportation-utilization chain, safe and efficient HS is the most challenging [189]. Among various HS strategies, solid-state storage in the form of high-capacity metal hydrides is one of the most compelling alternatives to compressed gas to improve the safety, efficiency, and compactness of systems. Metal hydrides can accommodate hydrogen atoms in the interstitial sites of the lattice, which makes the structure type and volume important factors in determining the storage capacity [189]. Hydrides of many transition metals and conventional alloys/intermetallics exhibit high capacity up to 2 Hydrogen per metal atoms (H/M) under moderate pressure and temperature conditions. Nonetheless, this advantage is frequently offset by unfavorable thermodynamics, i.e. high stability of the hydride resulting in elevated desorption temperatures, impeding the reversibility at moderate temperatures [190]. Despite many advances in this topic, up to now, none of the traditional metals/alloys/intermetallics fulfill the requirements for competitive storage media.

Alloying is recognized as one of the most effective strategies to enhance physical properties of materials. In contrast to conventional alloying, chemically complex and HEAs provide a vastly extended playground to discover new high-capacity metal hydrides. In the last two decades, studies on hydrogen absorption/desorption properties of HEAs were growing exponentially, with a focus on body-centered-cubic (BCC) alloys and hexagonal C14 Laves intermetallic phases [191–193]. The aim is to disclose the chemical composition—structure—storage properties relationship via addition/substitution of non-hydride forming and light-weighted elements. Multiple reports were devoted to tuning the thermodynamics of HE hydrides towards an optimum destabilization while minimizing the capacity loss [194, 195]. Besides, several studies addressed the maximization of reversible capacity during absorption/desorption cycling, the minimization of hysteresis between absorption and desorption reactions, and the control of H-induced defects into the lattice [196, 197]. However, the usual trial-and-error experimental approach makes the discovery of materials in the vast chemical space of HEAs challenging. Therefore, data-driven machine/statistical learning (ML) approaches supported by accurate first principles computational methods DFT have been proposed to successfully target HEAs with desired storage properties [195, 198, 199]. Despite many recent advances, fundamental understanding is still required to maximize the potential of HEAs and to bridge the gap towards real-life devices.

Current and future challenges

Several promising BCC and C14-type HEAs with capacity approx. 2 and 1 H/M, respectively, have been reported [192], however a clear understanding of the relationship between composition—structure—functional performance is still lacking. This is due to entangled correlations between functional and physicochemical parameters, including the exceptionally large compositional space, trade-offs between capacity and thermodynamics, and difficult to control degradation phenomena upon extended cycling.

The exploration of HEAs faces the vast number of elemental combinations in the compositional space, with more than 30 elements considered for this purpose [191]. Consequently, there is a need for computational methods, including data-driven ML and DFT approaches, to predict and rationalize correlations among properties and to extract straightforward descriptors. A strong dependence of the thermodynamics of metal hydride on a simple volume-based descriptor that can be computed from just the elemental composition without any knowledge of crystalline structure was identified by using an interpretable gradient boosting tree algorithm, irrespective of hydride phase [198]. Another recent ML study revealed that the thermodynamics of the first and second steps of absorption in BCC HEAs are correlated to the bulk modulus and the number of states in the d-band, respectively [199].

Metals can be classified as high- and low-affinity towards H as function of negative or positive enthalpy of hydride formation, respectively [192]. The use of high-affinity metals warrants high capacity, whereas the addition of low-affinity elements allows to tailor the enthalpy of HE hydride but often involves a capacity loss. Thus, HEA's selection is governed by compromises between thermodynamics, kinetics and capacity. For example, Al and Mo additions in the BCC TiVNb alloy (2 H/M) do not affect the kinetics while thermodynamics can be destabilized on the expenses of capacity [200].

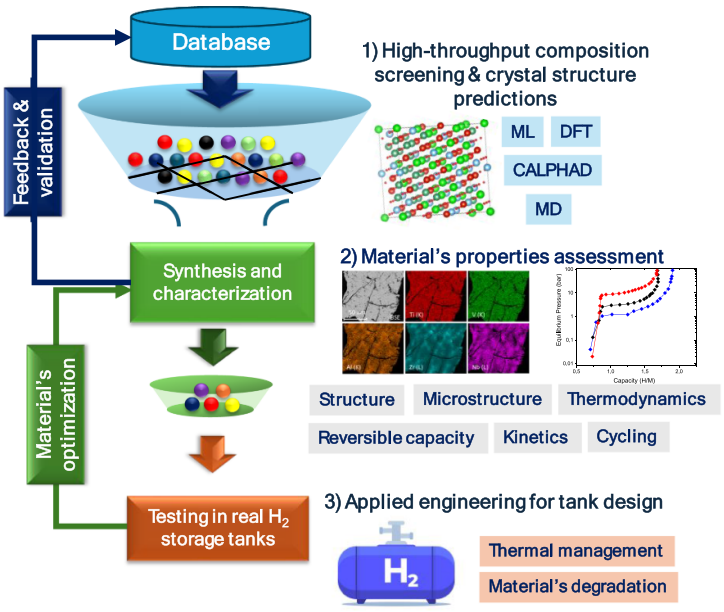

Figure 11. HEA's design strategy for development and integration in HS tanks : high-throughput composition screening & crystal structure predictions using a large variety modeling tools (CALPHAS, ML, DFT, MD), followed by materials property assessment (synthesis, physicochemical, hydrogen absorption/desorption and cycling properties) and testing under real-life scenarios (thermal management and material's degradation during extensive cycling need to be considered).

