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Research Progress of High Entropy Ceramic Materials

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Abstract

High-entropy materials (HEMs) have better mechanical, thermal, and electrical properties than traditional materials due to their special "high entropy effect". They can also adjust the performance of high entropy ceramics by adjusting the proportion of raw materials, and have broad application prospects in many fields. This article provides a review of the high entropy effect, preparation methods, and main applications of high entropy ceramic materials, especially exploring relevant research on high entropy perovskite ceramics. It is expected to provide reference for the promotion of scientific research and the development of further large-scale applications of high-entropy ceramic materials.

Keywords: *High-entropy ceramic materials; high entropy effect; preparation method; perovskite structure; research prog*

1 Introduction

With the rapid development of science and technology, higher requirements are being placed on the performance of materials. For ceramic materials, a single principal component ceramic material can no longer meet the performance requirements under harsh usage conditions. Inspired by the development of high-entropy alloys, scientists have introduced the concept of high-entropy into the field of ceramics. The concept of HEM originates from high entropy alloys (HEAs) In 2004, Ye [1] innovatively proposed HEAs. This alloy is typically made up of five or more metallic elements in equal or almost equal proportions. It is a single structured solid solution with multiple main elements. Its structure is ordered long-range, but its components are disordered. This new discovery breaks through the traditional design concept of single principal component materials and provides new ideas for the design of new materials. As the concept of high entropy gradually improves in the field of alloys, it is gradually introduced into the field of other materials.In 2015, Rostet al [2]. at North Carolina State University, one of the pioneers in the field of high entropy in the United States, successfully synthesized ($Mg_{0.2}Co_{0.2}Ni_{0.2}Zn_{0.2}Cu_{0.2}$)O. On considering the influence of the configurational entropy on its structure, it was named entropy stable oxide (ESO). After that, researchers at home and abroad have successively studied various high-entropy systems, including fluorite-structured [3], perovskite-structured [4], spinel-structured high-entropy oxide ceramics [5], as well as non-oxide high-entropy ceramics, such as nitrides [6], borides, and so on [7]. Although the research on high-entropy ceramics is hot, it is still in its infancy, and there is still a lot of work to be done.

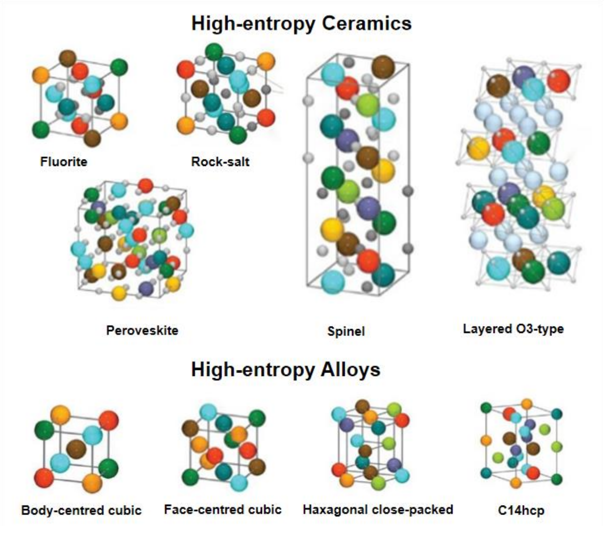

Figure 1 Structure of several high-entropy ceramics and alloys

2 Core Effects of High-entropy Materials

2.1 Thermodynamic high-entropy effects

The "entropy" of high-entropy ceramics is the configuration entropy (ΔS_{mix}), whose relationship with the number of components in the HEMs is shown in

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Figure 2 (a).According to Boltzmann's assumptions about the change in entropy and the degree of chaos, the greater the variety of elements [8], the greater the degree of chaos. If the effects of atomic vibrational grouping, electronic grouping, magnetic moment grouping, and other factors are considered at this point, the entropy change of HEMs is greater. Multiple major elements in each high-entropy material can be added in equal or unequal molar ratios, which leaves a wealth of scope for the design of high-entropy materials.The high-entropy effect can be used to explain the formation of HEAs with multiple principal elements as shown in Figure 2(b). On the one hand, the high mixing entropy at high temperature can effectively reduce the Gibbs free energy of the alloy system, thus stabilising the generated single phase; on the other hand, high entropy can reduce the electronegativity difference, inhibit the formation of compounds, and promote the mixing between elements. The solid solution phase in HEA will produce a strong solid solution strengthening effect, which can significantly improve the mechanical properties of the alloy such as strength and hardness [9].

