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Innovative Synthesis Strategies and Emerging Applications of High-Entropy Materials: A Comprehensive Review

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Abstract

High-entropy materials (HEMs) represent an emerging category of substances harnessing elevated configurational entropy to stabilize multiple elements within a single crystal lattice, resulting in distinctive physical properties suitable for applications in energy storage, and energy conversion. Initially, the fabrication of these materials involved methods that demanded elevated temperatures and extended synthesis durations. Nevertheless, there are still difficulties in successfully blending components at the atomic level inside the lattice, particularly when synthesizing nanomaterials. The rise of HEMs, characterized by their exceptional mechanical properties, stability under high temperatures, and robust chemical resilience, is anticipated to bring about significant progress in the enhancement of energy storage and conversion technologies. HEMs exist in diverse forms including bulk, films, belts, fibers, and powders. Consequently, the preparation methods encompass the fusion-cast process, physical vapour deposition, powder metallurgy, melt spinning, and more, which will be elaborated upon in the subsequent section. This review presented the evolution of synthesis strategies employed for different types of HEMs, elucidating their formation mechanisms. The synthesis techniques covered in this review include mechanical alloying, spark plasma sintering, arc melting, and various solution-based methods such as sol-gel and chemical vapour deposition. Each method is examined in terms of its principles, advantages, and limitations, providing insights into the factors that influence the composition, microstructure, and phase stability of HEMs.

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1. Introduction

Advanced materials have drawn much attention due to their unique properties and applications.[1-4] The two most significant issues facing mankind in the twenty-first century are the efficient and sustainable conversion and storage of energy and environmental preservation.[5] Energy storage is very important for reducing carbon emissions.[1] Changing from using fuels and gases to using sustainable energy.[6] To minimize environmental disasters, many attempts have been made to create innovative technologies for ecologically friendly and reliable energy conversion and storage systems.[7] Nevertheless, the quest for a more sustainable society faces major scientific and technical limitations that will need major contributions from the community of scientists.[8] In actuality, we are looking for devices that can contain a lot higher energy density and generate a lot of high-density power. Hybrid supercapacitors are an excellent choice as they merge the high energy storage capacity of batteries with the powerful performance of supercapacitors in a single device.[9] Batteries such as lithium-ion batteries (LIBs) are used a lot for storing energy and can be found in things like electric cars, phones, and robots.[10-14]

On the other hand, designing strong and high-performance catalysts for electricity is an entirely novel challenge.[15] For example, in recent years, converting renewable energy into high-energy-content chemical species via electrochemical and/or photochemical water-splitting processes has become an extremely hot issue. In this context, one of the main objectives of materials chemistry and nanotechnology is the development of cost-efficient, abundant electrode materials that can catalyze the tetra-protonic and tetra-electronic reactions of the

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oxygen evolution reaction (OER),[16] the hydrogen evolution reaction (HER),[17,18] and the electrochemical reduction reaction of CO_2(CO_2RR).[9]

Entropy was not considered as useful as enthalpy for material design until recently when an idea of high-entropy alloys (HEAs) was proposed. Following some previous ground research, concurrent experiments by Yeh *et al.* and Cantor *et al.*, who created a unique family of multi-component alloys, presented the notion of HEA in 2004. HEAs are multi-principal element alloys with a high configuration entropy that are made up of at least 5 elements with equal or nearly identical atomic fractions.[19,20] High-entropy materials (HEMs) have received increased attention in recent breakthroughs in the development of promising materials for energy technologies due to their unique concept of design and unique characteristics.[21] The HEM family includes several different types of materials, including HEAs, high-entropy oxides (HEOs), high-entropy hydroxides (HEHs), and several more. The idea of high entropy has no limits to traditional inorganic compounds and is not defined just by compositional disorder and element number.[5]

In 2015, the theory of HEOs was initially presented. HEOs have a single-phase crystal structure and are made up of at least five metal cations.[22] Recently, the concept of high-entropy has been introduced into some common coordination materials, such as Prussian blue analogues (HE-PBAs)/hexacyanoferrates (HE-HCFs) and other metal-organic frameworks (HE-MOFs), as well as metal-glycerolates (HE-glycerolates) and metal-polyphenol coordination polymers (HE-PCPs), which may open up new opportunities in energy storage application.[5,23-25] HEOs are projected to bring in a new stage of development for the electrochemical storage of energy systems. The high-entropy and hysteretic dispersion mechanisms can keep the lattice structure and restrict electrode volume growth during the cycles. HEOs with different ion radii cations have intrinsic lattice distortion, which prevents electrode breaking.[6]

The idea of high entropy has also been used with other materials like intermetallic, oxides, sulfides, nitrides, metal-organic framework materials, layered hydroxides, and chalcogenide fluorides. All of these are called HEMs. This review presented the evolution of synthesis strategies employed for different types of HEMs, each method is examined in terms of its principles, advantages, and limitations, offering insights into the factors influencing HEMs composition, microstructure, and phase stability.

2. Theory of high-entropy materials

When studying the analytical behavior of the system's microscopic elements in the 1870s, Ludwig Boltzmann created the word "entropy". According to Boltzmann's proposition connecting the possibility of a particular state's existence to the entropy of a system in that state, one can determine entropy using the following Eq. (1).[26]

$$S = k_B \, ln \, \omega \tag{1}$$

where S is the entropy contribution to the system for the given state, k_B is Boltzmann's constant (1.3×10^{-23} J K^{-1}) and ω is the probability that a given state can exist.[27]

The degree of disorder in a system is indicated by the thermodynamic parameter entropy. The entropy increases with the system's degree of disorder. HEAs are where the term "high entropy" first appeared. Since the configurational entropy of mixing (ΔS_{mix} from Eq. (2)) rises when additional equimolar primary components are added to the alloy system, Yeh *et al.* established the idea of HEAs.[19]

$$\Delta S_{mix} = -R \sum c_i \ln c_i = -R \sum \frac{1}{n} \ln \frac{1}{n} = R \sum \ln n \tag{2}$$

where n is the number of elements, c_i is the concentration of component i and R is the gas constant.[8]