As HEAs have only recently been proposed for HS [191], research has mainly concentrated on maximizing storage capacity and fine-tuning thermodynamic properties, since activation is usually straightforward. However, cycle-life stability, reversibility, and degradation are essential features for real applications, irrespective of the material. Formation of many dislocations and vacancies have been related to the capacity loss during cycling in TiVCrNb [201]. Moreover, local structure and atomic environment are essential for the preferential H occupancy of interstitial sites and consequently, for the reversible capacity [202]. Thus, identification of major contributions to material's degradation and capacity loss will enable the exploration of compositional modifications to minimize defect formation and dynamics and to maximize cycling performance.

Advances in science and technology to meet challenges

The most promising advances that will push HEA's towards applications include development of high-throughput composition screening & crystal structure predictions using a large variety of modeling tools including computational thermodynamics CALPHAD, data-driven ML, quantum-mechanical DFT and molecular dynamics, followed by materials property assessment and testing under real-life scenarios (figure 11).

Recent modeling and experiments suggest that to address the thermodynamic and capacity challenges associated with using HEAs for HS, it is necessary to investigate complex interdependencies and correlations between various materials properties and performances [203, 204]. The valence electron concentration seems to be an important factor affecting the hydrogen capacity for both BCC alloys and Laves type intermetallics [192, 205]. Other entangled properties are involved, such as bulk modulus [199], local structure [202], electronegativity [195], and atomic radius mismatch with larger values leading to greater local distortion and reduced capacity [192]. The development of analytical semi-empirical models for prediction of hydride's thermodynamics [203, 206] is also desirable, in particular addressing enthalpy-entropy compensation effects [207]. Consequently, computational methods will certainly enable extracting useful descriptors and correlations among properties that will rationalize our basic understanding and predict optimum HEAs compositions for HS.

The degradation of cycling performance is a general metal hydride issue due to large volume expansion during H-induced phase transformation. In this respect, advanced ex situ and *in situ* experimental methods will help clarifying the H-induced defects formation and dynamics, while numerical methods are less widespread to tackle this issue. Positron annihilation spectroscopy and Doppler broadening measurements can provide important insights into defects formation that degrades the cycling

properties. Along these lines, it would be useful to explore whether small additions of elements could enhance the stability while minimizing the capacity loss.

The use of state-of-the-art experimental techniques such as, atom probe tomography and large-scale instruments (synchrotron radiation, neutron sources), will enrich our fundamental knowledge. Besides, new preparation and processing tools such as, severe plastic deformation [208] should be explored to achieve better control over local composition and microstructure.

Finally, future research directions in the HEA's arena for HS should include the use of earth-abundant elements to decrease the cost and the availability of raw elements, coupled with the development of greener and less energy intensive alloy preparation methods. The resolution of these challenges will allow to bridge the gaps between fundamental understanding and applied engineering towards practical tank design based on HEA's.

Concluding remarks

HEAs are promising candidate materials for HS due to their unique structural and chemical properties, which could be tuned in a wide range by selecting synergistic combinations of 'hydriding' and 'non-hydriding' elements. Recent studies revealed intricate relationships between composition, structure, atomic size of involved elements, number of valence electrons, electronegativity and HS performance. A key challenge is optimizing HEA thermodynamics to achieve the best balance between destabilization and capacity, while also addressing degradation during cycling, hysteresis minimization, and hydrogen-induced defects. The vast compositional space of HEAs offers tremendous opportunities for data-driven ML and DFT approaches to predict material properties and identify effective descriptors of performance. For example, ML studies have demonstrated that the thermodynamics of hydrogen absorption in HEAs correlate with a volume-based descriptor computed from just the elemental composition. Despite these advances, fundamental understanding remains incomplete, particularly concerning the complex interplay of physicochemical parameters, such as atomic size mismatches and enthalpy-entropy compensation effects. Addressing these challenges will require the application of high-throughput screening methods, innovative preparation techniques such as additive manufacturing, and *in-situ* characterization techniques to bridge the gap between theoretical research and the practical implementation of HEAs in HS tanks.

