

A perspective on high-entropy two-dimensional materials

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Abstract

In past decades, high-entropy (HE) materials, containing five or more elements with approximately equal atomic ratio, are extensively investigated due to their desirable properties in a series of applications. Recently, HE two-dimensional (2D) materials have become promising materials, which not only endow the advantages from their bulk form but also exhibit unusual properties due to their 2D features. So far, the HE 2D transition metal carbides (MXenes), dichalcogenides (TMDs), hydroxalicates (LDHs), and oxides have been successfully synthesized and performed well in different electrochemical reactions, which is originated from the synergistic effect of multicomponents and atomic thin characteristics. Here, the challenges on processing, characterization, and property predictions of HE 2D materials are emphasized. Finally, viable strategies, advanced processing, fundamental understanding, in-depth characterization of new HE 2D materials are proposed.

KEYWORDS

2D materials, high-entropy, lattice distortions, MXenes, strong stress

1 | INTRODUCTION

Since the concept of “high-entropy (HE) alloy (HEA)” was proposed in 2004, it has been rapidly extended to other compounds, covering oxides,¹ diborides,² nitrides,³ sulfide,⁴ and carbides.⁵ Based on the equation of $S = R \ln N$, in which R represents the gas constant and N is the quantity of metal components, HE material (HEM) is defined as the material with $S > 1.5R$, indicating that at least five kinds of metal need to be contained.^{6,7} Compared to the HEA, HE compounds exhibit structural diversity, showing more possibilities by achieving desirable properties for various applications such as supercapacitor, electromagnetic

interference shielding, rechargeable batteries, and electrochemical catalysis. The HE phenomenon makes it possible to accommodate several metal atoms into one solid solution of oxide, sulfide, nitride, phosphide, carbide, and so on. The same composition of metals in various compounds also brings different intrinsic properties.

With advanced nanotechnology, a variety of HE nanostructures have been constructed and widely studied due to their favorable catalytic behaviors for many key electro- and thermocatalytic reactions, involving hydrogen evolution reaction (HER),⁸ oxygen reduction reaction,⁹ oxygen evolution reaction (OER),¹⁰ CO₂ reduction reaction (CRR), CO oxidation,¹¹ and NH₃ decomposition.¹² Among them,

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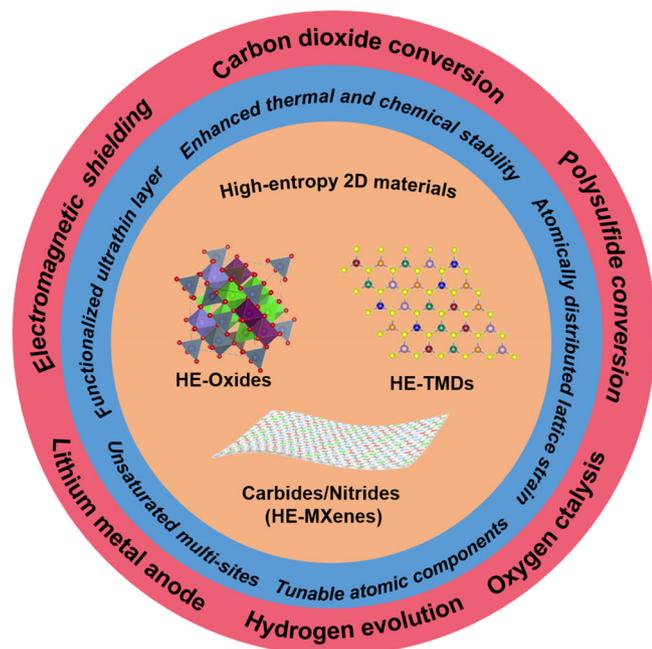


FIGURE 1 Schematic illustration of the unique properties and potential applications of HE 2D layers

two-dimensional (2D) materials have attracted extensive investigation due to their unique properties compared to their bulk forms. Also, the 2D features make them easy to be assembled with other materials to prepare other functional composites. Stabilizing five or more elements into one or few atomic layers with single crystal structures opens a new direction for 2D materials design and finalization, in which the concepts of “2D” and “high-entropy” are properly combined. Inspired by the exfoliation of layered structures, it is reasonable to obtain the HE 2D materials via top-down synthesis strategies, as illustrated in Figure 1.

Very recently, the top-down strategies were used to exfoliate the layered materials to synthesize the HE 2D materials, which have been used for rechargeable batteries and catalysis.^{13,14} The random distribution and the infinite combination of metallic elements at atomic-scale thickness endow HE 2D materials with attractive features, such as good thermal and chemical stability, ultrathin layer with high mechanical strain, high flexibility with tunable functional groups, as well as abundant unsaturated multisites, unfolding the great potential in various research fields. Although there are only a few reports reported so far, these reports significantly expand the family of 2D materials. In this perspective, the challenges on synthesizing, characterization, and property anticipation are highlighted. Finally, viable strategies, novel synthesizing, in-depth understanding, and advanced characterization of new HE 2D materials are proposed.

2 | SYNTHETIC METHODS FOR 2D HIGH ENTROPY MATERIALS

Currently, three main strategies are widely used for the preparation of 2D materials: the top-down method, bottom-up method, and the topological conversion method.^{15–17} These synthetic methods are also applicable to the preparation of 2D HE materials. As a large amount of research work on 2D HE materials continues to be studied and reported, many new preparation methods are being proposed, enriching the methods for the preparation of 2D HE materials.

2.1 | Top-down method

The top-down method is widely used to delaminate the van der Waals layered structures.¹⁸ Among which, mechanical exfoliation and liquid-phase exfoliation are two common ways for synthesizing 2D HE materials.

