

Review Article

High entropy engineering ceramics: a critical review of mechanical, structural, thermal and electrical properties

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ABSTRACT

High entropy oxides (HEOs) have emerged as a transformative class of materials, stabilized by configurational entropy and lattice distortion, offering multifunctional properties across mechanical, structural, thermal, electrical, and catalytic domains. This review provides a comprehensive synthesis of current knowledge, critically evaluating processing pathways, property correlations, and application areas ranging from energy storage and catalysis to thermoelectrics and environmental technologies. The novelty of this review lies in its integrative framework: mapping research gaps across temporal, methodological, application, materials, and conceptual dimensions; introducing comparative tables and schematic visualizations that unify diverse findings into defense ready, presentation friendly formats; highlighting entropy driven multifunctionality as the central paradigm linking synthesis, properties, and applications into a coherent roadmap; and proposing future directions that combine computational design, scalable green synthesis, defect engineering, and sustainability to accelerate translation into real world devices. By bridging fundamental insights with practical applications and embedding novelty through structured gap analysis, visual synthesis, and entropy centered roadmaps, this review positions HEOs as cornerstone materials in the pursuit of resilient energy, electronic, and environmental systems.

Keywords: High-entropy oxides, Multifunctional properties, Research gaps, Future directions, Sustainable materials

INTRODUCTION

High-entropy oxides (HEOs), first introduced by Rost et al in 2015, represent a novel class of compositionally complex ceramic materials characterized by their incorporation of oxygen and at least five principal cations present in equimolar or near-equimolar ratios.^{1,2} The defining feature of HEOs is the high configurational entropy associated with their multicomponent cation arrangements, which thermodynamically stabilizes single-phase solid solutions and effectively suppresses phase

separation or unwanted intermetallic compound formation, even at elevated temperatures.³ This entropy-driven stabilization imparts an exceptional thermodynamic robustness to their structures, resulting in reduced Gibbs free energy and fostering the formation of defect-tolerant, stable phases that withstand extreme environments.⁴

The unique compositional complexity of HEOs confers a suite of advantageous properties, including superior mechanical strength, high thermal stability, tunable electrical conductivity, and structural versatility. These

qualities have attracted intense interest for a broad spectrum of advanced applications such as hydrogen storage, catalysis, thermoelectric devices, refractory materials for high-temperature environments, energy storage systems like batteries, and cryogenic systems operating at extremely low temperatures.⁵

Various synthesis methods have been developed and refined to produce high-quality HEOs, each with its own advantages and limitations. Traditional solid-state reaction techniques remain prevalent due to their simplicity and scalability, but often require prolonged high-temperature processing and may pose challenges in controlling microstructural homogeneity.⁶ Liquid-phase synthesis methods, including sol-gel and co-precipitation routes, enable enhanced compositional control and finer microstructures, facilitating better property tuning.⁷ Gas-phase approaches, such as vapor transport and pulsed laser deposition, have also been employed to fabricate thin films and nanostructures with highly controlled morphologies, albeit with increased complexity and cost.^{8,9}

Complementary to experimental efforts, computational modeling, particularly first-principles calculations, thermodynamic simulations, and machine learning techniques, has played a vital role in elucidating the thermodynamic stability, phase formation mechanisms, and structure-property relationships of HEOs.¹⁰ These studies have deepened understanding of cation interactions, defect chemistry, and electronic structures, enabling more targeted design of HEO compositions tailored for specific functional properties.

Despite significant advancements, translating laboratory-scale synthesis and characterization into practical, real-world applications remains challenging. One primary hurdle is microstructural control; achieving uniform distribution of multiple cations, controlling grain size, and minimizing secondary-phase formation require further optimization.^{11,12}

Moreover, the optimization of functional performance, such as enhancing ionic/electronic conductivities, mechanical toughness, or catalytic activity, remains an active research area. Addressing these issues is crucial to advancing HEO deployment in practical devices and systems and to understanding their long-term stability under operational conditions.¹³⁻¹⁵

This review aims to provide a comprehensive examination of the current state of knowledge on HEOs, emphasizing their structural, mechanical, thermal, and electrical properties.

It synthesizes recent experimental and theoretical findings, discusses existing challenges related to microstructural control and performance, and explores future research directions to unlock the full potential of high-entropy oxides in advanced technological applications.