Acknowledgments

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11. HE thermoelectric materials and applications

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Status

Thermoelectric (TE) technology, which enables the direct conversion between thermal and electrical energy, is regarded as a promising solution for addressing the energy crisis through waste heat recovery. TE materials exhibit substantial potential for applications in power generation and refrigeration systems. The power generation efficiency (η) of thermoelectric generators, and the coefficient of performance (COP) of thermoelectric coolers, are both determined by the average figure of merit ($Z\bar{T}$) of thermoelectric materials,

$$\eta = \frac{T_h - T_c}{T_h}\frac{\sqrt{1+Z\bar{T}}-1}{\sqrt{1+Z\bar{T}}+\frac{T_c}{T_h}}$$

$$\mathrm{COP} = \frac{T_h}{T_h - T_c}\frac{\sqrt{1+Z\bar{T}}-\frac{T_h}{T_c}}{\sqrt{1+Z\bar{T}}+1}$$

where, T_h and T_c represent the hot-side and cold-side temperature, $(T_h–T_l)/T_h$ is Carnot efficiency. Besides, figure of merit of TE materials can be defined as $ZT = S^2\sigma T/\kappa$, where S is Seebeck coefficient, σ is electrical conductivity, κ is thermal conductivity, and T is the absolute temperature. As depicted in figure 12, when $Z\bar{T}$ value surpasses 2, the conversion efficiency can approach that of conventional heat engines (approximately 25%). Therefore, rational thermoelectric design is the key to achieve a high $Z\bar{T}$ to secure high η and COP. Traditionally, the effect of increasing ZT value is achieved through band structure regulation [209] and phonon scattering enhancement [210].

In recent years, entropy engineering has been proven to be an effective method for improving the performance of TE materials, providing a novel approach to optimizing both electrical and phonon transport properties. The configurational entropy (ΔS) can be expressed as $\Delta S = -N_A k_B \sum_{i=1}^{n} x_i \ln x_i$, where N_A and k_B are Avogadro's number and Boltzmann constant, respectively, n is the number of substituted components, and x_i is the mole content of the ith component. Therefore, increasing the compositional complexity of a material enhances its configurational entropy, which in turn can improve the symmetry of its crystal structure. Materials with highly symmetrical crystal structures possess numerous equivalent sites in both real and reciprocal spaces. This facilitates the formation of highly degenerate band edges and multi-valley electron pockets, which are directly linked to the symmetry of the material. Such features contribute to an increase in the effective mass and Seebeck coefficient [211], ultimately leading to superior electrical transport properties. On the other hand, when multiple elements coexist at the same lattice position, the differences in their atomic mass and radii introduce fluctuations in both the mass and stress fields. These fluctuations cause solute atoms to act as scattering centers for point defects, disrupting the propagation of high-frequency phonons and subsequently reducing the lattice thermal conductivity [212]. Simultaneously, entropy engineering can trigger dynamic hysteresis effects, leading to lattice distortions in local microscopic regions. These nano- to micron-scale lattice distortions, in conjunction with point defects, establish a multi-level microstructure through the material. This structure enables broadband phonons scattering, significantly lowering the material's lattice thermal conductivity (κ_L). At present, the high-entropy strategy has demonstrated remarkable success in several material systems, including IV–VI compounds [212, 213], semi-Hessler alloys [214], liquid materials [215], and oxide-based ceramics [216] (figure 13).

Current and future challenges

The primary challenge in the entropy engineering applications lies in synthesizing stable single-phase materials with high configurational entropy. Introducing multiple components not only elevates configurational entropy but also boosts the enthalpy of the material due to enhanced lattice distortion

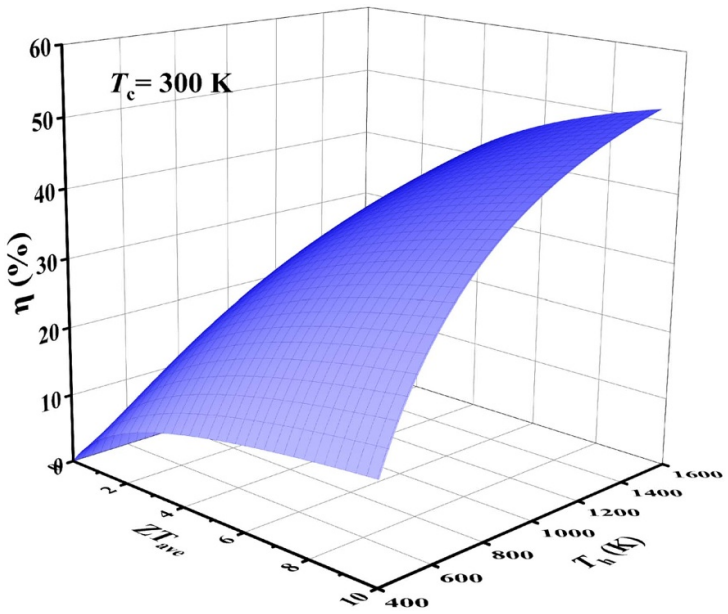

Figure 12. The relationship between thermoelectric conversion efficiency and the average figure of merit ($Z\bar{T}$), hot-side temperature.