According to this theory, the enthalpy formation (ΔH_{mix}) of intermetallic compounds can be offset by the thermodynamic contribution of high ΔS_{mix} in a system containing at least five equimolar elements to the Gibbs free energy (ΔG_{mix}) at high temperature, forming a single-phase solution of multiple elements ($\Delta G_{\text{mix}} \leq 0$ from Eq. (3)). HEAs consist of five or more elements in equal or very similar atomic ratios, with 5-35% of each element.[19,28] HEAs, compared to normal alloys, are often made of five or more elements in equal or very equivalent atomic ratios, with 5-35% of each element and multi-elemental alloys with $\Delta S_{\text{mix}} > 1.5R$ (entropy-based definition).[29,30]

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \tag{3}$$

Eq. (3) implies that an elevated entropy of mixing (ΔS_{mix}) results in a reduced Gibbs free energy of mixing (ΔG_{mix}), enhancing system stability, as long as the enthalpy of mixing (ΔH_{mix}) remains constant. Essentially, raising the configurational entropy (ΔS_{config}) in High Entropy Alloys (HEAs) allows for the stabilization of a single-phase crystal structure by increasing the number of elements randomly distributed on the lattice sites.[19]

Murty *et al.* categorized the materials with $\Delta S_{\text{config}} > 1.5R$ as "high entropy," $1.5R > \Delta S_{\text{config}} \sim 1R$ as "medium entropy," and $\Delta S_{\text{config}} < 1\ R$ as "low entropy" systems based on the entropy of the materials configuration.[31,32] HEAs have four main effects due to the existence of several components with distinct characteristics: (1) high entropy, (2) lattice distortion, (3) slow diffusion, and (4) cocktail effects.

The high entropy effect typically involves a qualitative assessment, highlighting the increased mixed configuration entropy in HEAs compared to that found in pure metals and intermetallic compounds.[19] Fig. 1 shows the properties and characteristics of HEA's.[33] The fundamental idea behind HEMs is the high-entropy effect. Yeh was the first to suggest this effect, which tends to reduce the number of phases and stabilize the high-entropy phases.[19,34] At higher temperatures, the configurational entropy (Eq. (1)) and stability of the single-phase structure are increased by many near-equimolar components. N-element alloy systems can generate up to N+1 phases in the thermodynamic high-entropy effect. HEAs, however, typically result in single-phase solid solutions as opposed to split phases or different intermetallic compounds. Lattice distortion is inevitably caused by variations in atom sizes, stemming from their inherent differences. Apart from these size variations, asymmetrical binding and electronic structures within the atoms and their immediate surroundings further contribute to this effect. This influence varies across the lattice, impacting bond energy and crystal structure, ultimately contributing to significant lattice distortion.[19]

These materials have greater hardness, reduced thermal and electrical conductivity, and reduced X-ray diffraction peak intensity and are all caused by lattice distortion.[35] The lattice distortion in high entropy alloys (HEA) is attributed to the random distribution of atoms in the crystal matrix.[36] Variances in atomic radii, chemical bonds, and local environments contribute to increased lattice distortion and defects within the structure. HEAs' microstructure and characteristics are mostly determined by a unique characteristic that significantly influences the lattice distortion effect.[37] This lattice distortion effect essentially gives HEAs their exceptional mechanical properties, including greater strength, ductility, and toughness.[38] In the hysteresis diffusion effect of kinetics, precipitates below tens of nanometers are commonly found in the interior of HEA, signifying sluggish diffusion and phase transformation rates.[39] Sluggish diffusion might be due to two factors. The first is that each element in the HEA diffuses differently, and the second is because the adjacent atoms at each lattice location are distinct. Moreover, HEA resists structural changes, such as grain coarsening or recrystallization, at elevated temperatures.

The sluggish diffusion observed in HEAs refers to the sluggish movement of atoms within the crystal lattice, resulting in slower diffusion rates compared to traditional alloys.[40] This reduced diffusion is caused by the high level of atomic disorder and the presence of multiple elements with different atomic sizes.[41] The slow diffusion phenomena in HEAs has several consequences. It first improves the mechanical and physical properties of the alloy by forming a stable solid solution with an equitable distribution of components. Second, it stops phase separation and dangerous intermetallic compound formation, both of which can reduce the alloy's performance.[42] Lastly, the slow diffusion effect helps HEAs become more stable and long-lasting by strengthening their resistance to grain formation. Ranganathan introduced the phrase "multimetallic cocktails" for the first time and emphasized the significance of alloys in the creation of alloys.[43] Phase distribution, form, boundary, and overall contribution of each phase's qualities to the component phases define the overall properties.[36] The cocktail effect of HEMs happens when the mixture contains many components with different characteristics. Such an alloy's eventual property is uncertain and does not equal the sum of its component attributes.[9]

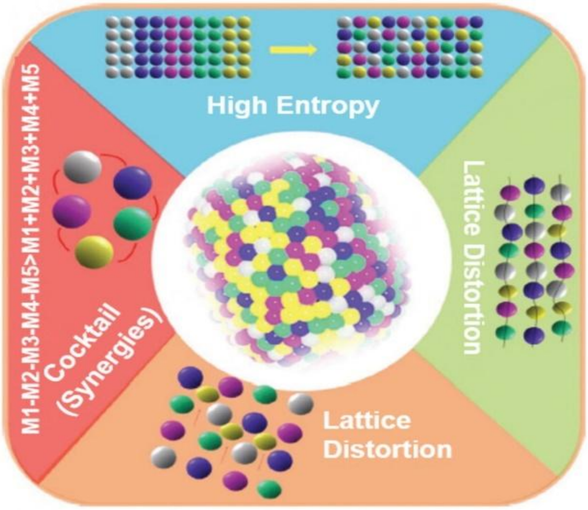

Fig. 1: Schematic illustration of the four effects of HEMs.[33]

3. Synthesis and structures of high entropy materials

A wide range of synthesis methods has been developed to fabricate high-entropy materials in different forms, each with distinct processing conditions, advantages, and limitations. Table 1 summarizes the key synthesis techniques commonly employed in the preparation of HEMs, including their temperature ranges, scalability, typical product forms, and representative references. A variety of structural forms, including hexagonal and cubic crystals with FCC and BCC lattices, may be found in HEAs. The FCC lattice is by far the most extensively explored HEM structure. Fig. 2 shows the different crystal structures of HEM's. The table provides an overview of reports on the synthesis and structures of HEA NPs with various crystal structures.