2.1.1 | Mechanical exfoliation

In 2004, Geim et al. used a simple mechanical exfoliation method to successfully exfoliate and observe monolayers of graphene from highly oriented pyrolytic graphite for the first time, making mechanical exfoliation an important method for the preparation of 2D materials.¹⁹ Typically, the lamellar 2D material is thinned by repeatedly peeling off via a transparent adhesive tape, which is transferred to a specific substrate such as SiO₂/Si. Mechanical exfoliation is also an effective method for preparing 2D HE materials with high cleanliness, high crystallinity, and thick atomic layers. For example, Qi et al. used the chemical vapor transport method to obtain a series of single-crystal, HE van der Waals materials, which were then exfoliated by transparent adhesive tape to obtain corresponding 2D HE materials such as dichalcogenides, halides, and phosphorus trisulfides.²⁰ Similar method was also reported to prepare (MoWVNbTa)₂S₂ by Cavin et al.¹⁴ However, the mechanical exfoliation method also suffers from some disadvantages. First, the monolayer yield of the exfoliated 2D HE materials is too low, and the product is usually a mixture of single- or few-layer nanosheets. Second, mechanical exfoliation is dependent on the experimenter. Thus the experimental accuracy, controllability, and reproducibility are low, making it difficult to obtain a specific size, shape, and thickness of 2D materials.

2.1.2 | Liquid-phase exfoliation

The key factor in liquid-phase exfoliation is the matching of the surface energy of the lamellar 2D material to that of

the solvent. The smaller the difference in surface energy between them, the more efficient the exfoliation will be. In general, liquid-phase exfoliation usually requires the use of physical and chemical assistance such as ultrasonication, ball milling, and ionic intercalation. For example, the liquid-phase exfoliation of HE MXene is usually accompanied by HF etching of “A” layer in MAX and LiF intercalation.^{13,21} Liquid-phase exfoliation is usually suitable for the large-scale preparation of HE 2D materials. However, the external forces during the liquid-phase exfoliation process will cause the fragmentation of 2D HE materials, resulting in small size, that is, below 5 μm . Besides, the strong force may also result in partial defects in HE 2D materials and affect their properties. Moreover, the exfoliated 2D HE materials have large specific surface area, which will inevitably adsorb solvents, surfactants, and polymers from the exfoliation process, causing contamination of the samples.

2.2 | Bottom-up method

The bottom-up method is based on the principle of constructing 2D HE materials from small molecules. According to the reaction environment, the bottom-up method can be divided into two types: the vapor-phase method and liquid-phase method. Among which, the liquid-phase method has been widely used to prepare 2D HE materials in high yields and quality. Liquid-phase method is a method in which a chemical reaction takes place in the liquid phase under specific experimental conditions. As the preparation reaction occurs in the liquid phase, the thickness and size of 2D HE materials can often be controlled by modulating the reactant precursors and experimental conditions. The 2D HE materials obtained by liquid-phase method have good dispersion in specific solvents and aqueous solutions, which can be used in a wide variety of systems and fields. Typically, the hydro-/solvent-thermal methods and coprecipitation method are two common liquid-phase methods to prepare HE 2D materials.

2.2.1 | Hydro-/solvent-thermal method

Under specific solvents or water as reaction media at high temperatures and pressure, the reactant precursors are dissolved and then react to crystallize and grow 2D HE materials. During hydro-/solvent-thermal reactions, precursor ions, solvent molecules, or surfactants can be inserted between layers during the bottom-up construction of 2D HE materials, resulting in larger layer spacing. For example, Wang et al. successfully developed a new type of HE layered double hydroxides (HE-LDHs) by hydrothermal

methods, and then exfoliated them by plasma etching into ultrathin HE-LDHs nanosheets with abundant defects.²² The resulted ultrathin defective HE-LDHs have five nearly equimolar metal compositions and possess a crystalline single phase with a layered structure. The hydro-/solvent-thermal methods for the preparation of 2D HE materials are simple, which can be scaled up to obtain a high yield of 2D HE materials with low cost.

2.2.2 | Coprecipitation method

Coprecipitation method refers to the addition of a variety of cationic metal salts to the solution so that they exist in a homogeneous phase in the solution. The 2D HE materials can be obtained from a variety of components with homogeneous precipitation. The chemical coprecipitation method not only allows for the refinement and homogeneous mixing of raw materials, but also has the advantages of a simple process, low calcination temperatures, short calcination times, and good product quality. The rapid synthesis kinetics of the coprecipitation process can overcome the thermodynamic solubility limitations of the different elements and facilitate the formation of 2D HE materials. For example, a range of HE hydrotalcites (HEH) from five to nine metal elements were prepared by Zou et al. using coprecipitation method.²³ The ultra-fast mixing and coprecipitation process of the metal salt solution ensures ultrashort reaction times to produce HEH. The fast nucleation and formation process can break the limits of the solubility product constant ($\text{p}K_{\text{sp}}$) of the metal ions and prevent undesired segregation.

Although the liquid-phase methods are simple and can be scaled up to prepare high-yield 2D HE materials with low cost and scalable, the liquid-phase methods are very sensitive to experimental conditions, such as precursor concentration, solvent system, surfactants or polymers, temperature. Moreover, the crystal structure of the obtained HE 2D materials is not complete due to the low reaction temperature of the liquid-phase method and most of the HE 2D materials are multilayered.

