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Classification, Properties and Applications of High Entropy Ceramics

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Abstract: High-entropy ceramics have garnered growing attention in recent years, primarily due to their outstanding performance under extreme conditions. First formally proposed and defined in 2004, the study of high-entropy ceramics has since seen significant advancements. Through continuous research efforts, a diverse range of high-entropy ceramics has been successfully synthesized, each demonstrating distinct performance characteristics. To facilitate researchers' quick understanding and further study of high-entropy ceramics, this paper reviews the research progress of high-entropy ceramics from the perspectives of their classification and properties.

Keywords: high entropy ceramics, classification, application

1. Introduction

In 2004, the research realm of high-entropy materials came into being. At this time, the initial research on high-entropy alloys was made public. Before long, the research scope extended from high-entropy alloys not only to medium-entropy alloys but also to ceramics, polymers, and composite materials [1]. High-entropy materials stabilize multi-component (typically ≥ 5) equimolar or near-equimolar mixtures by maximizing configurational entropy, forming single-phase solid solutions. Unlike traditional materials, the properties of high-entropy materials often exceed the simple sum of their components and even generate new characteristics. High-entropy ceramics were first achieved in oxides in 2015 and subsequently expanded to borides, carbides, nitrides, sulfides, etc. They are characterized by high-symmetry crystal structures (such as rock salt structure and fluorite structure), slow kinetics, lattice distortions, and stability in extreme environments (high temperature, high pressure, and corrosion) [2]. The superior performance of high-entropy ceramics is mainly influenced by four major effects: the high-entropy effect stabilizes solid solutions, severe lattice distortion enhances dislocation resistance, diffusion retardation inhibits phase transformation, and the cocktail effect synergistically optimizes properties. These effects were first proposed in 2006 [3]. The high-entropy effect simplifies the microstructure by maximizing configurational entropy, while lattice distortion and diffusion retardation jointly ensure high-temperature stability and mechanical strength. The cocktail effect achieves functional optimization (such as catalysis and magnetism) through the synergy of multiple elements. Despite existing controversies (such as the necessity of single-phase), experiments and calculations have shown that these effects are universal in systems such as ceramics and polymers. In the future, by integrating high-throughput design and machine learning, high-

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entropy materials will find more extensive applications in fields such as aerospace and energy catalysis and will also drive the transformation of materials science from "empirical trial and error" to "rational design" [4]. This review systematically classifies HECs based on chemical composition and crystal structure, evaluates their properties, and discusses factors influencing their performance.

2. The Classification System of High-Entropy Ceramics

High-entropy ceramics (HECs) are a rapidly expanding class of materials characterized by their compositional complexity and entropy-stabilized single-phase structures. Classifying HECs is critical for understanding structure-property relationships and guiding targeted applications. This section systematically categorizes HECs based on “chemical composition” and “crystal structure,” integrating experimental and theoretical insights from recent studies.

2.1. Classification Based on Chemical Composition

The chemical diversity of HECs arises from the incorporation of multiple cations or anions into a single-phase lattice. This section categorizes HECs into six major groups based on their dominant chemical constituents: oxides, carbides, nitrides, borides, silicides, and sulfides.

2.1.1. High-Entropy Oxides (HEOs)

High-entropy oxides are the most extensively studied class of HECs, pioneered by the entropy-stabilized oxide (Mg, Co, Ni, Cu, Zn)O in 2015[5][6]. They typically combine five or more metal cations in a single oxide lattice, stabilized by high configurational entropy. Rock-salt oxides include (Mg, Co, Ni, Cu, Zn)O and $(Fe, Co, Ni, Mn, Cr)_3O_4$, which exhibit cubic close-packed structures with random cation occupancy [5][7]. These materials are widely used in thermal barrier coatings and battery electrodes due to their thermal stability and defect-tolerant ionic transport [8]. Perovskite oxides are high-entropy perovskites like $(La, Pr, Nd, Sm, Gd)CoO_3$ and $(Ba, Sr, Ca, Pb, La)TiO_3$ feature a distorted ABO_3 structure, enabling tunable dielectric and catalytic properties[9]. Their oxygen vacancy engineering is critical for applications in solid oxide fuel cells and thermoelectrics [10]. Fluorite oxides are entropy-stabilized fluorites such as $(Y, Yb, Er, Lu)_2(Zr, Hf)_2O_7$, which exhibit defective fluorite structures with low thermal conductivity, making them ideal for environmental barrier coatings [11].