energy, often leading to thermodynamic instability. The scenario becomes even more intricate for multicomponent systems where three or more elements coexist at the same atomic site. Traditional trial-and-error approaches to preparing single-phase solid solutions require extensive resources, including significant manpower, time, and materials. Currently, predictive models for single-phase HE solid solutions, encompassing both parameter-based and free energy-based models, have achieved a maximum accuracy of only 72% [217]. This limitation underscores the urgent need to develop efficient stability prediction techniques tailored to multicomponent single-phase high-entropy TE materials. Simultaneously, based on the unique characteristics of each material system, it is necessary to design the optimal solid solution elements and solid solution amounts in a targeted manner. To facilitate the broader application of entropy engineering, it is essential to develop advanced prediction models. This includes creating more accurate computational frameworks that integrate thermodynamic and kinetic factors to predict material stability. Additionally, optimizing solid solution design is essential to tailor the selection and concentration of solid solution elements based on the unique properties of each material system, ensuring a balance between configurational entropy and thermodynamic stability. HEMs form strong lattice distortions and localized strain fields by randomly occupying lattice sites with multiple elements, significantly enhancing phonon scattering and reducing κ_L [218, 219]. However, the inherent structural disorder also acts as a scattering center for charge carriers, leading to a significant decrease in carrier mobility [220, 221]. Therefore, another key challenge in entropy engineering is to maximize the Seebeck coefficient, reduce κ_L, and minimize the adverse impact on carrier mobility. Achieving this balance requires a more refined understanding of how entropy influences both electrical and phonon transport. Further research is needed to elucidate the doping mechanisms and atomic arrangements in high-entropy materials, ultimately identifying the optimal trade-off between carrier and phonon scattering for enhanced TE performance.

It is worth noting that current study focuses on optimizing the performance of TE materials by leveraging the high-entropy effect. However, achieving high TE conversion efficiency requires not only high-performance TE materials, but also sophisticated TE module designs. Among these, selecting the appropriate interface material is critical, as it directly impacts thermal and electrical contact resistance, mechanical stability, and long-term durability.

Advances in science and technology to meet challenges

As previously discussed, the preparation of high-solubility single-phase solid solutions, as well as the identification of suitable solvable elements and their respective amounts, demands significant time and trial-and-error costs when relying solely on conventional trial-and-error methods. Notably, entropy, as a fundamental parameter, exhibits intrinsic gene-like characteristics, making it a key factor in material design. Materials Genome Initiative (MGI) model, such as high-throughput computation, database