2.3 | Topological conversion method

Our group innovatively proposed a new method for the synthesis of monolayer 2D materials via topological conversion method.²⁴ The conversion reaction is carried out by adding, removing, or replacing elements to the reactant precursor in a specific environment (such as high temperature). The structure or morphology of the reaction product is derived from the reactant precursor.^{25,26} The preparation of 2D materials by topological conversion has many advantages compared to traditional topological chemistry

such as good crystallinity and stable structure. Although topological conversion has not yet been reported to be applied to the preparation of HE 2D materials, it is believed that it will be the most promising strategy for the preparation of 2D HE materials soon.

3 | PROPERTY PREDICTIONS

It has been found that the HE 2D materials can inherit the favorable characteristics from their bulk form. Also, other unique properties originated from combination of “HE” and “2D features” are worth to be explored.

3.1 | Enhanced thermal and chemical stabilities of HE 2D materials

The stability of 2D materials is of great importance for various practical applications. It was found that the MXenes tend to be oxidized under humidity and oxygen conditions, which impede their applications in aqueous solution.²⁷ In addition, it is still challenging for 2D materials to maintain the long-term stability during the operation conditions in energy storage and conversion.²⁸ The entropy-stabilization concept may offer an opportunity for 2D materials to achieve high durability even under the harsh thermal and corrosive conditions. In some cases, the increase of metal elements in the atomic layer results in a maximum molar configurational entropy and minimizes Gibbs free energy, forming a relatively stable single-phase solid solution structure.²⁹ Exfoliating the HE layered structure is the most direct strategy to produce HE 2D materials. A $((\text{Ti}_{1/5}\text{V}_{1/5}\text{Zr}_{1/5}\text{Nb}_{1/5}\text{Ta}_{1/5})_2\text{CT}_x)$ with five types of transition metals in MX layers was first reported by Yang's group via selective etching of the HE MAX phases of $(\text{Ti}_{1/5}\text{V}_{1/5}\text{Zr}_{1/5}\text{Nb}_{1/5}\text{Ta}_{1/5})_2\text{AlC}$ (Figure 2A).¹³ As shown in Figure 2B and C, the flexible HE MXene nanosheet is featured with the characteristic (002) peak at $\sim 7^\circ$. Interestingly, the flexible HE MXene exhibits a high thermal stability over 212°C and superior corrosion-resistant behavior than that of monometal (Ti_2CT_x) and bimetal (TiNbCT_x) MXenes.

The layered van der Waal's materials also provide opportunity to get the HE atomic layer due to their weak inter-layer bonding.^{30,31} As illustrated in Figure 2D and E, Cavin et al. put forward a temperature T_0 to assess the stability of HE transition metal dichalcogenides (TMDs), where the T_0 is referred to the temperature of an equimolar HE alloys, above which ΔG of the solid solution is lower than that of all other possible mixtures.¹⁴ Then the authors synthesized the predicted HE TMDC alloy with five components (Mo, W, V, Nb, Ta) via chemical vapor transport method. This concept of entropy stabilization can also be identified in

HE 2D layers, which can be stabilized in severe operation situations such as corrosion, high temperature, and high electrochemical potential. For example, the HE selenide (Ti, V, Cr, Nb, Ta) Se_2 delivers superior robustness than its individual counterparts in acid (HNO_3), base (NaOH), and organic solutions.²⁰

3.2 | Atomically distributed lattice strains of HE 2D materials

The difference in size between each atom in a 2D HE material causes the lattice points to be shifted from their ideal positions. Since each metal in HE atomic layer has the same probability of occupying a lattice position, severe lattice distortions occur when the chemical ordering is neglected. It is well known that the most obvious effect produced by lattice distortion in HE layers is the enhancement of the mechanical properties. Take the HE MXene for example, we investigated the lattice strain of the HE MXene by geometric phase analysis, originated from the atomic-resolution HAADF image (Figure 3A and B). There are strong compressive and tensile lattice strains on HE atomic layers along the e_{xx} and e_{xy} directions, where the compressive strains may be attributed to the implantation of V atom due to its small atom radius. The tensile strains might be originated from the incorporation of Nb, Zr, and Ta into lattice, which possess bigger atom radius. In addition, the strain on the HE-MXene atomic layer is stronger than that on the bimetallic MXene layer, and the strain distribution on HE MXene is more extensive than that on TiNbCT_x . Such superior mechanical strains bring out wide application of HE-MXene on the dendrite-free Li metal anode (Figure 3C). As a result, the homogenous growth of dendrite-free lithium can be achieved on HE-MXenes, exhibiting a better cycling stability (up to 1200 h) than $\text{Ti}_3\text{C}_2\text{T}_x$ (<1000 h), and deep stripping–plating levels at 20 mAh/cm^2 (Figure 3D).

Other alkaline metal anodes, including lithium, sodium, and zinc are also widely studied as anode materials due to their high energy densities.^{32,33} Moreover, the formation of the random dendrites on the metal anodes due to their high electrochemical activity will cause the severe safety issues.^{34,35} Introducing the HE 2D materials with high lattice strain is promising to induce homogeneous nucleation and growth of various metals, improving the cycling stability of metal anodes.