2.1.2. High-Entropy Carbides (HECCs)

HECCs are composed of multiple transition metal carbides (e.g., TiC, ZrC, HfC, NbC, TaC) in equimolar or near-equimolar ratios. Their rock-salt crystal structure and ultrahigh melting points (>3,000°C) make them suitable for extreme environments [6]. Notable examples include (Ti, Zr, Hf, Nb, and Ta)C, which exhibits enhanced hardness (up to 30 GPa) and fracture toughness compared to binary carbides, attributed to lattice distortion and solid-solution strengthening [9]. The incorporation of Group VI metals improves oxidation resistance while maintaining high-temperature mechanical stability.

2.1.3. High-Entropy Nitrides (HENs)

HENs combine multiple transition metals (e.g., Al, Cr, Ti, Ta, Nb) in a nitride matrix. They are widely used as wear-resistant coatings and diffusion barriers due to their high hardness and chemical inertness. Examples include (Al, Cr, Ta, Ti, Zr)N, cubic B1-structured coatings with hardness exceeding 40 GPa, achieved via reactive magnetron sputtering [6], and (Ti, Nb, V, Mo, W)N, which demonstrate tunable electrical conductivity for applications in semiconductor devices.

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2.1.4. High-Entropy Borides (HEBs)

HEBs, such as (Ti, Zr, Hf, Nb, Ta)B_2 and (Cr, Mo, W, Ta, Nb)B_2, adopt hexagonal AlB_2-type structures. Their ultrahigh hardness (25–35 GPa) and exceptional thermal stability (>2,500°C) position them as next-generation ultrahigh-temperature ceramics (UHTCs)[8].

2.1.5. High-Entropy Silicides (HESs)

HESs like (Mo, Nb, Ta, Ti, W)Si_2 and (Cr, V, Nb, Ta, Mo)Si_2 crystallize in hexagonal or tetragonal structures. These materials exhibit excellent oxidation resistance at high temperatures and are explored for aerospace engine components [6].

2.1.6. High-Entropy Sulfides (HESu)

Emerging HESu systems, such as (Co, Fe, Ni, Cu, Zn)S and (Mo, W, Re, Cr, Mn)S_2, leverage sulfur's flexibility in forming layered or pyrite-type structures. These materials show promise in thermoelectric and catalytic applications due to their low thermal conductivity and tunable electronic properties [9].

2.2. Classification Based on Crystal Structure

The crystal structure of HECs governs their mechanical, thermal, and functional properties. This section categorizes HECs into five primary structural families.

2.2.1. Rock-Salt Structure

The rock-salt (NaCl-type) structure is prevalent in HECCs and HEOs and is characterized by a face-centered cubic (FCC) arrangement of cations and anions. Key features include cation disorder, the random occupancy of metal sites in (Ti, Zr, Hf, Nb, Ta)C enhances lattice distortion, leading to improved hardness and fracture toughness [9]. And anion sublattice stability, in oxides like (Mg, Co, Ni, Cu, Zn)O, oxygen vacancies stabilize the structure under reducing conditions, enabling applications in solid-state electrolytes [7].

2.2.2. Perovskite Structure

High-entropy perovskites (ABO_3) exhibit distorted cubic or orthorhombic structures due to cation size mismatch. This includes A-site entropy and B-site entropy. (Ba, Sr, Ca, Pb, La)TiO_3 shows relaxor ferroelectric behavior with high dielectric permittivity (>5,000), suitable for capacitors. (La, Pr, Nd, Sm, Gd)CoO_3 demonstrates enhanced oxygen evolution reaction (OER) activity due to synergistic electronic effects [7].

2.2.3. Hexagonal Structure

Hexagonal HECs, such as HEBs and HESs, are dominated by close-packed layers with anisotropic properties. (Ti, Zr, Hf, Nb, Ta)B_2 is AlB_2-type boride that exhibits anisotropic thermal expansion, requiring texture control during sintering. (Mo, Nb, Ta, Ti, W)Si_2 with $MoSi_2$-type silicide shows preferential grain growth along the c-axis, influencing oxidation resistance[6].

2.2.4. Spinel Structure

Spinel-structured HECs (AB_2O_4) combine multiple cations in tetrahedral and octahedral sites. For example, (Co, Cr, Fe, Mn, Ni)$_3O_4$ exhibits mixed ionic-electronic conductivity for battery cathodes

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and gas sensors. Non-stoichiometric compositions like (Mg, Al, Ga, Fe, Zn)Al_2O_4 enable tailored bandgap engineering for photocatalytic applications [6].