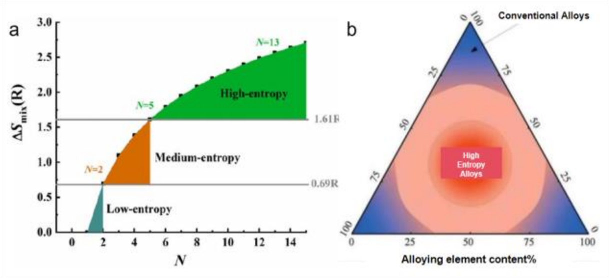

Figure 2 (a) Conformational entropy vs. number of group elements; (b) Relationship between the content of high entropy alloy element [9]

2.2 Effects of lattice distortion of structures

Unlike the lattice distortion of conventional materials [10], the elements in HEMs are numerous and often exist in equal molar ratios, so that all types of atom have the same chance to occupy fixed positions on the lattice. Since the various types of will be randomly distributed in the crystal structure, the different sizes of the atoms and the large difference in chemical bonding will cause more serious lattice distortions within the structure of the high-entropy materials. If the sizes of the atoms are very different, the lattice will not be able to maintain a normal structure because of the high degree of distortion, leading to the generation of amorphous phases.

2.3 Hysteresis-diffusion effects of dynamics

During the alloy material moulding process, the atoms will gradually converge from the chaotic arrangement in the molten state, and after the synergistic coordination and diffusion of the components, there will be a split phase, leading to easy nuclei formation in the alloys but not easy to grow [11].In high-entropy alloys, the large difference in atom size, the greater variety of elements, the more complex structure, the slower rate of internal diffusion and phase transitions and therefore the precipitation of nanometer-sized grains, which is not possible in conventional alloys, is often observed. This phenomenon results in HEAs that do not undergo grain growth and recrystallization at elevated temperatures, which has the potential to greatly improve the corrosion resistance of alloyed materials.

2.4 Cocktail effect

For HEMs, the basic properties of different components and the interactions between them can lead to the emergence of more complex properties of high-entropy materials. By changing the amount and type of added components, some of the properties of the material can be regulated, and even new certain properties can appear.The high-entropy effect allows high-entropy ceramics to remain single phase at extreme temperatures, pressures and chemical environments [9]. The kinetic hysteretic diffusion effect allows high entropy ceramics to be synthesized at high temperatures and maintain structural stability at room temperature. The kinetic hysteretic diffusion effect also leads to the formation of amorphous or nanocrystalline structures in high entropy ceramics, which endows them with corrosion-resistant properties. The low amorphous thermal conductivity is related to lattice distortion effects, and, together with the high hardness, it promotes the development of materials for thermal and environmental barrier protection. The lattice distortion induced by many elements of different sizes produces a solid solution strengthening leading to an improvement in their mechanical properties [12], mainly by preventing dislocation motion and modifying and eliminating the lattice slip regime through the resulting nanograin microstructure [13]. The suppression of grain coarsening at high temperatures has also been attributed to lattice distortion (LD) effects, where the distortion of the lattice increases the crystallization energy of the crystals, thus reducing the free energy gained by shrinking the surface area of the grains.

3 Method of Preparation of High-entropy Ceramic Powder

3.1 Solid phase reaction method

The solid-state reaction method often uses mechanical alloying technology to prepare powders, starting from elemental metal or metal ceramic powders. In this process, by using different types of metal ceramic powders and adding alcohol or stearic acid as process control agents, high energy is applied through high-energy ball milling equipment to produce partially or completely mechanically alloyed high-entropy ceramic powders. Wang et al [14]. successfully prepared single-phase (Hf, Zr, Ta, Nb, Ti) (C, N) with high entropy using various oxide powders in a flowing nitrogen environment, after high energy ball milling to provide higher energy to the powder, at temperatures above 1400 °C. This powder has a single-crystal cubic structure and high compositional uniformity. As the reaction temperature increases, the grain size of high-entropy carbonitrides increases, with the average particle size increasing from 0.31 to 1.26 μm. In addition, various anions can be introduced through solid-state reactions to adjust the composition and content. For example, amorphous BCN powder can be obtained by mechanically alloying equimolar graphite powder with metal nitrogen boron compounds for 10 h. After 24 h of high-energy ball milling, a nano-high entropy ceramic powder with a face-centered cubic rock salt structure was finally formed.