3.3 | Unsaturated multisites and vacancies of HE 2D materials

HE materials attract great attention in catalysis due to their unique electronic structures and tunable

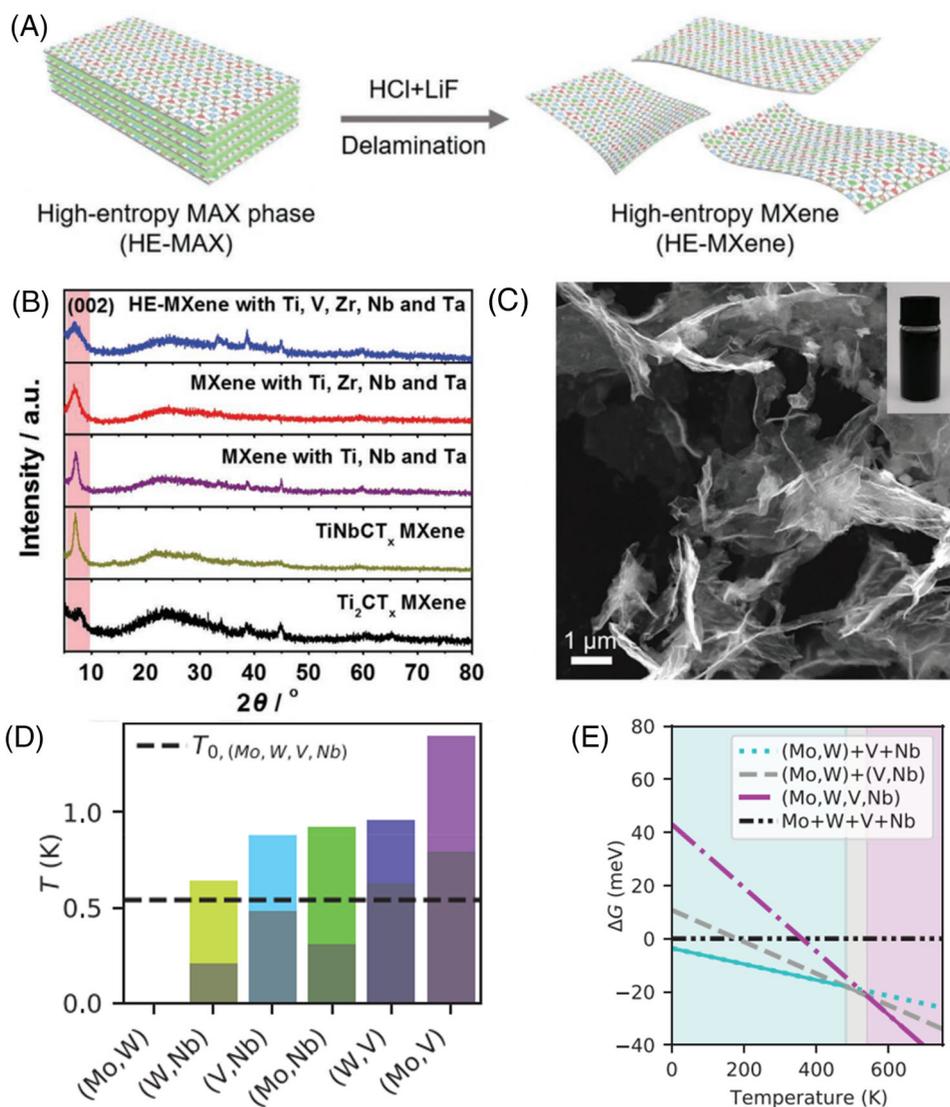


FIGURE 2 (A) Schematic illustration of the preparation of HE-MXene from HE-MAX ($\text{Ti}_{1/5}\text{V}_{1/5}\text{Zr}_{1/5}\text{Nb}_{1/5}\text{Ta}_{1/5}$)₂AlC via the hydrogen chloride/lithium fluoride etching approach.¹³ (B) XRD patterns of MXenes with Ti, V, Zr, Nb, and Ta species, exhibiting the characteristic (002) peak.¹³ (C) SEM, images of HE-MXene atomic layers, showing the ultrathin and single crystalline features. The inset in (C) shows a photograph of the HE-MXene atomic layers dispersed in water at a concentration of 1 mg/mL.¹³ (D) The miscibility temperatures of the six binary alloys with the greyed portions corresponding to T_0 , the temperature where the mixing free energy of the equimolar binary alloy becomes negative. The dashed line corresponds to T_0 for (Mo, W, V, Nb).¹⁴ (E) A plot of the equimolar free energy versus temperature for the partial decompositions that are stable at certain temperatures, the HEA and the mixture of individual components for reference. The solid line corresponds to the most stable phase or phase mixture at a certain temperature¹⁴

components, which have been investigated in HER, OER, CRR, and nitrogen reduction reactions. Most catalytic reactions involve multistep, which are accompanied with different reaction intermediates. Optimizing the adsorption energy is key to improve the kinetics of catalytic reaction.^{36,37} Adding different components into one material can synergistically for catalytic system.^{38,39} The unsaturated sites are often recognized as active sites for a series of catalytic reactions.⁴⁰ The HE 2D layers are not only featured with highly exposed basal plane but also abundant unsaturated multisites at edge, which

are ideal active sites for catalysis (Figure 4A). The 2D (MoWVNbTa)₂S₂ is successfully prepared and studied for the CO₂ reduction to CO, showing a high current density of 0.51 A/cm² at -0.8 V versus RHE, which is 10 times higher than Ag nanoparticles. The authors conducted the first-principles calculations, revealing that the multisites at edge deliver low average energy barriers for CO desorption, which is the rate-determining step in CO₂ conversion to CO (Figure 4B). The calculation results also suggest the V and Nb sites are favorable active sites compared to W and Mo due to their low CO desorption energy.