3.1 Alloys

Material scientists have employed a variety of methods, including additive manufacturing (AM) processing techniques, powder metallurgy (PM) methods, mechanical alloying (Solid state reaction), and melting and casting methods, to synthesize hybrid electrode assemblies (HEAs) within the last decades.[44] The simplest and most widely used technique for preparing the new high entropy alloy material is vacuum arc furnace casting. Then, using magnetron sputtering, the multi-element high-function alloy coating was effectively created. By using melted casting, forging, powder metallurgy, spraying, and coating techniques, block, coating, and film materials may be combined to create the new high-speed alloy material. However, achieving uniform atomic-scale mixing in alloy-based HEMs remains challenging due to phase segregation and melting point disparities among constituent elements. As

Table 1: Summary of typical synthesis methods for high-entropy materials and their characteristics.

Synthesis method	Temperature range	Scalability	Advantages	Limitations	Morphologies	Ref.
Ball milling	RT – 600 °C	High	Low cost; large-scale powder processing	Long processing time; potential contamination	Nanoparticles, precursors	[44-46]
Vacuum arc melting / casting	>1000 °C	Medium	Homogeneous melting; suitable for alloys	Limited composition control; high energy consumption	Bulk ingots, alloys	[44]
Spark plasma sintering (SPS)	800–1200 °C	Medium	Fast densification; phase control	Expensive; limited throughput	Dense bulk ceramics	[45]
Sol-gel	<400 °C	Medium–High	Excellent compositional homogeneity	Drying and calcination steps time-consuming	Nanoparticles, thin films	[45]
Wet chemistry (*e.g.*, co-precipitation, spray pyrolysis)	100–600 °C	Medium–High	Versatile; solution-based; scalable	May require post-sintering; precursor sensitivity	Powders, coatings	[53]
Pulsed laser deposition (PLD)	~500–800 °C	Low	Enables single-crystal thin films	High cost; limited substrate compatibility	Thin films	[45]
Microwave-assisted synthesis	100–300 °C	Medium	Energy-efficient; rapid synthesis	Not suitable for all compositions	Nanoparticles	[46,47]
Solvothermal/ hydrothermal	100–250 °C	Medium	Mild conditions; high crystallinity	Long reaction times; pressure vessel required	Nanostructures, MOFs	[53,54]
Electrochemical deposition	RT – 100 °C	High	Precise control over composition/thickness	Limited to conductive substrates	Coatings, films	[48]
Pulsed thermal decomposition	>1400 °C	Low	Rapid synthesis; metastable phase formation	Requires high-temp pulses; not yet scalable	Powders, nanoalloys	[48]

a result, ceramic-based high-entropy materials have gained attention as promising alternatives, offering improved compositional control and superior thermal stability.

3.2 Ceramics

HECs have been synthesized using different techniques: solid-state reaction, wet chemistry, and epitaxial growth. The most common synthesis method for creating HECs is a solid-state reaction, in which the precursor powders are mixed during ball milling or created using mechanochemistry, and the resulting powders are then sintered.[45] Ball milling is only utilized to mix the powder for HECs made from high melting point precursors, such as oxides and carbides; the production of HECs happens during sintering, which is essentially when the component inter-diffusion processes take place. The synthesis of HECs is accomplished by the use of wet chemical techniques such as reverse co-precipitation, nebulized spray pyrolysis, and phenol spray pyrolysis. HEC synthesis is typically carried out via epitaxial film growth. HEC oxides have only been prepared by pulsed laser deposition (PLD) thus far in the literature. This technique has the benefit of allowing for the growth of single crystals, which prevents grain boundary segregation and allows for the achievement of accurate elemental ratios.

3.3 Sulfides

There have only been a limited number of publications regarding the effective synthesis of high entropy/entropy-

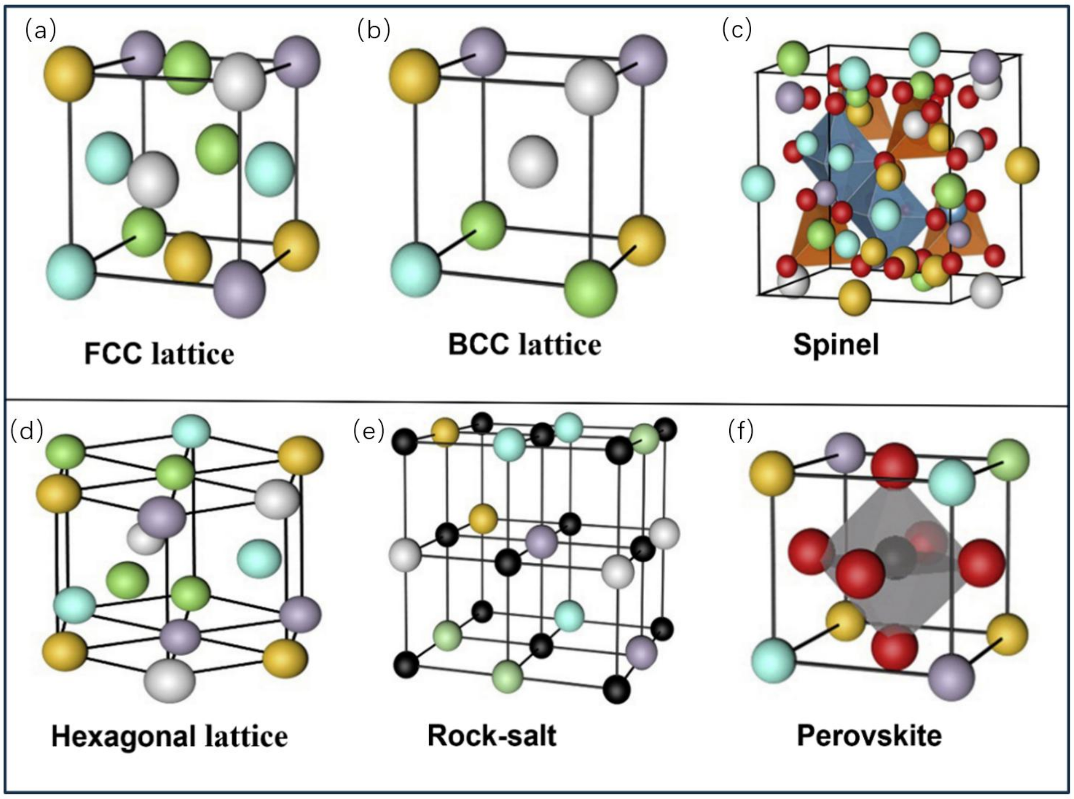

Fig. 2: Typical structures of HEMs: (a) FCC lattice structure, (b) BCC lattice structure, (c) Spinel structure, (d) Hexagonal lattice structure, (e) Rock-salt structure, and (f) perovskite structure.