2.2.5. Fluorite and Defective Fluorite Structures

Fluorite-type HECs (e.g., AO_2) and their derivatives are notable for oxygen ion mobility. There are entropy-stabilized fluorites, $(Y, Yb, Er, Lu)_2(Zr, Hf)_2O_7$ that feature a disordered anion sublattice, reducing thermal conductivity to <2 W/m·K for thermal barrier coatings [6]. Also, pyrochlore derivatives like $(Gd, Sm, Nd, Y, La)_2Zr_2O_7$ exhibit radiation tolerance, critical for nuclear reactor liners.

3. Key Properties of High Entropy Ceramics

High entropy ceramics (HECs) exhibit a unique combination of properties due to their compositional complexity, entropy stabilization effects, and structural diversity. These properties make them promising candidates for applications in extreme environments, energy storage, electronics, and catalysis. This section systematically discusses their mechanical, thermal, electrical, and chemical properties, as well as emerging functional behaviors, supported by experimental and computational studies.

3.1. Mechanical Properties

The mechanical robustness of HECs is a critical factor for their use in structural applications under high stress or temperature. High-entropy carbides (HECs), such as (Hf, Ta, Zr, Nb, Ti)C, demonstrate exceptional hardness (20–30 GPa) and fracture toughness (4–6 MPa·$m^{1/2}$), attributed to solid solution strengthening, lattice distortion, and grain boundary effects [11]. Machine learning potentials (MLPs), such as neuroevolution potentials (NEPs), have enabled large-scale molecular dynamics (MD) simulations to predict stress-strain responses and dislocation dynamics in HECs. For example, NEP-based MD simulations for (Ti, V, Cr, Mo, W)C systems revealed that increased compositional complexity enhances resistance to shear deformation and crack propagation, aligning with experimental nanoindentation results [12]. Spinel-structured high-entropy oxides (HEOs), like $(Co, Al, Fe, Ni, Ti)_3O_4$, exhibit compressive strengths exceeding 2 GPa due to their cubic close-packed oxygen sublattice and entropy-stabilized cation disorder. These materials also show improved wear resistance compared to conventional oxides, making them suitable for tribological coatings [13].

3.2. Thermal Properties

Thermal stability and conductivity are pivotal for HECs in thermal barrier coatings (TBCs) and high-temperature electronics. Entropy-stabilized oxides, such as (Mg, Co, Ni, Cu, Zn)O, retain phase stability up to 1,400°C, with thermal expansion coefficients (8–10 $\times 10^{-6}$ K^{-1}) matching those of substrates like nickel-based superalloys. Machine learning-aided studies on high-entropy carbides highlight ultralow thermal conductivity (2–3 W/m·K) due to phonon scattering from mass contrast and lattice distortion, which is critical for minimizing heat transfer in TBCs [11].

In contrast, graphitic carbon-containing HECs, such as Ti-Mg-Al-Zr-O composites, achieve enhanced thermal conductivity (15–20 W/m·K) via in situ graphitization during sintering, balancing insulation and heat dissipation needs [14].

3.3. Electrical and Dielectric Properties

The electrical behavior of HECs spans insulating, semiconducting, and metallic regimes, depending on composition and defect engineering. Spinel HEOs like $(Co, Al, Fe, Ni, Ti)_3O_4$ exhibit tunable

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resistivity (10^3–10^6 Ω·cm) and resistive switching behavior, where leakage current density decreases from 4.59×10^{-10} to 3.43×10^{-10} mhos/cm as sintering temperature increases. This property is leveraged in non-volatile memory devices [13].

Electroconductive HECs, such as (Ti, Mg, Al, Zr)O_2 composites, achieve metallic conductivity (10^2–10^3 S/cm) through oxygen vacancy generation and homogeneous graphitization. These materials also demonstrate a Hall carrier density of ~10^{21} cm^{-3}, comparable to doped semiconductors, enabling applications in electro-discharge machining and photothermal systems[14]. Dielectric properties are equally notable. For example, relaxor ferroelectric HECs like (Li, Ca, Sr, Ba, La)TiO_3 achieve colossal dielectric constants (>10^4) at low frequencies, driven by polar nanoregions and compositional heterogeneity[13].

3.4. Chemical Stability and Corrosion Resistance

The high-entropy effect confers exceptional resistance to oxidation and corrosion. For instance, (Cr, Mn, Fe, Co, Ni)$_3O_4$ spinel oxides maintain structural integrity in 1 M H_2SO_4 at 80°C for 100 hours, with mass loss <0.5%, outperforming conventional stainless steels[13]. Similarly, HECs like (Hf, Ta, Nb, Zr)C show negligible oxidation weight gain (<1%) at 1,200°C in air due to dense oxide scale formation[11].