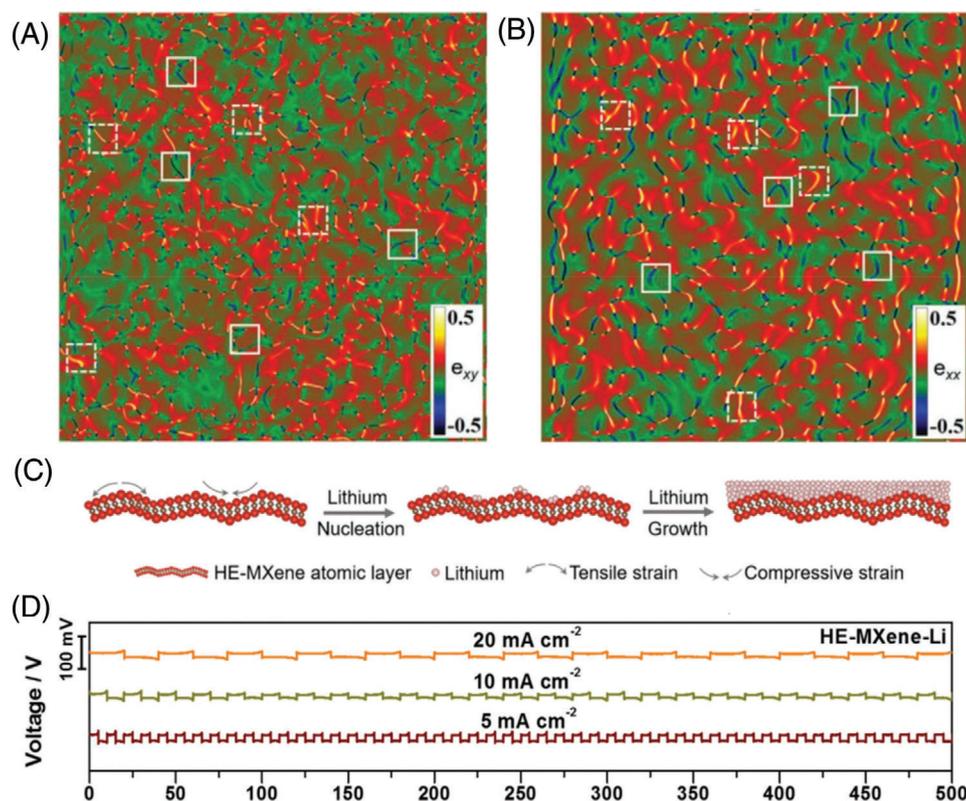


FIGURE 3 Strain distribution of e_{xx} (A) and e_{xy} (B) for HE-MXene derived from $(\text{Ti}_{1/5}\text{V}_{1/5}\text{Zr}_{1/5}\text{Nb}_{1/5}\text{Ta}_{1/5})_2\text{AlC}$, showing strong strains, where the color from green to dark blue represents compressive strain degree and the color from red to bright yellow refers to tensile strain degree in the data bar.¹³ (C) Schematic illustration of the nucleation and growth behavior of lithium guided by strains on HE-MXene atomic layers.¹³ (D) Galvanostatic cycling of HE-MXene-Li at different deep stripping and plating capacities for 5, 10, and 20 mAh/cm² at 1.0 mA/cm²¹³

Our group also reported a HE CN-MXene consisting Ti, V, Zr, Nb, and Ta via selective etching $(\text{Ti}_{1/3}\text{V}_{1/6}\text{Zr}_{1/6}\text{Nb}_{1/6}\text{Ta}_{1/6})_2\text{AlC}_x\text{N}_{1-x}$ MAX phase, exhibiting high catalytic activities for lithium polysulfide (Figure 4C).²¹ As shown in Figure 4D, a high-rate performance of 702 mAh/g at 4C can be achieved by the Li-S battery composed of HE CN-MXene-coated separator toward sulfur cathode, which is superior to those of $\text{TiVC}_{1/2}\text{N}_{1/2}\text{T}_x$ (589 mAh/g), $\text{Ti}_2\text{C}_{1/2}\text{N}_{1/2}\text{T}_x$ (491 mAh/g), HE-MXene (185 mAh/g), and MXene (403 mAh/g). Also, the distinct reduction peaks can be observed at 0.12, -0.15, and -0.5 V for HE CN-MXene (Figure 4E), corresponding to S_8 to S_6^{2-} , S_6^{2-} to S_4^{2-} and S_4^{2-} to S^{2-} , respectively, demonstrating the catalytic potential for LiPSs conversion on HE CN-MXene. It has been also demonstrated that the metals partially bonded with N in HE CN-MXene are crucial for catalyzing the polysulfides conversion. This work highly suggests the advantages of unsaturated multisite in accelerating multistep involved reactions.

The vacancies are also widely studied for a range of catalytic reactions.^{41,42} Owing to the chemical activities' differences of various metals in the HE 2D materials, vacancies and defects can be easily obtained by selective

etching or post-treatment process. After energy dispersive spectrometer (EDS) elemental analysis of the transition metal elements in the prepared HE MXene, we found that a significant decrease in the relative content of Zr and Nb occurred compared to that in the MAX precursors (Figure 5A). Besides, high-angle annular dark field (HAADF) photographs similarly confirmed the presence of vacancies. In addition to the introduction of cationic vacancies in HE 2D materials, anionic vacancies can be achieved as well. For example, Wang et al. reported a low-temperature O_2 plasma etching strategy to synthesize oxygen vacancy-rich HE oxides nanosheets.⁴³ High-energy electrons collide and can transfer energy to oxygen molecules, producing excited state oxygen radicals with higher chemical activity than oxygen molecules. Therefore, the HE layered double hydroxide (LDH) precursors can be transformed into single-phase spinel-type HE oxides using low-temperature O_2 plasma etching. Both the X-ray photoelectron spectroscopy spectra and Electron paramagnetic resonance results confirmed that the prepared HE oxide nanosheets were featured with abundant oxygen vacancies. The low-temperature O_2 plasma technology enables the synthesized HE oxides with nanosheet