stabilized metal sulfides. These synthetic approaches needed complex, dangerous, time-consuming, and high-temperature processes.[46] According to one report, almost 60 hours of ball milling are required to produce a homogenous distribution of the necessary components within the (CuSnMgGeZn)S and (CuSnM-gInZn) system. Under a different approach, the reaction vessel must be prepared by annealing quartz with extremely dangerous HF, and then the elemental powder must be heated to a high temperature for 120 hours.[47] It has also been reported that (CrMnFeCoNi)Sx is synthesized by pulsed thermal decomposition of metal salts and thiourea, with a first 6-hour drying process, extremely high annealing temperatures (~1650 K) over brief time intervals (~55 ms), and a quick quenching mechanism.[48] Only Schaak *et al.*[48] have, to the best of our knowledge, documented a low-temperature synthetic approach for high entropy. Cu-Zn-Co-In-Ga-S.

3.4 Dichalcogenides

More recently developed HE metal chalcogenides have an anionic sub-lattice consisting of sulfide, selenide, and telluride ions (S, Se, Te, or a mixture of the three). Among the more recently synthesized HE metal chalcogenides, HE metal telluride and mixed chalcogenide materials have also been reported, although HE metal sulphide or disulphide compounds are the most abundant. Numerous methods have been employed to synthesize HE metal chalcogenides, including ball-milling, elemental annealing, solvothermal annealing, and pulsed thermal decomposition.

3.5 Metal-organic framework

Metal-organic frameworks have shown wide ranges of applications.[1,49-52] A simple ambient temperature solution phase approach was used by Zhao *et al.* to create a high entropy MOF. The produced HE-MOF-RT exhibits a large BET surface area, a sheet-like nanostructure, and a near-equimolar element ratio. With an overpotential of 245 mV at 10 mA cm^2, HE-MOF-RT demonstrates exceptional electrocatalytic activity for OER.[24] (HE-MOF) has been logically synthesized using a moderate solvothermal approach by Xu *et al.* with five metals (Ni, Co, Fe, Zn, and Mo). The quintuple metal in the HE-MOF could have an additive impact, and its two-dimensional array structure could improve electrical conductivity and encourage reactant, intermediate, and product transfer, leading to good electrocatalytic OER activity.[53] Hu *et al.* studied the production mechanism of a porous hollow high-entropy metal-organic framework (MOF-74) made of Mn, Fe, Co, Ni, Cu, and Zn using a one-pot hydrothermal technique.[54] At room temperature, Ling-Guang Qiu *et al.* developed the synthesis of four well-known MOFs.[55,56]

In recent years, low-temperature synthesis strategies have garnered increasing attention for fabricating high-entropy materials due to their energy efficiency, environmental friendliness, and compatibility with metastable phase formation. Microwave-assisted methods, for example, have been applied to synthesize sulfide-based HEMs with shorter reaction times and better particle size control, avoiding prolonged high-temperature processing. Electrochemical deposition has emerged as a promising route for preparing high-entropy MOFs and thin films, offering precise control over stoichiometry and morphology at ambient conditions. These emerging techniques present clear advantages over traditional solid-state or melt-based routes, especially in terms of reducing thermal budget and enabling direct integration onto substrates. Notably, McCormick and Schaak demonstrated a low-temperature molten salt synthesis pathway that stabilizes high-entropy chalcogenides with controlled elemental ratios and tunable electronic properties,[56] showcasing the transformative potential of non-conventional methods.

4. Applications of high entropy materials

HEMs have many desirable characteristics that make them highly promising for use in energy conversion, storage, and catalysis, *etc*, as shown in Fig. 3.[57-59]

4.1 Catalysis

Although research on the catalytic performance of HEMs is still in its earliest stages, their exceptional activities and promising results are sparking interest for further investigation across a range of catalytic applications. Studies reveal that HEAs not only outperform single-atom catalysts but also exhibit superior activity and stability compared to binary and ternary alloys. A particularly attractive strategy is to harness HEAs made from non-noble metals. These materials are not only more cost-effective and plentiful, but they also deliver catalytic activities and stabilities that match or even surpass those of their noble metal counterparts.

HEMs, particularly HEOs, are ideal for applications needing high-temperature stable catalysts, as their entropic contribution at high temperatures helps minimize Gibbs free energy. Additionally, HEOs serve as excellent supports for dispersing single noble metal atoms on their highly active sites. This dual functionality enhances catalyst activity while reducing material costs by minimizing the use of precious metals.[60] Binary and ternary transition metal alloys are efficient catalysts but often struggle with miscibility gaps and limited compositional flexibility. Adding two or three suitable elements to create HEA catalysts resolves these issues, enhancing activity and stability. HEAs show great potential for both OER and ORR. It is possible to improve Pt-based noble-metal catalysts by adding more components by using a high-entropy strategy.[61] This not only boosts their catalytic capabilities through synergistic effects but also reduces the noble metal content, thereby lowering overall costs. Four proton-coupled electron transfers with sluggish kinetics are involved in the OER. In both acidic and alkaline conditions, this half-reaction-driven reaction yields molecular oxygen.