3.5. Functional Properties

Based on its unique properties, HECs are applied to various situations. HEOs, in particular, are emerging as electrodes for supercapacitors and batteries. Inverse spinel (Cu, Ni, Zn, Al, Fe)$_3O_4$ exhibits a specific capacitance of 450 F/g at 1 A/g, attributed to multi-cation redox activity and high surface area[13]. High-entropy layered oxides, such as (Li, Ni, Co, Mn, Al)O_2, deliver lithium-ion diffusion coefficients ~10^{-10} cm²/s, enabling stable cycling at high voltages[13].

HEOs like (Co, Fe, Mn, Ni, Cu)$_3O_4$ demonstrate superior catalytic activity for oxygen evolution reactions (overpotential: 280 mV at 10 mA/cm²) due to their tailored d-band centers and oxygen vacancy clusters[13]. Photothermal HECs, such as Ti-Mg-Al-Zr-O composites, achieve 92% solar-to-thermal efficiency under 1 kW/m² irradiation, driven by broad-spectrum light absorption and localized surface plasmon resonance [14].

Bioinert HECs like (Zr, Ti, Nb, Ta, Mo)O_2 show excellent cytocompatibility (cell viability >95% after 72 hours) and antibacterial properties (>99% reduction in *E. coli* colonies), positioning them for orthopedic implants and antimicrobial coatings.

4. Emerging Applications of High-Entropy Ceramics

The unique properties of high-entropy ceramics (HECs), including exceptional thermal stability, mechanical robustness, and tunable electronic structures, have positioned them as transformative materials across diverse industrial and technological domains. This section explores their most promising applications, supported by experimental and theoretical advancements while addressing challenges and opportunities for future development.

4.1. Thermal Barrier Coatings (TBCs) for High-Temperature Systems

HECs, particularly high-entropy oxides (HEOs) and carbides (HECCs) are revolutionizing thermal barrier coatings (TBCs) used in gas turbines, aerospace engines, and nuclear reactors. Traditional TBCs, such as yttria-stabilized zirconia (YSZ), face limitations in phase stability and sintering resistance above 1200°C. HEOs like (Gd, Y, Sm, La, Nd)$_2Zr_2O_7$ exhibit superior thermophysical properties due to their entropy-driven phase stabilization, sluggish diffusion kinetics, and severe

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lattice distortion. These characteristics reduce thermal conductivity (≤1.5 W/m·K) and enhance sintering resistance, extending TBC lifetimes under cyclic thermal loads[15]. For instance, entropy-stabilized fluorite oxides, such as $(Hf_{0.2}Zr_{0.2}Ce_{0.2}Sn_{0.2}Ti_{0.2})O_2$, demonstrate negligible phase decomposition even after 1000 hours at 1500°C, outperforming conventional YSZ[15].

HECCs, including (Ti, Zr, Hf, Nb, Ta)C, are also emerging as TBC candidates due to their ultra-high melting points (>3500°C) and oxidation resistance. Their multi-principal cation sublattice mitigates crack propagation under thermal stress, a critical advantage for next-generation hypersonic vehicle coatings [16].

4.2. Energy Storage and Conversion Systems

High-entropy ceramics are redefining energy storage technologies, particularly in capacitors and battery electrodes. For example, high-entropy relaxor ferroelectric ceramics (e.g., $Sr_{0.2}Ba_{0.2}Pb_{0.2}La_{0.2}Na_{0.2}NbO_3$) exhibit ultrahigh energy storage densities (>12 J/cm^3) and efficiency (>90%) due to their nanoscale compositional heterogeneity, which disrupts long-range polar order and enhances breakdown strength (Fig. 3)[17]. This makes them ideal for pulsed power systems and electric vehicle inverters.

In lithium-ion batteries, HEOs like $(CrMnFeCoNi)_3O_4$ stabilize cycling performance by suppressing cation migration and oxygen loss at high voltages. Their configurational entropy buffers structural strain during lithiation/delithiation, achieving >90% capacity retention after 500 cycles. Additionally, high-entropy sulfides (e.g., $(Mo, W, Re, Ru, Os)S_2$) are being explored as catalysts for hydrogen evolution reactions (HER), leveraging their tunable d-band centers and corrosion resistance[18].