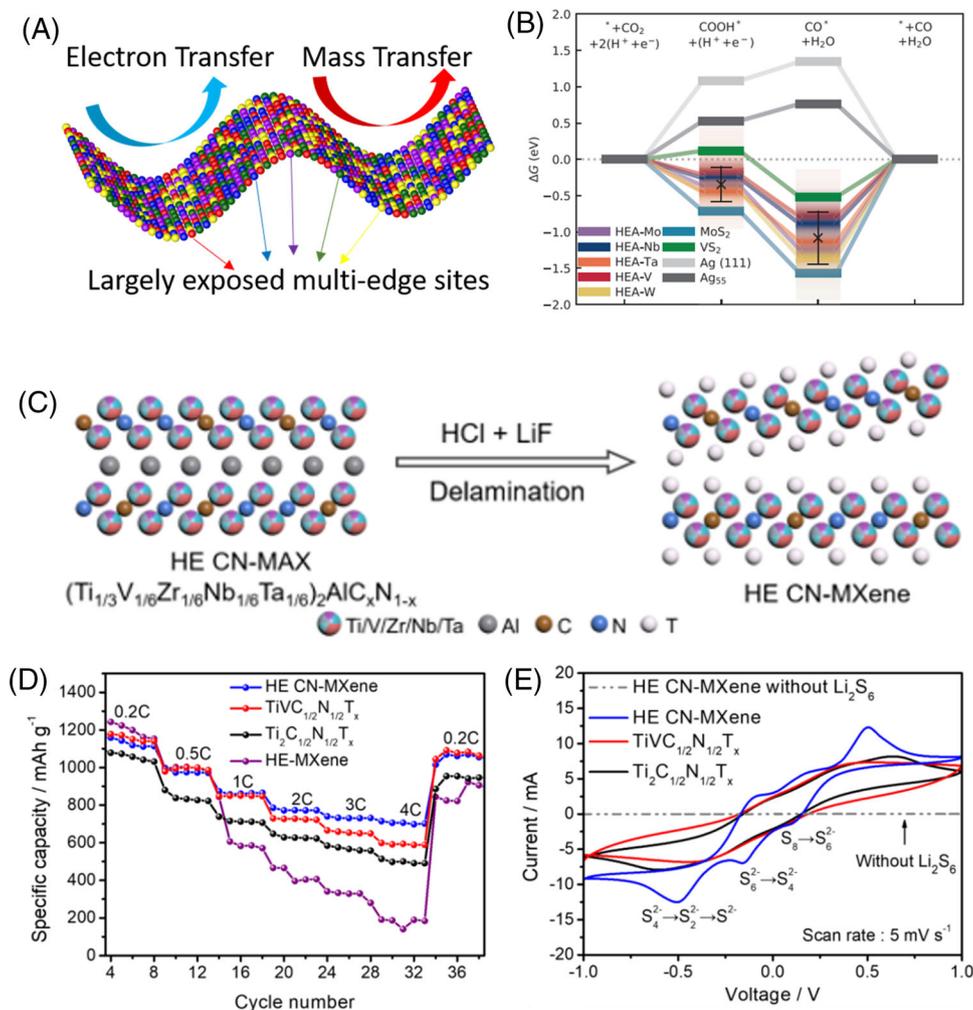


FIGURE 4 (A) The scheme of high-entropy 2D layers with abundant unsaturated multisites. (B) Free-energy pathway of CO_2 reduction at the equilibrium condition potential.¹⁴ (C) Schematic diagram for preparing HE CN-MXene by selectively etching HE CN-MAX in a HCl/LiF mixture and subsequent delamination treatment.²¹ (D) Rate capabilities of Li-S batteries with $\text{Ti}_2\text{C}_{1/2}\text{N}_{1/2}\text{T}_x$, $\text{TiVC}_{1/2}\text{N}_{1/2}\text{T}_x$ and HE CN-MXene functional separators from 0.2 to 4 C.²¹ (E) CV curves of HE CN-MXene²¹

form, plenty of oxygen defects and high specific surface area, improving the electrocatalytic activity. Due to multielements randomly distributed in the basal plane, the different cation vacancies may be generated and exhibit unusual properties.

3.4 | Ultrathin functionalized HE 2D materials

Owing to five or more metallic elements being uniformly distributed in a solid solution state on one or several atomic layers, the HE 2D materials possess ultra-thin thickness. Also, the large exposed basal plane with high surface area enables easy functionalizing with external chemical or physical atmosphere via covalent bonding to modify the properties.⁴⁴ shown in atomic force microscope (AFM)

images (Figure 5B), the obtained HE CN-MXene exhibits a thickness of 1.2 nm. In addition to HE-MXenes, a series of HE van der Waals materials (dichalcogenides, halides, and phosphorus trisulfides) were reported by Qi et al.²⁰ The thinnest area of as-prepared (Mn, Fe, Co, Ni)PS₃ nanosheet can reach to 2.5 nm, approximately the height of four stacking layers (Figure 5C). Besides, HE oxides nanosheets with an average thickness of 20 nm were reported.