Despite these advances, the long-term durability of HEA

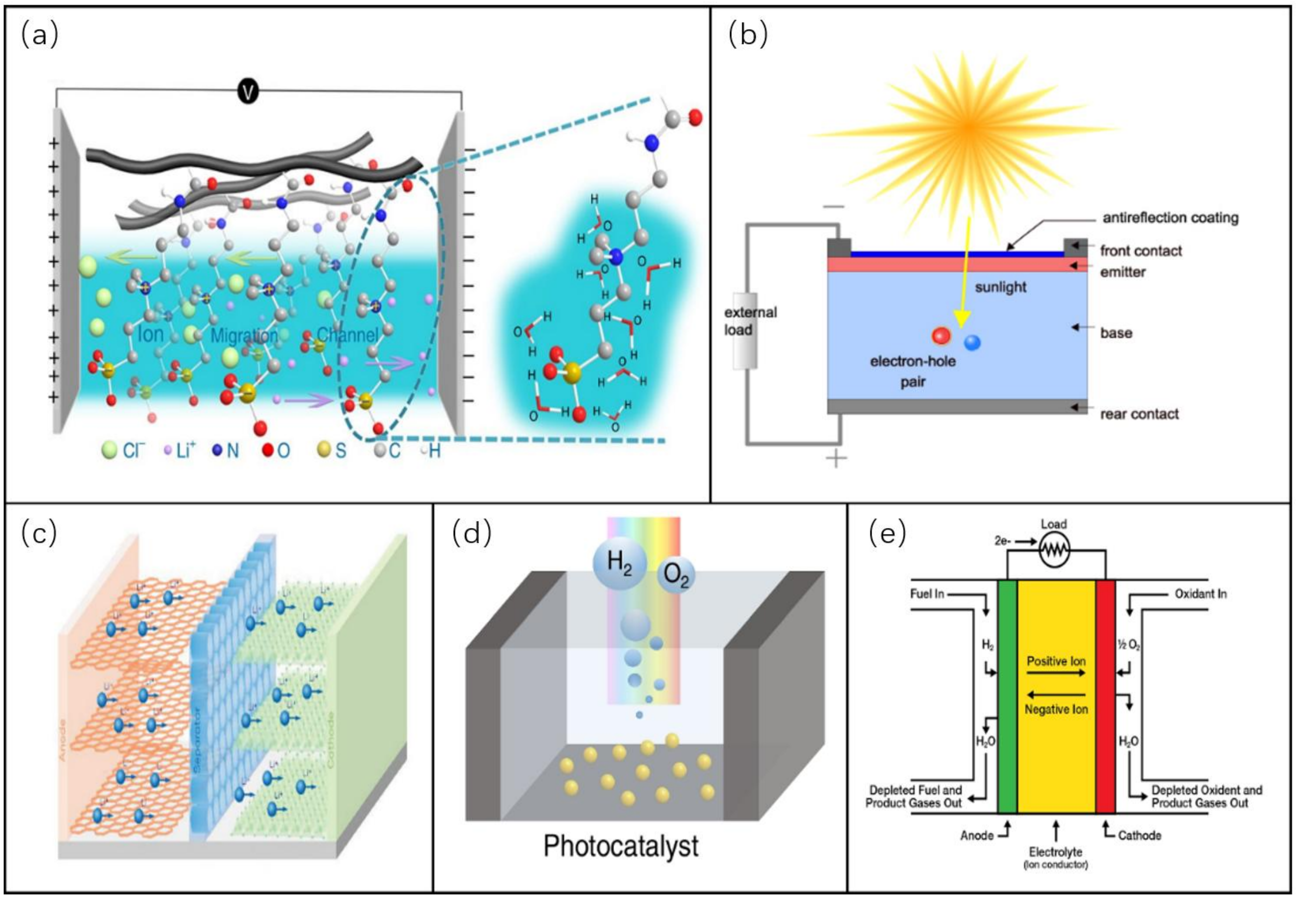

Fig. 3: Various applications of HEMs: (a) supercapacitor,[57] (b) lithium ion battery,[58] (c) catalysis,[59] (d) fuel cells, (e) solar cell.