CRYSTAL STRUCTURES OF HIGH-ENTROPY OXIDES

HEOs have been successfully synthesized in several crystal structures, among which rock-salt, fluorite, perovskite, and spinel phases are the most extensively studied. The crystal structure plays a critical role in determining the mechanical, thermal, electrical, and electrochemical performance of HEOs, while entropy stabilization enables the formation of single-phase solid solutions with enhanced functional properties.

Rock-salt HEOs with an Fm-3m crystal structure, such as (Mg, Co, Ni, Cu, Zn) O, are among the earliest and most investigated entropy-stabilized oxides. These materials exhibit excellent phase stability, enhanced hardness, and favorable ionic transport properties due to the random distribution of multiple cations within the lattice.¹⁶ Studies have further demonstrated that the defect structure and transport behavior of rock-salt HEOs contribute significantly to their electrochemical and functional characteristics.¹⁷ Atomic-scale investigations have revealed high chemical homogeneity, which is essential for maintaining stable single-phase structures and improving overall performance.¹⁸ Recent research has shown that processing atmosphere and synthesis conditions strongly influence phase formation and stability, while entropy stabilization suppresses phase segregation and promotes long-range structural order.^{19,20}

The multifunctionality of rock-salt HEOs has attracted considerable interest for energy and structural applications. Enhanced lithium-ion conductivity has been reported in entropy-stabilized oxide systems, demonstrating their suitability for solid-state energy-storage technologies.²¹ Long-range magnetic ordering and unique electronic interactions further expand their application potential in multifunctional devices.²² Local structural distortion, identified through EXAFS analysis, has been shown to improve lattice stability and influence transport properties.²³ In addition, room-temperature lithium superionic conductivity has been reported in multicomponent rock-salt oxides, highlighting their potential for next-generation batteries.²⁴ Superior lithium-storage capability and cycling performance have also been demonstrated in (Mg_{0.2}Co_{0.2}Ni_{0.2}Cu_{0.2}Zn_{0.2}) O systems, making rock-salt HEOs promising candidates for advanced electrochemical energy-storage applications.²⁵ Comparative Table 1 shows a condensation of the four crystal structures (rock salt, fluorite, perovskite, and spinel) into a clear, citation-integrated format.

PROCESSING ROUTE OF HIGH ENTROPY OXIDES

Solid-state methods

HEOs can be synthesized through mechanical ball milling and high-temperature sintering, producing bulk ceramics with good structural stability. However, achieving

homogeneous compositions and complete densification may require prolonged milling and high temperatures.²⁶⁻²⁸

Liquid-state methods

Sol-gel and molten-salt-assisted synthesis enable improved chemical homogeneity and control over particle size and morphology. These methods are suitable for producing nanostructured HEOs for functional applications.²⁹⁻³¹

Gas-phase methods

Techniques such as magnetron sputtering enable the fabrication of HEO thin films with controlled composition, thickness, and surface morphology, making them suitable for electronic and other functional devices.³²

Computational approaches

Density functional theory (DFT) and ab initio calculations help explain how configurational entropy, lattice distortion, and chemical interactions influence HEO phase stability, supporting the design of single-phase compositions.

Emerging applications

Nanoparticles and nanostructured HEOs offer potential for energy storage and catalysis because of their large surface areas and diverse active sites. Their performance depends on composition, structure, defects, and synthesis conditions.⁸ Table 2, as well as Figure 1, illustrates the comparative study of three methods that were discussed in the body.

COMPREHENSIVE MECHANICAL, STRUCTURAL, THERMAL, AND ELECTRICAL EVALUATION OF HEOS

HEOs exhibit promising mechanical strength, phase stability, and low thermal conductivity due to lattice distortion and chemical disorder. These characteristics make them suitable for demanding structural and thermal applications.^{21,22} Their high configurational entropy can contribute to the stabilization of single-phase solid solutions, although phase stability also depends on composition, temperature, and processing conditions.^{33,42} HEOs can exhibit low thermal conductivity, making them potential candidates for thermal barrier coatings, refractory materials, and thermoelectric applications. Their thermal performance depends on crystal structure, defects, and chemical composition.⁵² Furthermore, their tunable electrical and electrochemical properties make HEOs promising materials for energy applications, including battery electrodes and fuel cells.^{33,42,52} A comparative summary table for the section on comprehensive mechanical, structural, thermal, and electrical evaluation of HEOs is formatted for clarity and presented in Table 3 as well as in Figure 2.