Based on different synthesis methods, HE 2D materials with different functional groups can be obtained. For example, by etching the HE-MAX in HCl/LiF mixture solution, HE-MXene terminated with O and F species are successfully prepared by us, which can be confirmed by the Fourier transform infrared spectroscopy (FTIR) spectra (Figure 5D).⁴⁵ The O and F species were also detected for the preparation of HE CN-MXene (Figure 5E). Like previously reported, MXenes with diversified functional

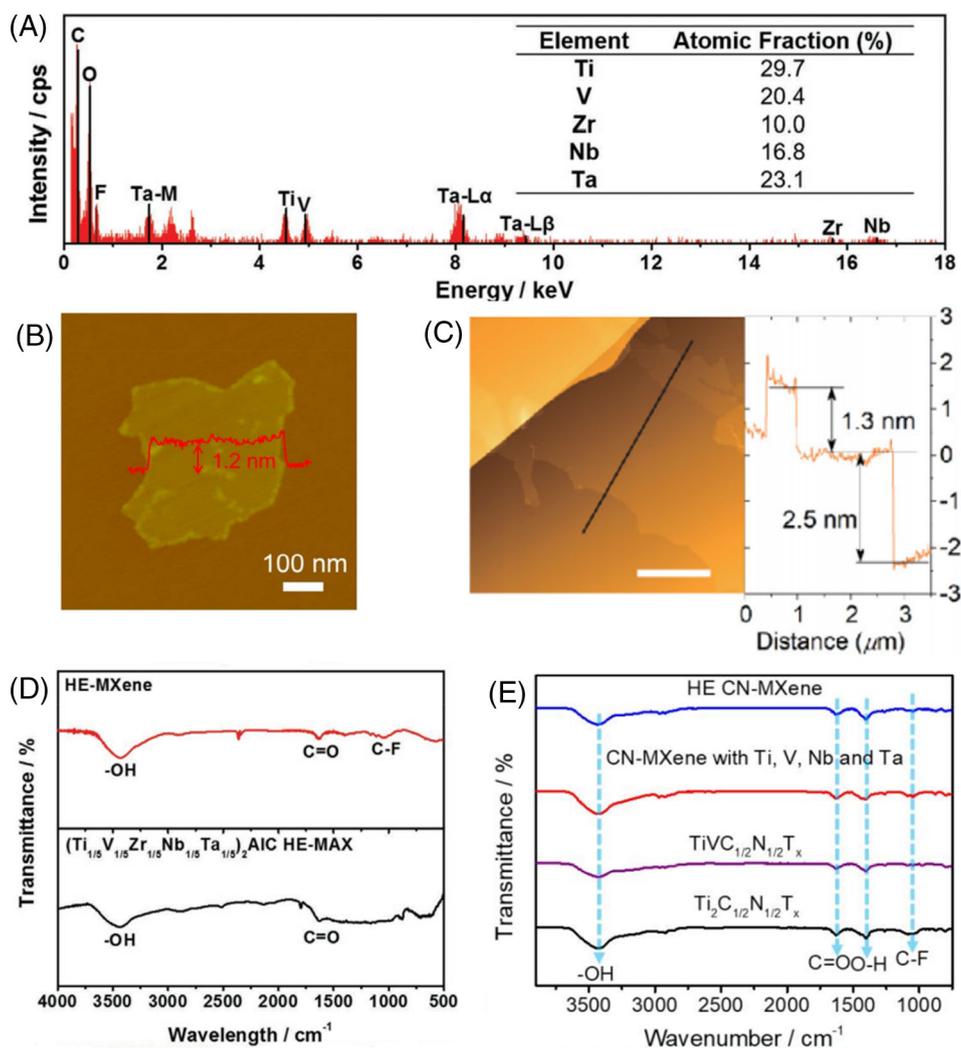


FIGURE 5 (A) The EDS spectra of HE-MXene, disclosing the contents of transition-metal elements in the range of 5–35 at.%.¹³ (B) AFM image and corresponding height analysis of HE CN-MXene, disclosing many ultrathin layers with a thickness of 1.2 nm.²¹ (C) Atomic force microscope (AFM) topographic image of the exfoliated HEX thin films and their cross-sectional profile along the black line.²⁰ (D) FTIR spectra of HE-MAX ($\text{Ti}_{1/5}\text{V}_{1/5}\text{Zr}_{1/5}\text{Nb}_{1/5}\text{Ta}_{1/5}$)₂AlC and the corresponding HE-MXene.¹³ (E) FTIR spectra of HE CN-MXene, showing the characteristic stretching vibrations of -OH, C = O, O-H, and C-F²¹

groups in different etching environment, we believe that it can be further extended to the preparation of HE MXenes containing various functional groups such as -Cl, -Br, -S, -I, or -P.^{46,47}

The surface functions have a great impact on the intrinsic properties, thus, affecting their performance for various applications. It was found that the superconductivity of niobium carbide MXenes can be governed by their surface groups, which is generated from the lattice strain caused by different functional terminations.⁴⁸ Since, the HE-MXenes already possessed the lattice strain, the further change of surface function may lead to unusual superconductivity. Also, an enhancement of electrochemical performance can be observed in oxygen-terminated Nb₂C MXene, which has abundant active sites.⁴⁹ The HE-MXenes have the

potential in various catalytic reactions due to their element's diversity in one solid solution. The change of terminations can further increase the active sites to improve their overall performance.