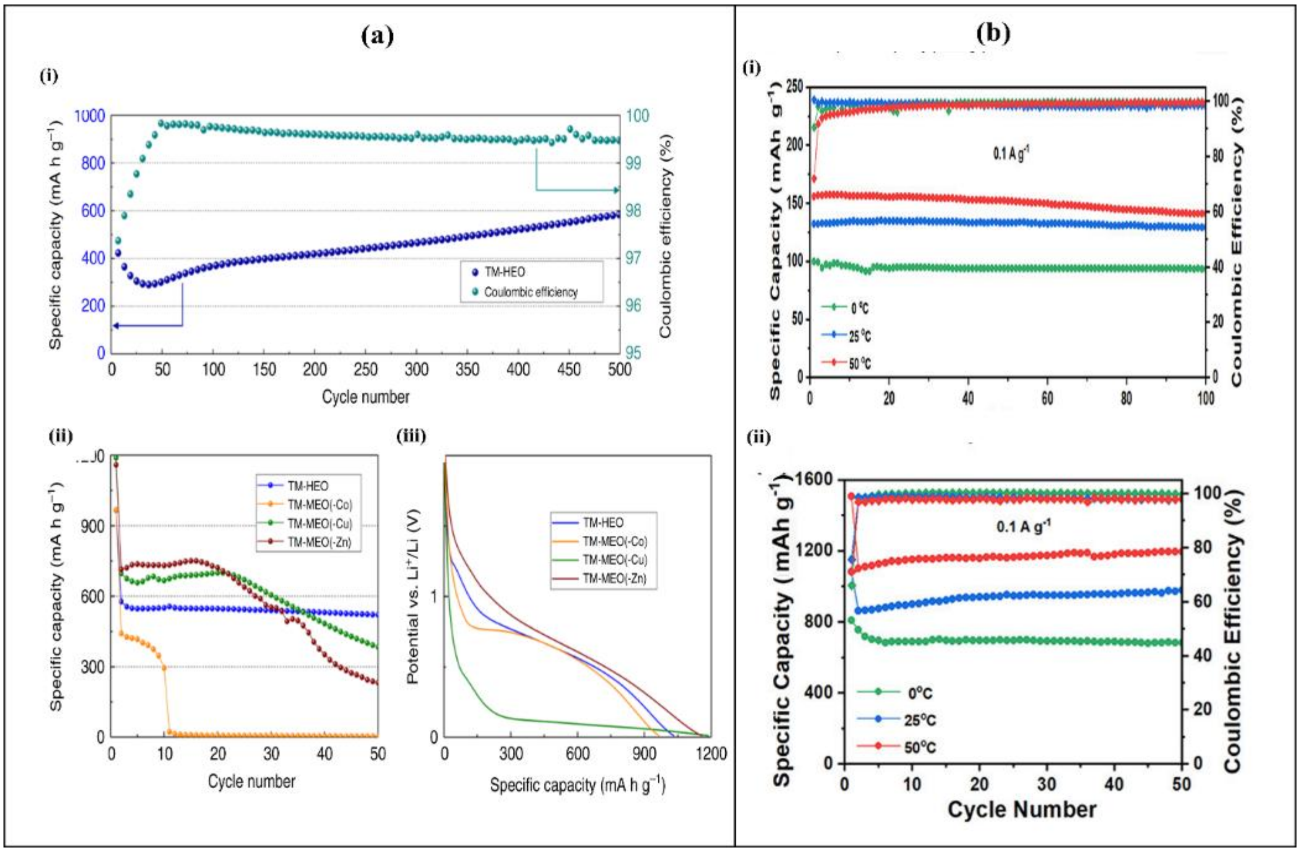

Fig. 4: (a) An analysis of the several medium and high entropy oxides under investigation. (i) long-term cycling stability at 200 mA g^{-1} with consistent coulombic efficiency. (ii) stable capacity at 50 mA g^{-1}, whereas materials lacking Zn and Cu suffer severe degradation, and the material without Co fails after 10 cycles. (iii) Discharge (lithiation) profiles of the first cycle for the different compounds.[71] (b) cycling performance of CCFMNO electrode. (i) long-term cycling stability at various temperatures and current densities of 0.1 A g^{-1}, (ii) long-term cycling performance at different temperatures.

catalysts in corrosive environments—particularly acidic media—remains a critical challenge. Surface passivation, caused by oxide layer formation, elemental leaching, or structural reconstruction, can lead to a gradual decline in catalytic performance. Addressing this issue requires careful surface and compositional engineering. Incorporation of corrosion-resistant elements such as Pt or Ir has been shown to enhance stability, while protective coatings like conductive polymers or graphene layers can serve as diffusion barriers against aggressive species. Tailoring the electronic structure through entropy-tuned composition can also suppress undesirable surface reactions. Recent studies suggest that dynamic surface restructuring or in-situ regeneration may further prolong catalyst lifetime, making HEAs more viable for applications such as acidic water splitting and PEM fuel cells.

4.2 Batteries

Batteries have revolutionized our society and widen different applications. HEMs are a new class of materials science that is gaining popularity due to their remarkable energy storage capabilities. Because of their structural stability, these equimolar multielement compounds have larger charge capacities, higher ionic conductivities, and a longer cycle life.[62,63] The preferred power source for a variety of uses, including electric cars and portable devices, is rechargeable batteries. Because of their superionic conductivity, HEMs, especially HEOs, were first taken into consideration for batteries. Promising solid electrolytes, Bérardan *et al*. created single-phase HEOs with a rocksalt structure that demonstrated quick Na mobility and strong Li-ion conductivity at ambient temperature.[64] High-capacity electrodes in Li/Na-ion batteries can be constructed with HEAs, and their performance can be optimized through compositional tuning. They are also perfect for the air electrodes in Li-air and Na-air batteries due to their excellent electrocatalytic activity for OER and ORR.[65] Even while research on high-entropy polymers is still in its early stages, it may lead to the creation of innovative solid polymer electrolytes for Li/Na-ion batteries that have better cycling stability and ionic conductivity.[66] Polymer composite electrolytes have been seen to exhibit potential as solid electrolytes by the incorporation of various metal oxides, ceramic nanoparticles, nano-flakes, and MOFs. In the future, the design and research of novel high-entropy polymer-composite materials will be made easier by the benefits of these materials high entropy form.[67-69] An entropy-stabilized spinel oxide, $(Co_{0.2}Mn_{0.2}V_{0.2}Fe_{0.2}Zn_{0.2})_3O_4$, has been developed by Shisheng Hou *et al*. as a potential anode for LIBs. Moderate volume change and highly reversible capacities of approximately 900 mAh g^{-1} at 0.2 A g^{-1} for 500 cycles and approximately 500 mAh g^{-1} for 2000 cycles at 3 A g^{-1} are presented by this material. This study highlights the potential of HE-TMO anodes for advanced LIBs with high capacity and long cycle life.[70]

In 2018, Sarkar *et al*. synthesized the $(Co_{0.2}Mn_{0.2}V_{0.2}Fe_{0.2}Zn_{0.2})_3O_4$ rock-salt compound using the NSP method and demonstrated its effectiveness as an anode, achieving a capacity of approximately 500 mAh/g at 0.1 A/g, as shown in Fig. 4a(i). They also tested medium-entropy derivatives of the high-entropy parent structure, but these exhibited a faster capacity decay compared to the HEO (Fig. 4a(ii)). Additionally, they performed medium-entropy derivatives of the parent structure with a higher entropy, but all of them showed a faster rate of capacity erosion than the HEO. The electrochemical performance of three of the 4-cation systems without Zn, Cu, or Co, and the 5-cation TM-HEO are compared in Fig. 4a(iii).[71,72]

Fig. 4b(i) shows cycling performance of CCFMNO electrode, which indicates that at various temperatures and current densities of 0.1 A g^{-1}, capacity degeneration does not occur during 50 cycles. This observation may be explained by the strong structural stability of HEMs at very high temperatures. Fig. 4b(ii) shows long-term cyclingperformance at different temperatures, After 100 cycles at 50, 25, and 0 °C, respectively, the high reversible capacities of 141 mAh g^{-1} (capacity retention 91%), 132 mAh g^{-1} (capacity retention 98%), and 100 mAh g^{-1} (capacity retention 94%) were found.[71] Overall, the design of high-entropy materials for battery electrodes and electrolytes leverages features such as compositional flexibility, lattice distortion, and ionic conductivity. These same principles also apply to HEMs in supercapacitors, where electrochemical performance is similarly governed by surface redox activity and structural stability. A comparative summary of key performance metrics is provided in Table 2.

While HEOs exhibit excellent capacity and cycling stability as battery anodes, volume expansion during charge-discharge cycles remains a major issue. Lattice distortion inherent to multi-element compositions helps alleviate this challenge by distributing internal strain and buffering structural changes, thus suppressing particle pulverization and maintaining integrity. In solid-state batteries, poor interfacial contact between HEM electrodes and solid electrolytes often limits ion transport. Interface engineering approaches such as introducing buffer layers, surface coatings, or compositionally graded interfaces have been shown to enhance both mechanical and electrochemical stability. For sulfide-based high-entropy materials, low intrinsic electronic conductivity is another barrier to practical use. Strategies including conductive carbon coating, heterostructure formation, and aliovalent ion doping have demonstrated effectiveness in improving conductivity while preserving the material's multicomponent characteristics. These mechanisms and design principles are essential for improving the electrochemical performance of HEMs in next-generation energy storage systems.