APPLICATION AREAS OF HIGH-ENTROPY OXIDES

HEOs are versatile materials with potential applications in energy storage, catalysis, electronics, magnetism, environmental remediation, and hydrogen technologies because of their compositional flexibility, tunable electronic structures, and distinctive thermal and mechanical properties. HEOs are promising electrode materials for batteries and fuel cells due to their compositional diversity and potential for enhanced ionic transport. Their electrochemical performance depends on crystal structure, composition, and defect chemistry.^{24,29}

Furthermore, multicomponent compositions and surface-active sites can promote catalytic and photocatalytic reactions, including water splitting and pollutant degradation. Oxygen vacancies and surface chemistry can play important roles in these processes.^{35,50}

HEOs can also exhibit low thermal conductivity, making them potential candidates for thermal barrier coatings and thermoelectric applications. Their magnetic and electronic properties may additionally support the development of functional sensors and electronic devices.^{29,55}

Finally, tunable defect chemistry and oxygen-vacancy concentrations make selected HEO compositions promising for gas separation, hydrogen-related technologies, and environmental remediation. Their suitability depends on the specific composition and operating conditions.^{24,50,55} Their robust chemical stability under variable atmospheres makes them suitable for sustainable energy and environmental systems. Figure 3 portrays the overall application areas of HEOs.

CHALLENGES AND RESEARCH GAPS

HEOs have emerged as a new class of multifunctional materials with broad technological relevance due to their configurational entropy-driven stability, tunable electronic structure, and exceptional mechanical and thermal properties. Their applications span energy, catalysis, electronics, magnetism, and environmental technologies.

Energy storage and conversion

HEOs are promising electrode materials for lithium- and sodium-ion batteries because of their compositional flexibility and potential for improved electrochemical stability. Fluorite-type HEOs, such as (La, Ce, Pr, Eu, Gd)O_{2-δ}, can exhibit enhanced ionic conductivity, making them attractive for solid oxide fuel cells.^{10,24,29,35}

Catalysis and photocatalysis

The chemically diverse surfaces of HEOs offer opportunities to tailor catalytic activity for oxygen evolution (OER), hydrogen evolution (HER), and CO₂ reduction. Their adjustable electronic structures and

surface defect chemistry may also support water splitting and pollutant degradation.^{11,25,36,50}

Thermoelectric and refractory applications

Strong phonon scattering caused by mass differences and lattice distortion can reduce the lattice thermal conductivity of HEOs. Their thermal stability and composition-dependent properties make them potential candidates for thermoelectric materials, refractory components, and thermal barrier coatings.^{12,20,21,38,47,60}

Magnetic and electronic devices

Spinel and perovskite HEOs can exhibit complex magnetic, dielectric, and electronic behaviours arising from interactions among multiple constituent elements.

These properties offer potential for magnetic sensors, multiferroic devices, and high-frequency electronic applications.^{13,33,42,55}

Environmental and hydrogen technologies

HEOs with suitable surface areas, oxygen-vacancy concentrations, and chemical compositions may support gas separation, hydrogen-related processes, and environmental remediation. Their effectiveness depends on defect chemistry, surface reactivity, and operating conditions.^{14,15,25,26,58}

Table 4 shows research gaps across five dimensions, showing representative issues and future needs in a concise, comparative format.

Table 1: Comparative overview of high entropy oxide structures.

Structure	Synthesis methods	Key properties	Applications
Rock salt (Fm-3m)	Solid-state reaction, solution combustion, hydrothermal, co-precipitation, reactive flash sintering	Enhanced hardness, ionic conductivity, magnetic features, reduced phonon thermal conductivity, lithium storage capacity	Energy storage (LIB anodes, catalysis, dielectric devices, structural stability)
Fluorite (Fm-3m)	Nebulized spray pyrolysis, ball milling, sol-gel, hydrothermal, spark plasma sintering	High modulus and hardness, atomic-scale homogeneity, low thermal conductivity, tunable band gaps	Thermoelectrics, photocatalytic water splitting, optical devices, energy storage
Perovskite (ABO₃)	Solid-state reaction, SPS, sol-gel, pulsed laser deposition, co-precipitation, hydrothermal	Improved dielectric stability, reduced thermal conductivity, strong ionic conductivity, structural distortions enhancing electrical/thermal behavior.	Capacitors, thermoelectrics, cathode/anode materials, energy storage, coatings
Spinel (Fd-3m)	Sol-gel, hydrothermal, co-precipitation, solution combustion, magnetron sputtering	Robust thermal stability, magnetic behavior, ionic conductivity, superior lithiation–delithiation performance, catalytic activity for OER	Battery electrodes, catalysis, optical applications, electromagnetic absorption