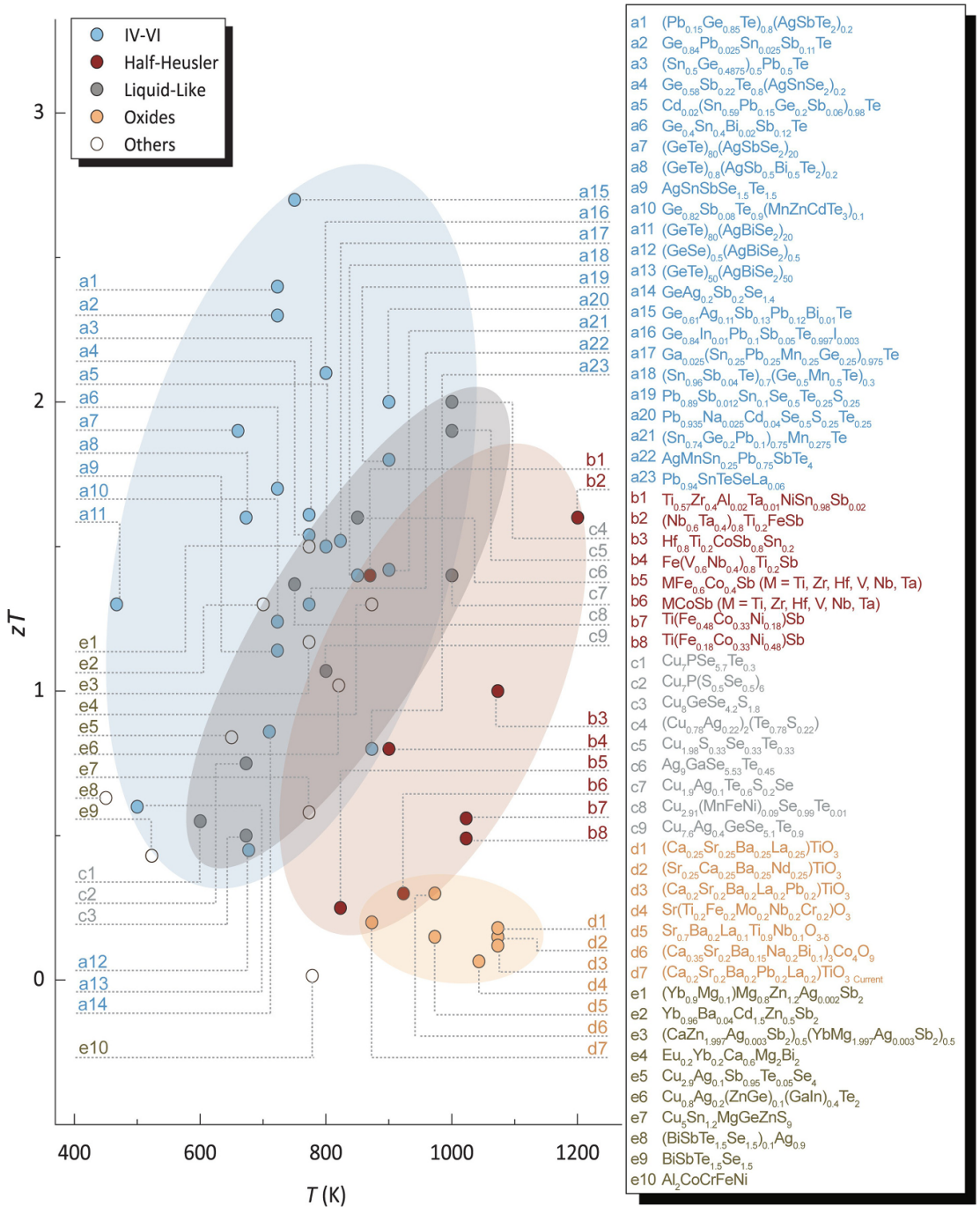

Figure 13. Peak ZT values of the typical high-entropy thermoelectric materials. Reprinted from [211], Copyright (Year), with permission from Elsevier.

construction, and ML, have expedited material discovery and design by integrating theoretical model, computational simulations, and experimental validation [222]. Consequently, leveraging the synergy between high-entropy TE materials and MGI methodologies will greatly accelerate the discovery and application of high-performance TE materials. To enhance exploration efficiency, the incorporation of high-throughput experimental techniques is essential, enabling the rapid screening and optimization of novel TE materials.

Moreover, the inherently homogeneous single-phase structure of high-entropy TE materials enables the growth of large-sized single crystals. The high-entropy effect and sluggish diffusion kinetics further support the feasibility of single-crystal growth, allowing these materials to retain a single-phase structure with exceptional thermodynamic stability. In this context, advanced characterization techniques, such as angle-resolved photoemission spectroscopy and inelastic neutron scattering, can provide deeper insights into their electrical and thermal transport mechanisms, thereby guiding efforts to decouple thermal and

electrical transport [223]. Additionally, three-dimensional reconstruction techniques based on high-resolution STEM offer novel perspectives on the microstructure of high-entropy TE materials, further directing the optimization of their TE transport properties.

Currently, the integration of TE materials optimized for distinct temperature zones through serial connections has proven to be an effective strategy for maximizing the average *ZT* across the entire operational temperature range. The adoption of high-performance high-entropy TE materials has significantly enhanced the conversion efficiency of TE modules, demonstrating their immense potential for practical applications. For instance, Jiang *et al* designed a segmented module incorporating high-entropy PbSe as one of the HT TE legs, achieving a remarkable conversion efficiency of 12.3% at a temperature difference of 507 K [212]. Subsequently, the researchers replaced all HT TE legs with high-entropy materials, specifically, a p-type $Ge_{0.61}Ag_{0.11}Sb_{0.13}Pb_{0.12}Bi_{0.01}Te$ alloy and a n-type $Pb_{0.997}In_{0.003}Te_{0.996}I_{0.004}$ alloy. This modification further improved the module's performance, achieving a record experimental conversion efficiency of 13.3% under a temperature difference of 506 K [213]. With the exceptional conversion efficiencies demonstrated by current high-entropy TE devices, this technology is poised to advance toward commercial viability.

Concluding remarks

In recent years, high-entropy thermoelectric materials have emerged as a research hotspot in the thermoelectric field due to their unique multi-principal-element design concept. By introducing five or more principal elements to form high-configuration-entropy systems, these materials demonstrate significant potential for reducing lattice thermal conductivity and modulating electronic band structures. However, further performance enhancement of high-entropy thermoelectric materials still requires overcoming core challenges, including synergistic optimization of electron-phonon transport, stability under extreme environments, and low-cost, large-scale fabrication. Looking ahead, with advancements in computational materials science, *in-situ* characterization techniques, and intelligent manufacturing processes, high-entropy thermoelectric materials are expected to achieve broader application and commercialization.

Acknowledgments

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12. HEMs in turbines

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Status

Gas turbines are used for aviation propulsion and industrial power generation. In view of the use of HEM in turbines, the hot section is considered the most promising area. The reasons are: (i) The number of lightweight metals and, therefore, lightweight HEAs competing with Al-, Mg-, or Ti-alloys in the LT sections is limited. (ii) Lightweight HEAs competing with ferritic steels show promising properties, but steels and ceramic composites have pricing that is too competitive. (iii) The turbine's efficiency highly depends on the temperature of the hot gas in the turbine which is most easily increased by introducing materials capable of withstanding higher working temperatures. Commonly, Ni-based superalloys are used, however, gas temperatures exceeding the melting point of the Ni alloys are reached [224]. (iv) These components require thermal insulation as protection against the harsh environment in which context HEOs are discussed. Other HE ceramics such as nitrides, carbides, or borides are not a viable option, as they oxidize when exposed to harsh conditions. To date, thermal barrier coating (TBC) systems consist of a ceramic top coat, typically yttria-stabilized zirconia (YSZ) with low thermal conductivity providing thermal insulation, and a bond coat consisting of MCrAlY (M=Ni, Co, NiCo) providing oxidation and corrosion resistance [225].