4.3 Supercapacitor

While the operating mechanisms of supercapacitors differ from batteries, many HEMs demonstrate excellent charge storage performance in both applications due to their shared structural advantages. Particularly, high-entropy oxides and alloys have shown promising electrochemical behaviors across these platforms. Among high-power energy storage technologies, supercapacitors are highly promising due to their nearly infinite cycle life, higher power and energy density than batteries and dielectric capacitors.[73-76] ECs come in two types: electrical double-layer capacitors (EDLCs) and pseudo-capacitors, each operating on distinct mechanisms. In EDLCs, energy storage occurs via electrostatic interactions in the double layer between the electrode surface and electrolyte. Carbon nanomaterials are used as electrodes, offering high power density and long cycling life but limited energy density.[77] Conversely, pseudo-capacitors operate through faradaic charge transfer at the electrode-electrolyte interface, providing higher energy density but with reduced cycling life.[78] Capacitor electrodes can be produced by modifying HEOs derived from TM-metal oxides for pseudo-capacitance. Additionally, carbide and sulfide HEMs are promising electrode materials for EC devices, having proven effective in their binary and ternary forms.

Positive electrodes made of Fe-Co-Ni-Cu-Zn HEA and negative electrodes made of AC (activated carbon) were created by Gobinda Chandra Mohanty *et al*. After an additional analysis of the mass loading, the corresponding CV (three electrode systems) of AC and HEA with a potential window of -1.0 to 0.0 V and 0.0 to 0.5 V at 50 mV s^{-1} are recorded, as shown in Fig. 5a.

GCD measurements range from 0.8 to 1.5 V at 2 A g^{-1} (Fig. 5b); no discontinuity is seen. GCD plots were collected at different current densities ranging from 0.5 to 5 A g^{-1}, as the charge-discharge plots remain stable up to 1.5 V.[79] To create HEOs $(FeCo\text{-}NiCrMn)_3O_4$ for use as supercapacitor cathode materials, Zhang *et al.* used the dealloying process. Cyclic voltammetry (CV) measurements were employed to assess the electrodes' electrochemical properties. Fig. 5c displays the $(FeCoNiCrMn)_3O_4$ curves at a scan rate of 10-100 mV/s and a voltage window of 0-0.6 V. Fig. 5d indicates how the GCD curve explains the electrochemical properties of HEOs. These are reported for various current densities ranging from 1 to 10 A/g for a voltage window of 0-0.46 V. Specific capacitances of 639, 595, 578, 578, 552,523 and 508 F/g have been obtained for current densities of 1, 2, 3, 5, 8, and 10 A/g, in that order. The material possesses a high specific capacitance, as demonstrated by electrochemical experiments (639 F/g at 1A/g).[80]

Table 2: Summary of performance metrics of high-entropy materials in batteries and supercapacitors.

Application	Material	Specific Capacity/Capacitance	Cycle Life	Current Density	Key Features	Ref.
LIB anode	$(Co_{0.2}Mn_{0.2}V_{0.2}Fe_{0.2}Zn_{0.2})_3O_4$	~900 mAh/g @ 0.2 A/g	2000 cycles	0.2–3A/g	Lattice distortion, stable structure	[70]
LIB electrolyte	Rocksalt HEOs (Na/Li)	-	Stable cycling	-	High Li/Na-ion conductivity	[64]
Supercapacitor	$(FeCoNiCrMn)_3O_4$	639 F/g @ 1 A/g	>10,000 cycles	1–10 A/g	Pseudocapacitance, redox-active sites	[80]
Supercapacitor	FeCoNiCuZn HEA	-	Stable Galvanostatic charge-discharge (GCD) cycling	2 A/g	High voltage window, excellent retention	[81,82]