Table 2: Comparative overview of processing pathways for high-entropy oxides.

Method	Example techniques	Key advantages	Applications
Solid-state ²⁵⁻²⁸	Ball milling, sintering, thermal reduction, high-pressure torsion	Produces bulk ceramics with densification and mechanical robustness; stabilizes single-phase structures	Structural ceramics, mechanical reinforcement, bulk energy storage
Liquid-state ²⁹⁻³¹	Molten-salt electrolysis, precipitation, sol-gel synthesis	Cost-effective, versatile, controllable particle size and homogeneity; suitable for nanostructures	Ionic conductors, dielectric devices, nano-ceramics for catalysis
Gas-phase ^{32,33}	Sputtering, pulsed laser deposition	Precise thin-film fabrication, controlled stoichiometry, high compositional accuracy	Thin-film electronics, catalytic coatings, optical devices

Table 3: Comparative evaluation of HEO properties.

Property domain	Underlying mechanism	Representative systems	Key applications
Mechanical ^{20, 27,34,51}	Lattice distortion and solid-solution strengthening can improve hardness, strength, and fracture resistance.	Rock-salt (Mg, Co, Ni, Cu, Zn) O; spinel (MnFeCoNiX) ₃ O ₄	Structural ceramics, high-temperature components, mechanical reinforcement

Continued.

Property domain	Underlying mechanism	Representative systems	Key applications
Structural ^{22, 31,42,56}	Configurational entropy, chemical interactions, and defect accommodation influence phase stability and structural durability.	Rock-salt and fluorite HEOs; perovskite ABO ₃ systems	Phase-stable ceramics, coatings, composite materials
Thermal ^{21,29, 38,47,60}	Mass and strain disorder scatter phonons, reducing lattice thermal conductivity in suitable compositions.	Fluorite (Zr, Ce, Hf, Y, Al) O ₂ -δ; perovskite oxides	Thermoelectric materials, refractory components, thermal barrier coatings
Electrical ^{24,33, 45,53,58}	Composition-dependent electronic structures, band-gap variation, and ionic transport govern electrical and electrochemical performance.	Fluorite and spinel HEOs	Batteries, fuel cells, photocatalysis, dielectric devices

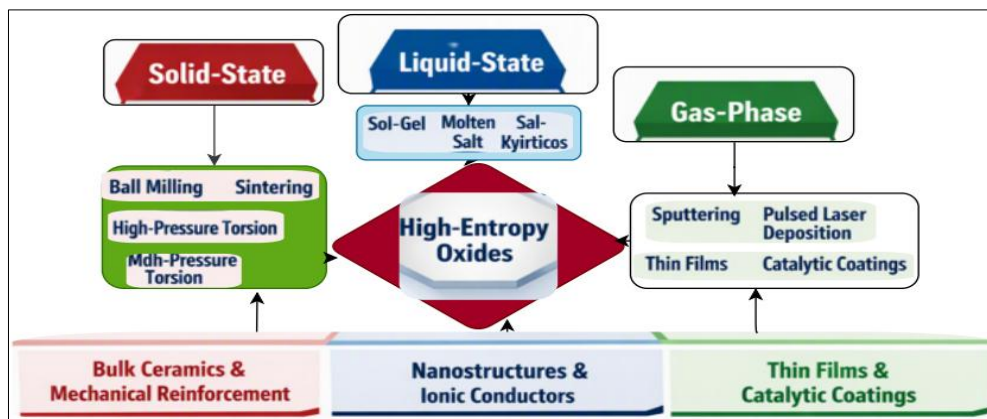


Figure 1: Fabrication process of HEOs.