The surface temperature of the TBC and, therefore, of the component is limited to 1200 °C in long-term use due to the metastability of the YSZ top coat. The combustion temperature is up to 250 °C higher due to the use of active cooling. However, while the higher combustion temperature results in higher efficiency, active cooling reduces the turbine power. To reduce climate impact, gas turbines are being continuously optimized by turbine design, higher temperatures, and the combustion of alternative fuels. All approaches require new materials that have either lower density to enable lighter turbines, or stability at higher temperatures, or corrosion-resistance to water vapor due to the combustion of sustainable aviation fuels, or all of the above. HEMs are discussed for all three layers in the literature. Refractory metal-based HEAs are being investigated as future materials beyond Ni-based superalloys [226]. Promising alternative TBC materials are HEOs based on ZrO_2 and HE rare earth zirconates; HE aluminates, hafnates, niobates, and cerates, are also being investigated [227]. The developments, advantages, and challenges are discussed in the following chapters and are summarized in figure 14.

Current and future challenges

Considering development of a layer compound (figure 14), on the one hand, each material component must be developed independently, and on the other hand, all materials must ultimately form a complete assembly. In environments exceeding 40% of the material's melting temperature, the microstructure evolution highly depends on diffusion-activated processes, but the microstructure must be stable enough to maintain the material's properties during long-term exposure. This excludes interesting compositions that are metastable at room temperature where the advantageous effect of the elemental mixing is frozen but prone to destabilization by fast diffusion at high temperatures.

Structural alloys: HEAs with significant volume fractions of precipitates or ceramic particles show the best balance of strength, creep resistance, damage tolerance, and corrosion and oxidation resistance [228]. For very high temperatures, the main challenge is to find a composition precipitating a second phase with appropriate phase stability and a strengthening mechanism that is competitive with the Kear-Wilsdorf locks in Ni-based superalloys. Another option is finding an alloy composition capable of resisting such high temperatures that the decrease of precipitate strengthening over homologues temperature is not tremendous. Such alloys are mostly refractory alloys with a ductile to brittle transition temperature due to their body-centered cubic crystal structure [229]. In this case, the main challenge is the development of alloys that are ductile at room temperature, which is relevant for processing.

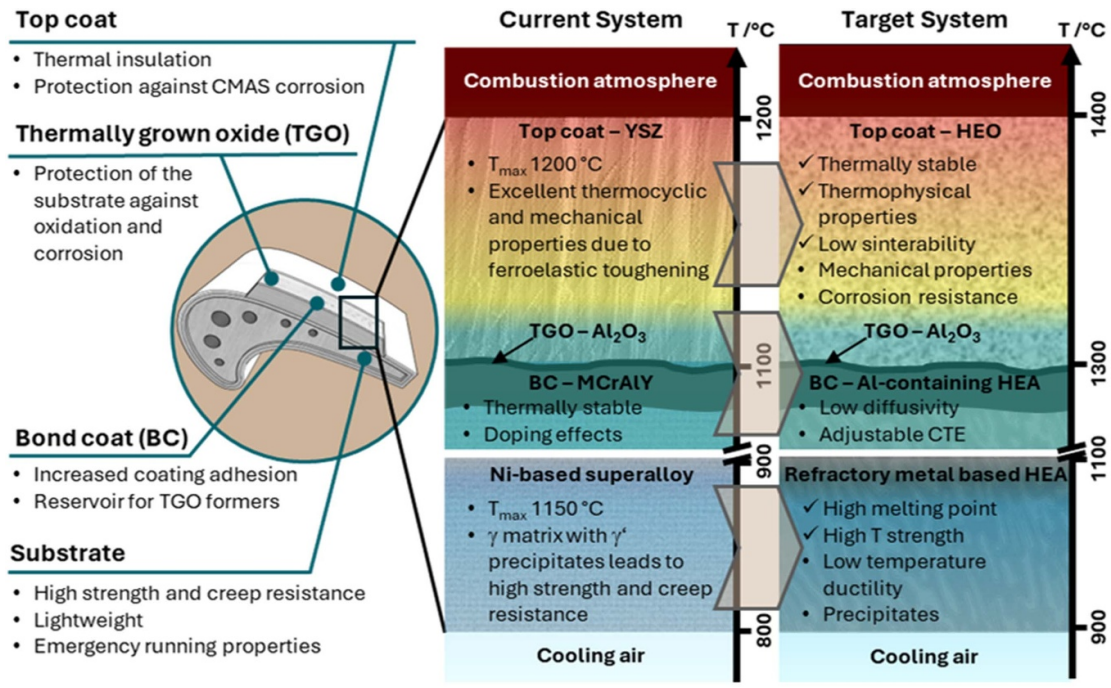

Figure 14. Schematic structure of the parts in the hottest section of a turbine. The requirements for each component are given on the left, the current structure is given in the middle, and the target system is given on the right. In this illustration, all components in the target system are substituted with HEMs; however, the individual compositionally complex components could also be combined with conventional materials in the future.