3.2 Preceramicpolymer pyrolysis

Although the solid phase reaction method is widely used in material preparation, it also has some shortcomings, such as the high reaction temperature, long reaction time, difficulty in accurately controlling the proportion of the product, and easy introduction of impurities during the preparation process [15]. To overcome these challenges, researchers have developed a new method for synthesizing ceramics with high entropy under relatively mild conditions, drawing on the strategy of precursor preparation of ceramics. This method includes the use of the sol-gel method and coprecipitation method to realize the mixing of raw materials at the atomic level, thereby reducing the energy required for the synthesis of HEMs. Moreover, sufficient calcination is carried out at relatively low temperatures to remove excess crosslinking agents, precipitants, or solvents, achieving a low-temperature synthesis of high-entropy ceramics. Compared to the atoms in solid powders, the powder prepared by precursor conversion has a significant improvement in atomic diffusion rate, resulting in higher entropy ceramic powders with more uniform composition distribution and more unified crystal structure. The precursor pyrolysis method not only has the advantages of mild reaction conditions, controllable product particle size, and purer products but also faces problems such as low yield, complex process, high equipment requirements, and difficulty in reinterring product particles.

3.2.1 Sol-gel method

The Sol-gel method is a technology that uses compounds containing highly chemically active components as precursors. In this method, the raw materials are mixed uniformly in the liquid phase and undergo hydrolysis and condensation chemical reactions to form a stable transparent sol system in the solution. With the passage of time, the colloidal particles in the sol gradually polymerize slowly to form a three-dimensional network structure of gel [16], which is filled with solvent that has lost its mobility. After drying and sintering solidification processes, materials with molecular to nanoscale substructures were finally prepared. Zhang et al [17]. successfully prepared ($La_{0.2}Y_{0.2}Nd_{0.2}Gd_{0.2}Sr_{0.2}$) CrO_3 ceramic powders ($La_{0.2}Y_{0.2}Nd_{0.2}Gd_{0.2}Sr_{0.2}$) using the sol-gel method. This method not only overcomes the limitation that the 34conventional solid phase method requires high-temperature calcination at more than 1300 ℃, but also realizes the fine and uniform distribution of powders and the uniform dispersion of elements.

3.2.2 Coprecipitation method

The coprecipitation method is a technology that adds an appropriate precipitant to the electrolyte solution containing various ions, to initiate the reaction to generate homogeneous precipitates, and then thermally decomposes these precipitates to obtain high-purity nano powder materials. The advantages of this method are twofold. First, it can be directly obtained from nanopowder materials with uniform chemical composition through chemical reaction in solution. Second, the method facilitates the preparation of nanopowder materials with small particle size and uniform distribution.Wang et al [18]. successfully prepared multi-metal MOF precursors by chemical precipitation method at room temperature, and obtained the general formula through the treatment with high entropy alloy CoNiCuMnAl@C nanocatalysts. This catalyst not only exhibits excellent redox reaction (OER) performance but also exhibits excellent durability.

3.3 Molten salt method

The molten salt method uses various low melting point salts as reaction media to dissolve the reactants in the molten salt for the reaction. Compared to the diffusion rate of atoms in solid-phase powders, the diffusion rate of salt atoms in the molten state at high temperatures is significantly increased, which can significantly shorten the preparation time of powders. After sufficient reaction, the salt substances were dissolved and wash with appropriate solvents to obtain HETMCC powder. This method not only has a simple process and low synthesis temperature, but can also obtain powder with uniform composition. Chu et al [19]. synthesized nano ($Ta_{0.25}Nb_{0.25}Ti_{0.25}V_{0.25}$) C with single-phase rock salt structure using four types of metal powders and carbon powders as raw materials and potassium chloride as molten salt medium under first-principles guidance at 1300 ℃. The powders exhibited good compositional uniformity.

4 The Application of High Entropy Ceramics

The high entropy effect significantly improves the mechanical performance of high entropy ceramics (HECs), pushing them beyond the traditional boundaries of mixture rules and theoretical predictions [20]. This emerging paradigm in material design, coupled with infinite possibilities of component and microstructure combinations, provides HECs with performance characteristics that traditional materials cannot match. The potential applications of this family of materials are extensive, spanning various fields from structure to functionality. Specific applications include ultra-high temperature thermal protection and insulation for hypersonic aircrafts, thermal protection and environmental barrier coatings for engine components [21], radiation-resistant materials for nuclear facilities [22], wear-resistant coatings for cutting tools, materials for electromagnetic wave absorption and interference shielding [23], anodes for rechargeable batteries, catalysts for clean energy and environmental protection, thermoelectric devices, and supercapacitors, as illustrated in Figure 3.