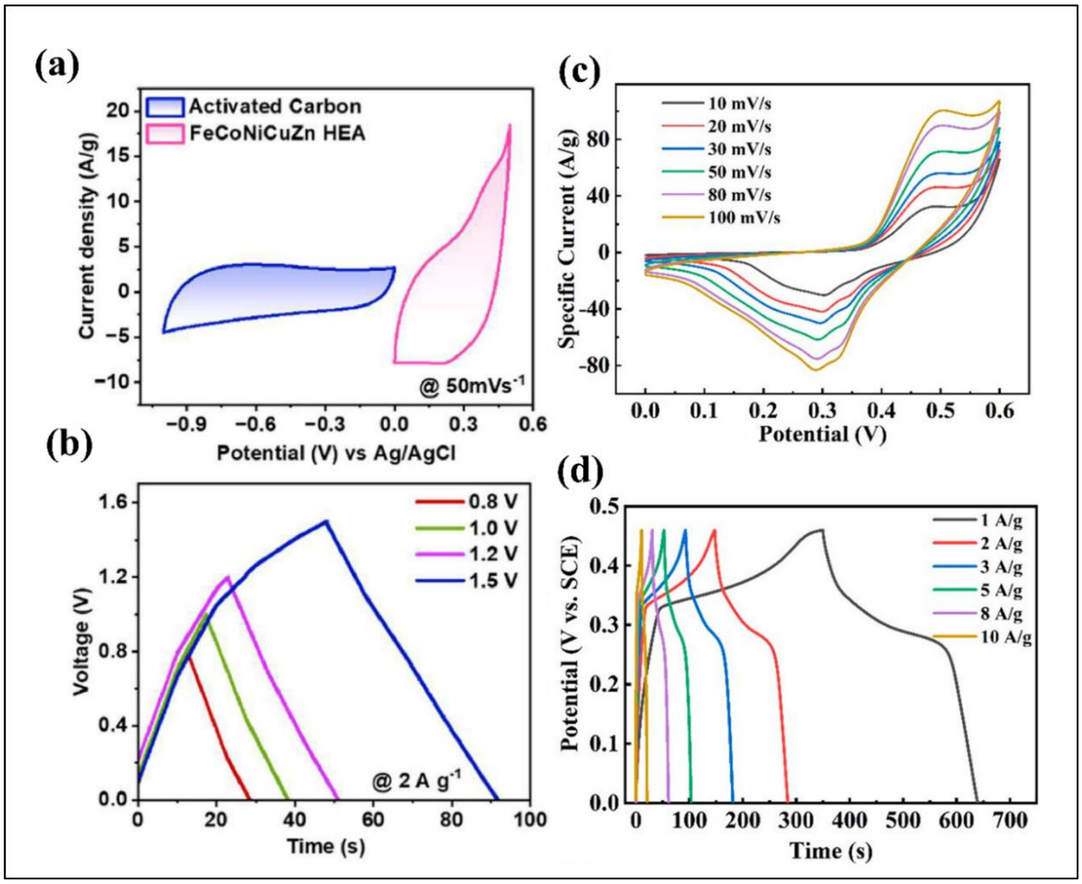

Fig. 5: (a) Fe-Co-Ni-Cu-Zn HEA and AC CV at 50 mV s^{-1}, (b) GCD curves between 0.8 and 1.5 V, (c) CV curves of $(FeCoNiCrMn)_3O_4$, (d) GCD curves of $(FeCoNiCrMn)_3O_4$.

4.4 Solar cells

Solar energy is a clean, unlimited renewable energy source, but due to material costs and efficiency problems, solar cells are still expensive.[81,82] Finding low-cost, abundant, and effective materials for various solar cell types is an ongoing research challenge. HEMs can enhance solar cells by boosting efficiency and lowering costs. Tailoring HEAs allows precise tuning of optical properties and bandgap reduction in semiconductor materials for thin-film solar cells. The high-entropy concept extends to various materials, including halides, offering superior properties over binary and ternary forms. Developing single-phase crystalline high-entropy halide perovskites (ABX_3) as quantum dots for light-sensitized perovskite solar cells can greatly enhance performance.

Metal oxides with large bandgaps, such as TiO_2 and ZnO, are critical in dye-sensitized solar cells (DSSCs) and PSCs.[83] Cell performance can be improved by increasing their bandgap and optical characteristics via partial doping. However, single-phase structures and high synthesis temperatures pose challenges for ternary oxides. These concerns are resolved by entropy-stabilized single-phase HEOs, which have a wide compositional range.

4.5 Fuel cells

Fuel cells have attracted increasing attentions due to their wide applications.[84,85] With simultaneous ORR and HOR processes, high-temperature solid oxide fuel cells (SOFCs) can directly convert chemical energy from a variety of flexible fuels,[86] such as hydrogen, hydrocarbons, and ammonia, into electrical energy with high efficiency and low emissions (up to 85%).[87] HEOs are increasingly frequently selected for SOFC cathodes because of their superior qualities of high activity and stability. Because the complicated compositions of SOFCs using perovskite HEOs never approach homogeneous mixing using the solid-state method, the sol-gel method has emerged as the favoured method.[88]

Though significant compositional space needs to be explored for optimal performance, HEMs have shown remarkable potential as anode and cathode electrocatalysts for fuel cells. Compared to binary oxides and doped cerium oxides with less additional elements, fluorite HEOs, which contain equimolar cerium and other rare earth elements, show more oxygen vacancies.[89] They serve as interesting solid electrolytes for solid oxide fuel cells because of their strong oxygen conductivity and stability at high temperatures. They are also suitable for dual-phase ceramic-molten carbonate membranes and porous ceramic supports in molten carbonate fuel cells due to their comparable chemical and mechanical stability to YSZ.[90]

5. Conclusions and perspectives

Nowadays, a large variety of organic and inorganic materials are covered by the idea of high-entropy, which may be efficiently applied to the design and development of numerous novel materials with desired features. By adjusting entropy, which is the degree of unpredictability, one can investigate the mechanical and functional characteristics of materials for a range of purposes by varying both compositional complexity and structural disorder. HEMs, which are characterized by their entropy-driven stabilization, include metallic alloys as well as oxides, diborides, carbides, and sulfides. These materials are emerging novel materials with intriguing characteristics and outstanding durability. A single-phase solid solution forms thermodynamically more readily when there is a high ΔS_{mix} along with the mixing of five or more near-equimolar metal elements or ions.

The development of a single-phase crystalline solid solution is influenced by a number of other thermodynamic, structural, and kinetic properties, according to further research. Entropic phase stability, lattice distortion, slow diffusion, and cocktail effects are some of the remarkable functional aspects of HEMs that have drawn attention in addition to their well-known mechanical properties.

In this review, high-scalability synthesis approaches were the primary concern, we also examine a number of scalable synthesis techniques. Given their low cost, straightforward precursors, quick processing periods, and capacity to synthesize vast quantities of materials, ball milling, aerosol, microwave, and electrochemical synthesis are the synthesis technologies that hold the most promise for future industrial scale. This overview attempts to provide an overview of the fundamental ideas of HEM and investigates the methods for producing HEM as well as the economical industrial production of these materials for potential uses in the future.

HEMs exhibit superior characteristics as catalyst materials or catalyst supports. HEMs have been studied as cathode/anode electrocatalysts for a variety of reactions, including the HER, OER, FAOR, EOR, ORR, and MOR. They have also been studied as catalysts for the burning of methane, ammonia breakdown, CO oxidation, CO_2 reduction, and azo dye degradation. HEMs show significance in the energy storage industry as materials for storing hydrogen, electrodes for batteries, and capacitors.

Despite these advancements, several critical challenges remain unresolved. Large-scale and cost-effective synthesis of HEMs continues to be hindered by high-temperature requirements, complex procedures, and batch-to-batch variability. Additionally, identifying optimal compositions in multi-element systems is difficult due to the vast compositional space and nonlinear interactions among elements. Progress in this field increasingly relies on interdisciplinary approaches. Machine learning and data-driven methods are being explored to predict promising element combinations, optimize synthesis parameters, and uncover hidden structure–property relationships. Integration of high-throughput experimentation and AI-guided modeling can significantly accelerate materials discovery and development. Furthermore, developing energy-efficient and environmentally friendly synthesis methods is crucial for promoting sustainable and industrially viable applications of HEMs.

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Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

CRediT Statement

Zhiwei Li: Conceptualization, Writing – original draft, Writing – review & editing, Supervision, Project administration, Resources. **Kejia Zhang**: Writing – review & editing. **Wenkai Zhang**: Writing – review & editing, Funding acquisition. **Tao Ding**: Supervision, Resources, Project administration, Funding acquisition.

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