Bond coat: HEAs are promising bond coats due to their low diffusivities. Most of these systems also aim to form α-Al_2O_3 scales as protective thermally grown oxides which is considered most promising [230]. The future challenge is finding an appropriate composition to link the substrate to the top coat.

Ceramic top coat: HEOs show extremely low thermal conductivities [231] and are thermally stable to high temperatures [232]. The challenge is that their composition and thermal expansion coefficient must match the underlying material—reactions and interdiffusion processes, e.g. with the TGO as well as thermal mismatch should be avoided [233]. Furthermore, HEOs must show adequate mechanical properties, corrosion resistance, and long-term stability without phase transformations to temperatures exceeding 1200 °C as well as low sintering tendency which are known to introduce stresses in the coating that cause spallation. Additionally, HEOs must be affordable with respect to pricing and availability of their containing elements.

If competing with the current state-of-the-art materials, production routes, and scalability must be investigated. For alloys, suitable casting techniques and machinability are needed. For ceramics, the deposition of coatings using industrial processes such as thermal spraying or electron-beam vapor deposition is necessary.

Advances in science and technology to meet challenges

Due to the huge unknown compositional space, high-throughput experimental methods (e.g. diffusion couples, thin-film material libraries, laser metal deposition) and simulations (e.g. thermodynamic and atomistic calculations, empirical correlations) are needed [68]. To speed up investigations, machine-learning-assisted (ML) material development will be used in the future to narrow down necessary experiments and reduce the number of time-consuming tests. Long-term experiments are necessary for HT applications such as turbines to determine thermodynamic stabilities, kinetics, and the related change of the material's properties over time. Besides classical forward simulations and property predictions, ML is also necessary to evaluate the process-structure-property relationships by inverse design. However, high-quality and consistent databases are lacking in executing reliable predictions. Open-source composition-structure-property databases play a huge role in making data available for accelerating ML-aided material development [234]. The quality and comparability of the stored data and determined properties must be guaranteed by measurement standards.

Structural alloys: Refractory metal based HEAs are candidates to be used in the hottest sections due to the high melting points of the base elements and their HT strengths [235]. The primary challenge of room temperature ductility was recently found to be, in principle, possible to overcome [226, 236]. To achieve sufficient creep resistance, appropriate precipitates must be identified. Additionally, alloys with face centered cubic crystal structure developed from the Cantor system can be potentially used as disc or blisc materials [105]. These alloys have lower densities than conventional alloys with higher specific strength up to 900 °C [237]. The first results on oxidation resistance show the ability to form protective Al_2O_3-scales.

Ceramic top coat: HEOs are competitive with the state-of-the-art top coats, as their single-phases are stable at higher temperatures [232] and they show promising resistances against metallic interdiffusion and CaO–MgO–Al_2O_3–SiO_2 (CMAS) attack [238, 239]. Simultaneously, HEOs offer low thermal conductivities in some cases less than 1 W m^{-1} K^{-1} due to their compositional complexity [225, 231]. Recent studies show that HEOs can be processed using the well-established thermal spraying processes used to coat conventional systems [240]. To reduce the environmental impact and production costs of the materials, a feasible strategy would be to use the variety of elements in natural ores for the synthesis of HEOs without having to purify them beforehand.

It should be noted here that individual components can be replaced by HEMs if they have better properties than the conventional materials.

Concluding remarks

HEMs and compositional complex materials (name rather used for multi-phase materials) are highly attractive for applications at high temperatures and harsh conditions, such as those encountered in turbines, as the entropy term gains influence at high temperatures and thus stabilizes the materials thermodynamically. Multicomponent compositions promise new outstanding properties, such as high creep resistance and HT strength in alloys or corrosion resistance and low thermal conductivity in TBCs. However, there is no evidence of huge leaps due to the so-called HE effect in terms of material properties. Nevertheless, promising new compositions have been found, particularly for HEOs, which have very low thermal conductivities and for which the coefficient of thermal expansion could be tailored, an important prerequisite for the application. This shows that each material class has its own challenges, such as the balanced mechanical properties of HEAs or the processability of HEOs. In order to tackle these in a targeted manner, the industry must now partner with the scientific community to identify fields of application and achieve progress in terms of technical readiness. Advances in database development and the use of ML will accelerate the development of new HEMs, to find a way into application.