3.5 | Tunable atomic components of HE 2D materials

In addition to the above-mentioned features, the HE 2D materials also possess other special properties by changing the composition of elements in the atomic layer.⁵⁰ The desired performance can be achieved by adding certain elements.⁵¹ For example, by introducing the Mn, Fe, Co, Ni elements in the HEM, the obtained (Mn, Fe, Co, Ni)PS₃

nanosheets exhibited a rich magnetic behavior. Superconductivity and metal-insulator transition have been also detected in HE tellurides and sulfides, respectively.²⁰ The HE 2D materials not only inherit the unique intrinsic properties from their bulk form, but the 2D features may help to further regulate the properties as well.⁵² Thermoelectric technology generates electricity from waste heat, but suffers from the low performance of thermoelectric materials. Jiang et al. reported an n-type PbSe-based HE materials, showing the figure of (zT) value of 1.8 at 900 K.⁵³ The largely distorted lattices lead to the unusual shear strains with strong phonon scattering, resulting in a much lower lattice thermal conductivity. The lattice distortion is caused by adding the heteroatoms with different radius. The properties can be intentionally tailored by different atoms. This work demonstrates the entropy engineering strategy to improve the thermoelectric behaviors of materials. If the thickness of can be further reduced, the unique band structures can be further regulated, which motivates large amounts of research efforts. Overall, the components variation in HE materials provides plenty of opportunities to obtain the desirable properties of materials, which could be magnified in their 2D systems.

4 | CONCLUSION AND OUTLOOKS

In this perspective, we summarize the recently reported HE 2D materials such as 2D HE MXenes, metal chalcogenides, LDHs, metal oxides. These 2D HEMs with versatile compositions, ultrathin feature, highly exposed surface, abundant surface chemistry, as well as tunable physical and chemical properties, have been widely applied in the fields of electronics, catalysis, and energy storage. We believe that HE 2D materials can be further extended to fabricate many new kinds of atomic layers. But it remains challenging in fabrication and application of HE 2D materials. The investigation of compatibility and diffusion rate of different metal atoms will help to better understand synthesizing mechanism. Although it has been proved that the change of composition of HE 2D materials leads to tailored properties, the identification of the role of each metal elements in various applications is still complicated. As illustrated in Figure 6, it is motivated to explore the underlying reasons and accordingly, take advantage of the unique properties of HE 2D materials for practical applications.

4.1 | Theoretical calculations

As the composition variations make it huge family of HE 2D materials, the established database is key to rationally guide the synthesis of HE 2D materials and regulate their

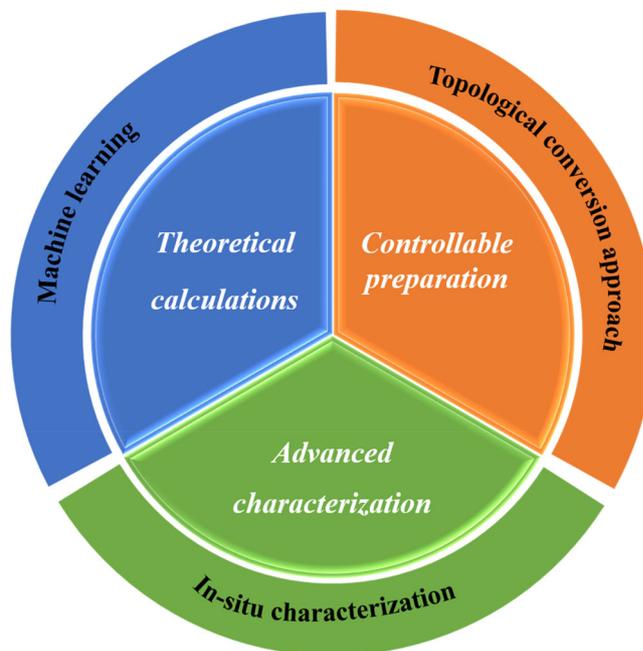


FIGURE 6 Future development of high-entropy 2D materials

properties. The compatibility of different atoms in one solid solution is highly appreciated before the experiments. The machine learning tools aim to predict the possibilities of different elements accommodated in one solid solution. Moreover, the machine learning can also link the material's features and properties, which provide the rules of phase or element selections.

4.2 | In-depth understanding of the formation/working mechanism of HE 2D materials

There are many experimental results, which exhibit that the transition metal layer of MAX phases and 2D MXenes could be successfully converted to 2D TMCs. However, there is no direct evidence and convincing results about the conversion mechanism of HE 2D materials such as the atomic migration path and the variation of transition metal layer number during the conversion process. The intermediate state between the precursors and HE 2D materials should be collected and more advanced characterization methods such as atomic-resolution scanning transmission electron microscopy and ultrathin sectioning characterization associated with focused ion beam, are urgently needed for the following inspection of subtle structure of the intermediates. Also, it is key to identify the main component via in situ techniques when different components perform synergistically, which helps to further optimize the performance of HE 2D layers in various applications.

4.3 | Finding new ways to synthesize HE 2D materials and enrich the system

Currently, the reported methods for synthesizing HE 2D materials mainly focus on mechanical exfoliation, liquid-phase exfoliation, and hydrothermal methods, resulting in a small lateral size due to the anisotropy of the stresses on the atoms in all directions of the plane. Topological conversion method is a promising method to prepare HE 2D materials. For example, topological conversion of layered non-van der Waals solid HE MAX phases under high temperatures in an atmosphere containing sulfur group atoms is likely to yield a series of 2D HE transition metal sulfides with high monolayer rates and controlled phases. Besides, the system of HE 2D materials are mainly limited to MXenes, TMDs, LDHs, and metal oxides, many other HE forms of 2D materials have not been reported such as metal organic frameworks MOFs.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Yanglansen Cui: writing and editing. **Yongzheng Zhang:** writing and editing. **Zhenjiang Cao:** writing and editing. **Jianan Gu:** editing. **Zhiguo Du:** editing. **Bin Li:** editing. **Shubin Yang:** conceptualization, organization, writing and editing.

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