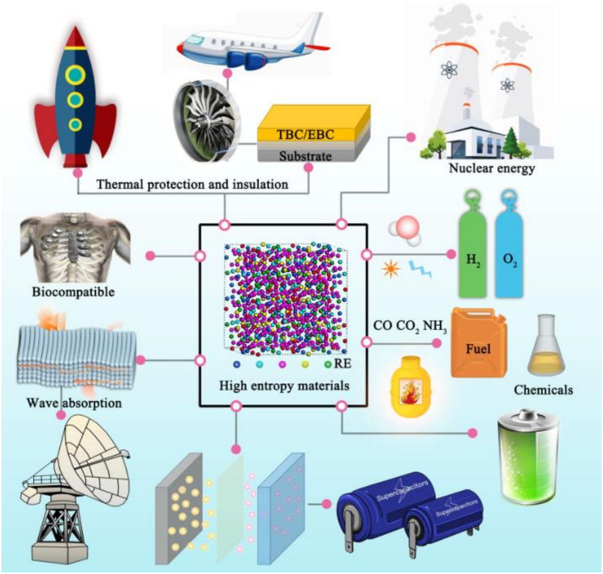

Figure 3 From applications in ultra-high temperature structures to energy and catalytic functional applications

4.1 Supercapacitor

Supercapacitors are currently the world's largest mass-produced double-layer capacitors, mainly divided into carbon-based, metal oxide, and polymer supercapacitors based on electrode materials. Among them, carbon-based supercapacitors are particularly outstanding due to their wide applications, and their performance mostly depends on factors such as the characteristics of carbon materials, the specific surface area of electrode materials, particle size distribution, conductivity, and electrochemical stability. By introducing HEMs into carbon-based electrode materials, it is possible to increase the active sites and surface area, improve the electrochemical stability, and thus achieve higher capacity and current density. Jin et al. successfully synthesized (CrMoNbVZr) N using a mechanochemical soft urea method, with a specific capacitance between 230 and 54 F/g, significantly better than carbon-coated tin oxide nanofibers (118 F/g) under the same conditions.

4.2 Thermoelectric materials

When disorder is introduced into the material, its thermal conductivity can be effectively reduced [25]. This reduced lattice thermal conductivity makes high-entropy ceramic semiconductors ideal materials in the field of thermoelectric. Zhang et al [26]. Successfully prepared a novel high-entropy perovskite-type ceramic $(Ca_{0.2}Sr_{0.2}Ba_{0.2}La_{0.2}Pb_{0.2})TiO_3$ (abbreviated as CSBLP) using the solid-state reaction method. This high-entropy ceramic exhibits both long-range structural order and short-range chemical disorder, with uniformly distributed nanosized grains ranging from 4 to 6 nanometers in its microstructure. Benefiting from the increased configurational entropy, the CSBLP ceramic demonstrates a high Seebeck coefficient (| S | 272 μV/K at 1073 K) and low thermal conductivity (κ is 1.75 W/m•K at 1073 K) after annealing at 1300 °C. This study demonstrates the feasibility of effectively reducing the thermal conductivity of thermoelectric oxides and improving their thermoelectric performance through the design of high-entropy compositions.

4.3 Catalyzer

High-entropy alloys, because of their excellent corrosion resistance, have surpassed traditional transition-metal alloys and find wide applications as electrocatalysts in acidic or alkaline environments. These alloys exhibit good catalytic performance in reactions of oxidation (methanol, ammonia, and carbon monoxide), decomposition (ammonia), and reduction (oxygen). The different atomic bonding in high-entropy ceramics provides outstanding catalytic activity, and by adjusting their unique geometric structure or pore structure to increase surface area, their performance can be further enhanced. Riley et al [27]. synthesized high-surface-area high-entropy oxides (HEO) (CeLaPrSmY) O_2-y using a sol gel method in the presence of polymer complexing agents, followed by calcination at lower temperatures. This high-entropy ceramic maintains a single-phase structure even at high temperatures and exhibits activity without the need for expensive platinum group metals. Furthermore, Riley et al. also compared the catalytic performance of (CeLaPrSmY)O_2-y synthesized by solid-phase synthesis with that synthesized by the sol-gel method with CeO_2 in the oxidation-reduction reaction of carbon monoxide. The results showed an improvement in the catalytic activity of both HEOs; compared to the solid-state samples, the sol-gel method-prepared HEOs exhibited higher activity because of their larger surface area.