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13. Conclusions and outlook

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Compositionally complex and high-entropy materials are rapidly transitioning from a fascinating concept to a solid reality with strong impact across multiple application fields, notably energy technologies. This roadmap shows that multi-principal-element compositions offer advantages in catalytic activity, charge transport, structural and chemical stability, and durability across batteries, solid oxide fuel and electrolysis cells, electro- and photocatalysis, HS, thermoelectrics, and turbomachinery. Such advantages arise from intrinsic properties of HEMs and CCMs such as entropy-enabled phase stability, lattice distortion, multi-site synergy, sluggish diffusion, and compositional flexibility, which have already yielded substantial performance gains in harsh operating environments and opened routes to replace critical raw materials with abundant alternatives. However, despite remarkable recent advances, systematic correlations between disorder, local chemistry, and device-level metrics remain incomplete, indicating the need for targeted theory, high-quality data, and operando experimentation to turn current promises into robust technologies.

As detailed in this roadmap, the field now benefits from complementary modeling strategies, such as spectral and supercell approaches and thermodynamic density-of-states descriptors that predict synthesizability and map disorder–property landscapes. These tools reduce design risk in vast compositional spaces but require broader validation for ionic, defect-rich, and multiphase systems central to energy. In parallel, high-throughput experiments based on combinatorial micro-libraries, automated characterization, and closed-loop active learning, have matured to the point that composition-performance-stability maps can be reliably generated, enabling accelerated design of CCMs catalysts, electrode active materials and coatings, among other materials. As a result, the challenge progressively shifts from finding new compositions to verifying microstructures and processing conditions to translate thin-film or micro-library optimized compositions into bulk and device architectures. The chapters dedicated to each energy application in this roadmap highlight these trends towards device implementation of HEMs and CCMs: batteries leverage compositional complexity to improve cycling stability and suppress deleterious phase transitions while enabling superionic conductors; SOCs benefit from HEO cathodes and compositionally complex electrolytes that demonstrate durability and tolerance to CO_2/Cr poisoning; electrocatalysis and photocatalysis exploit multi-site ensembles and quasi-continuous adsorption spectra that couple activity with resilience; HEA hydrides provide tunable thermodynamics and high capacity supported by emerging machine-learning descriptors; entropy-engineered thermoelectrics achieve record module efficiencies by jointly shaping electronic bands and phonon transport; and HEO thermal-barrier coatings deliver ultra-low thermal conductivity and compositional latitude for CTE matching in gas turbines for energy conversion.

Despite the increasing number of articles dedicated to the implementation of complex materials in devices, important knowledge gaps persist, including the origins of improved stability in single-element versus compositionally complex systems, the quantification of defect chemistry, and the characterization of short-range order and surface evolution under operando conditions. Practically, there is also a need for energy- and cost-efficient routes to scale synthesis with microstructural control from nano-sized to micro-sized particles, and for systematic treatment of recycling and sustainability considerations in multi-element materials. Addressing these bottlenecks, together with the generation of standardized durability protocols and performance data to enable rigorous benchmarking, is essential for technology readiness and responsible deployment.

To address these challenges in CCMs and HEMs, we outline research priorities that reflect the collective discussion presented in this roadmap. On the theoretical side, a priority is advancing thermodynamic-kinetic synthesizability through extensions of DEED/POCC frameworks to include defects, interfaces, and finite-temperature phonon contributions, together with uncertainty quantification and transfer learning across different families of complex materials. An excellent complement to theory will be the emerging use of accelerated materials discovery. HEMs are ideally suited for these approaches because their vast compositional space is impossible to fully explore with conventional frameworks. Future closed-loop discovery schemes should integrate active learning, advancing toward the ultimate goal of deploying self-driving labs. To improve model reliability, coherent and comprehensive datasets should be systematically shared in FAIR repositories, including negative results that are

currently missing in the literature. Building on accelerated discovery, a major priority is the development of energy-efficient and scalable synthesis routes with tailoring capabilities for grain size, texture, and defect-density control, with direct translatability from thin-film or micro-library optima to bulk powders and coatings. At the device level, new metrics are needed to quantify the impact of composition, microstructure, and interfaces on performance. Such descriptors will accelerate co-optimization of HEMs under real-world conditions. Equally important is deliberate engineering of interfaces and architectures to mitigate interdiffusion, tailor space-charge regions, and stabilize microstructures under thermal and mechanical loads, coupled with operando multimodal characterization to resolve transport and degradation mechanisms alongside standardized accelerated tests. Finally, sustainability should be embedded by design through element families, phase-selective leaching, tolerant remanufacturing routes, and life-cycle analyses that quantify ecological costs per stored kilowatt-hour or per operating hour, with a clear preference for earth-abundant chemistries.

To conclude, compositionally complex and high-entropy materials have moved beyond curiosity to become a cornerstone of next-generation energy technologies. Their unique ability to combine stability, performance, and flexibility opens unprecedented opportunities for innovation across energy storage and conversion systems. Looking ahead, success will depend on turning complexity into predictability, scaling breakthroughs into practical solutions, and embedding sustainability at every stage. This roadmap is both a snapshot of progress and a call to action. By uniting theory, experimentation, and industrial implementation, CCMs and HEMs can help shape a cleaner, more resilient energy future.

Data availability statement

No new data were created or analysed in this study.

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