4.3. Wear- and Corrosion-Resistant Coatings

Thermal spraying techniques, such as plasma spraying and cold spraying, are now employed to deposit HEC coatings for extreme wear and corrosion environments. High-entropy carbide coatings (e.g., (Ti, V, Nb, Mo, W)C) synthesized via supersonic flame spraying achieve hardness values exceeding 30 GPa, surpassing binary carbides like TiC or WC. Their multi-elemental matrix reduces abrasive wear rates by 40–60% in mining machinery components [19]. In marine environments, HEO coatings like $(Al, Cr, Fe, Co, Ni)_3O_4$ demonstrate exceptional pitting resistance due to their passive oxide layers, which self-heal under chloride exposure. Electrochemical tests reveal corrosion currents <10^{-8} A/cm^2, comparable to Hastelloy alloys[15].

4.4. Structural Components in Extreme Environments

HECs are critical for applications demanding simultaneous mechanical strength and thermal resilience. For instance, HECCs such as (Ti, Zr, Hf, Ta)C are being integrated into nuclear reactor cladding due to their low neutron absorption cross-sections and radiation tolerance. Their high entropy impedes defect clustering under irradiation, reducing swelling by >50% compared to SiC[16]. In aerospace, HEOs like (Mg, Co, Ni, Cu, Zn)O are used in hypersonic vehicle leading edges, where their high entropy stabilizes the cubic phase under aerodynamic heating (>2000°C). Their fracture toughness (≥4 MPa·m$^{1/2}$) is attributed to crack deflection at nanoscale compositional fluctuations [15].

4.5. Catalytic and Functional Applications

The "cocktail effect" in HECs enables unprecedented catalytic activity. For example, high-entropy perovskite oxides (e.g., $(La, Pr, Nd, Sm, Eu)MnO_3$) exhibit 20-fold higher oxygen evolution reaction (OER) activity than single-component perovskites due to optimized, e.g., orbital filling and lattice

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strain. Similarly, high-entropy MXenes (e.g., $(Ti, V, Nb, Mo, Ta)_2C$) show promise in CO_2 reduction, with Faradaic efficiencies >80% for methane production[18]. Functional applications include thermoelectrics, where high-entropy silicides (e.g., $(Mg, Ca, Sr, Ba, Eu)Si_2$) achieve ZT values >1.2 at 800°C via phonon scattering at distorted lattices [18].

4.6. Challenges and Future Perspectives

Despite this progress, challenges remain. First, most HECs are lab-scale products. Spraying techniques require optimization to control stoichiometry in non-equiatomic systems [19]. Moreover, the long-term stability of metastable HECs under operational conditions (e.g., redox cycles) needs further validation [15]. Additionally, machine learning (ML) models, as demonstrated for HECCs [16], must expand to multi-anion systems (e.g., oxynitrides).

These limitations can be overcome through future research, which includes multi-functional HECs and combining entropy stabilization with topological defects for synergistic properties. Bioactive HECs, exploring high-entropy phosphates for bone implants and quantum materials, designing high-entropy superconductors via entropy-mediated electron-phonon coupling.

5. Conclusion

High-entropy ceramics (HECs) represent a groundbreaking paradigm in materials science, characterized by their compositional complexity, entropy-stabilized structures, and exceptional multifunctional properties. Over the past two decades, significant progress has been made in classifying HECs into diverse systems (e.g., oxides, carbides, nitrides) and understanding their structure-property relationships. These materials exhibit extraordinary mechanical robustness and electrochemical performance. Their unique properties stem from entropy-driven effects, including lattice distortion, diffusion retardation, and the "cocktail effect," enabling applications in extreme environments). However, critical challenges impede their widespread adoption. Scalable synthesis remains a bottleneck, as lab-scale methods like spark plasma sintering struggle with stoichiometric control during industrial-scale production. Metastable phases risk degradation under operational stresses, while the reliance on rare elements (e.g., Hf, Ta) and energy-intensive processing escalate costs. Additionally, gaps in standardized definitions, long-term stability data, and mechanistic understanding of anion sublattice disorder or B-site entropy effects hinder systematic optimization.

Future advancements hinge on interdisciplinary innovation. Emerging synthesis techniques, such as flash sintering and laser additive manufacturing, promise energy-efficient fabrication of metastable HECs. Ultimately, HECs transcend traditional material limitations by harmonizing conflicting properties—strength with low thermal conductivity and corrosion resistance with catalytic activity. Addressing challenges in scalability, cost, and standardization will require global collaboration among academia, industry, and policymakers. With continued innovation in synthesis, characterization, and computational tools, HECs are positioned to redefine performance benchmarks in energy, aerospace, and beyond, ushering in a new era of entropy-dominated materials engineering.

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