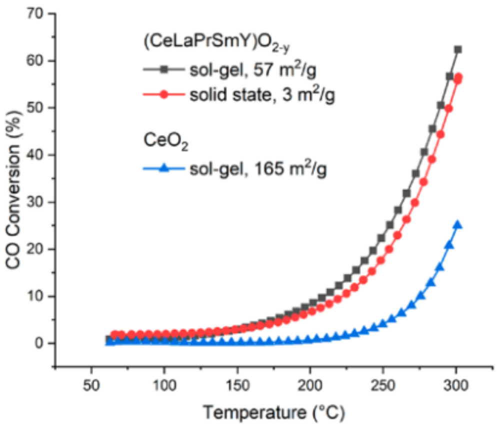

Figure 4 CO oxidation activity of (CeLaPrSmY) O_2 y solid and sol gel samples and CeO_2 sol gel samples with the corresponding specific surface area

4.4 Thermoelectric materials

The lower lattice thermal conductivity of HEMs makes it possible for them to be used as semiconductors in thermoelectric devices. In 2018, Roychowdhury et al [28]. successfully prepared a high entropy selenide (AgBiGe) Se with a single phase rock salt structure in a closed environment using a melting reaction method and studied it using GeSe and $AgBiSe_2$ as raw materials. The experimental results show that at 677 K, its optimal thermoelectric value ZT reaches the maximum value of 0.45, demonstrating its excellent thermoelectric performance. And at 677 K, the thermal conductivity of the material is 3.8 W/m ·K, which drops to 0.43 W/m ·K at 300K.

4.5 Dielectric materials

In high-entropy ceramics, multiple sublattice structures exist, with each structure being occupied by multiple elements. These sub-lattices exhibit not only long-range periodicity, but also their structural distortions affect electronic behavior, dipole response, and band structure. Simultaneously, the random distribution of elements leads to compositional disorder, thereby reducing short-range order. This characteristic makes the dielectric properties of the material easy to manipulate. The dielectric constant, as an indicator of the degree of polarization of dielectric materials in an external electric field, increases with a stronger polarization degree. Zhou et al [29]. Studied the dielectric properties of the high entropy perovskite oxide $Ba(Zr_{0.2}Ti_{0.2}Sn_{0.2}Hf_{0.2}Me_{0.2})O_3$ (where Me represents Nb^{5+} and Ta^{5+}). They found that within the frequency range of 1 kHz to 1000 kHz, the dielectric constant and dielectric loss decreased slightly with increasing frequency. Within the temperature of 303 K～473 K, this material exhibited good dielectric constant and loss stability at specific frequencies. Specifically, within the frequency range of 1 kHz to 1000 kHz, the dielectric constant of $Ba(Zr_{0.2}Ti_{0.2}Sn_{0.2}Hf_{0.2}Me_{0.2})O_3$ ranged approximately 90～113, while the dielectric loss remained between 120～140.

5 Research Progress in High Entropy Perovskite Ceramics

The basic structure of the perovskite oxides is ABO_3, which is similar to HEA. In this structure, the anionic sublattice is occupied by oxygen elements, and the introduction of different cations into the cationic sublattice will complicate the structure. Its calculation method is:

$$\Delta \text{Smix} = \text{-R}[(\sum_{i=1}^{n} c_i \ln c_i)_{cation-site} + (\sum_{i=1}^{n} c_i \ln c_i)_{anion-site}]$$

n is the number of constituent elements, ci is the molar fraction of the i-th component, and R is the gas constant. The difference from HEAs is that the unique A and B cation sites in the perovskite structure need to be considered separately. Due to the ability of perovskite structures to accommodate a large number of different cation substitutions, different systems can be constructed by the high entropy of a single cation site or the simultaneous high entropy of two cation sites [30]. Current research mostly focuses on high-entropy systems with single-cation sites.

Jiang et al. [31] prepared a high entropy perovskite oxide (HEPO) system for the first time, fixing Sr and Ba at the A site and trying to fix four elements and replace the other four elements at the B site, resulting in a total of 13 high entropy perovskite ceramics at the B site. By analyzing the difference in particle size (& B) and tolerance factor (t), it was found that a t value less than 1.03 is a necessary condition for the formation of a single phase structure. Early research mainly focused on the synthesis methods and structural characterization of HEPOs. Chellali et al. [32] and Sharma et al. [33] respectively used atomic probe microscopy (APT) and electron energy loss spectroscopy (EELS) techniques to observe the powders prepared with high entropy perovskite oxide (HEPO) and single crystals, and found that all elements were uniformly distributed at the atomic level. Sakar et al. [34] successfully synthesized five high entropy perovskite ceramics at sites A and B, as well as a kind of ten-element system. The existence of an entropy stabilization effect in HEPOs was demonstrated through cyclic heat treatment in (5A0.2) MnO_3. Zhong et al [35]. proposed a new concept through phase diagram calculations, stating that the "high entropy" characteristics of HEPOs involve multiple species such as anions, cations, and defects, and their component concentrations are difficult to directly determine, which is fundamentally different from HEAs.

Afterward, researchers changed their focus from exploring the basic structure of HEPOs to exploring their performance. Zhang et al. [26] prepared thermoelectric ceramics $(Ca_{0.2}Sr_{0.2}Ba_{0.2}La_{0.2})$ TiO_3 with low thermal conductivity using phonon engineering techniques.

Thermal conductivity at 1073 K was 2.5 W/(m·K), and thermal conductivity was further reduced to 1.75 W/(m·K) by introducing Pb at the A site. In the field of dielectric materials, high-entropy A-site perovskite materials have also received widespread attention. Zhong et al. [35] pointed out in their study that the lattice distortion present in high-entropy perovskite oxides of site A significantly affects their antiferroelectric phase transition behavior. Xiong et al [36]. observed that $(Ca_{0.25}Sr_{0.25}Ba_{0.25}Pb_{0.25})\ TiO_3$ ceramics exhibit low loss (tan) within 25～125 ℃, The coexistence of long-range ferroelectricity and quasi relaxation behavior. Pu et al [37]. simultaneously found 1.02 J/cm^3 in $(Na_{0.2}Bi_{0.2}Ba_{0.}2Sr_{0.2}Ca_{0.2})$ TiO_3 ceramics $(Na_{0.2}Bi_{0.2}Ba_{0.2}Sr_{0.2}Ca_{0.2})$ The high discharge energy density and the significant adiabatic temperature change of 0.63 K introduce high entropy perovskite oxides (HEPO) into the field of electric heating for the first time. Liu et al [38]. observed good thermoelectric properties and adiabatic temperature variation in TiO_3 ceramics in a hexagonal system $(Bi_{1/6}La_{1/6}Na_{1/6}K_{1/6}Sr_{1/6}Ba_{1/6})$ ΔT_{max} reaches 0.14 K. In addition, Wang et al. [39] successfully prepared relaxation ferroelectric ceramics of $(Ca_{0.25}Sr_{0.25}Ba_{0.25}La_{0.25})$ TiO_3 using flash firing technology. The material has a slender hysteresis loop and a discharge energy storage density of 0.684 J/cm^3. Although research on the structure and properties of high-entropy perovskite oxides is still in its early stages, they have shown broad application prospects in the field of dielectric ceramics.

6 Conclusion and Outlook

Compared to traditional ceramic materials, high entropy ceramic materials exhibit significant advantages in corrosion resistance, oxidation resistance, thermal stability, and high hardness, making them widely applicable in fields such as national defense, aerospace, and new energy. However, the preparation and theoretical research of high-entropy ceramics still face some challenges:

(1) Synthesis difficulty: The synthesis of high entropy ceramics usually requires complex preparation conditions, such as high temperature, high pressure, or specific gas environments, which limit their wide range of applications.

(2) Cost issue: The production of high-entropy ceramics often relies on multiple rare or expensive elements, which may result in production costs much higher than those of traditional ceramics.

(3) Performance variability: Although some ceramics with high entropy exhibit excellent performance, not all combinations can achieve the expected results, and the consistency and predictability of performance still need to be improved.

(4) Insufficient theoretical research: The theoretical foundation for the stability and performance of high-entropy ceramics is not yet mature, and more theoretical and experimental research is needed to fully understand their behavior and properties.

In summary, although high-entropy ceramics have shown many promising application areas as an emerging material category, they still face significant challenges. Future research work should focus on improving preparation techniques, reducing production costs, and deepening the understanding of the essential properties of these materials.

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