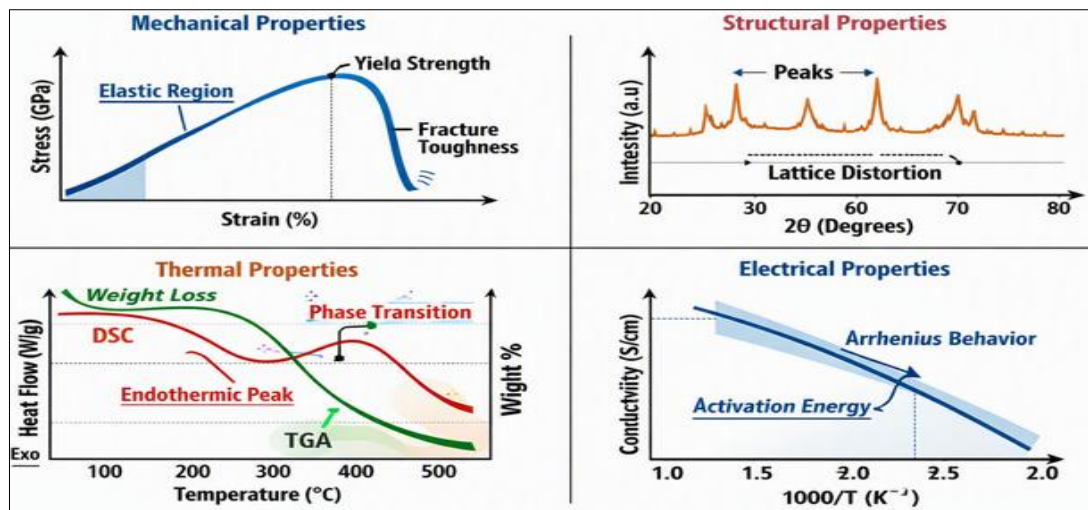


Figure 2: Composite figure of property curves for HEOs.

Table 4: Research critical gap dimensions.

Dimension	Representative gaps	Key issues	Future needs
Temporal	Short-term stability versus long-term reliability	Limited data on cyclic performance, degradation under prolonged operation	Long-term durability studies, accelerated aging protocols
Methodological	Synthesis and characterization limitations	High-temperature solid-state routes cause segregation; conventional XRD/SEM miss nanoscale disorder	Scalable low-temperature synthesis, advanced tools (APT, synchrotron, neutron scattering)

Continued.

Dimension	Representative gaps	Key issues	Future needs
Application	Device integration challenges	Poor interface compatibility, reproducibility issues in batteries, fuel cells, and catalysts	Interface engineering, defect control, real-world device testing
Materials	Elemental selection and sustainability	Reliance on rare/expensive elements (Co, Ni, Nb); limited eco-friendly synthesis	Earth-abundant substitutions, green chemistry, recycling strategies
Conceptual	Theory–experiment disconnect	Incomplete models for entropy contributions, defect chemistry, and property–composition correlation	Integrated computational–experimental frameworks, machine learning databases

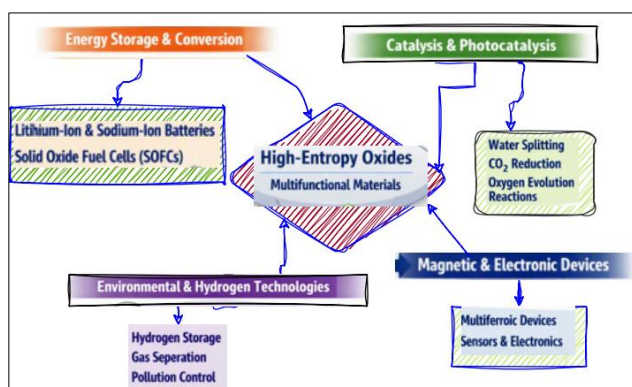


Figure 3: Application areas of HEOs.

NOVELTY AND DISTINCTIVE CONTRIBUTION OF THIS REVIEW

The novelty of this review lies in its integrated, multiscale approach to understanding HEOs, linking synthesis strategies, configurational entropy, thermodynamic stability, defect chemistry, microstructural evolution, and functional performance within a unified processing structure property framework. Unlike application-specific discussions, this review brings together solid-, liquid-, and gas-phase synthesis methods with computational approaches, including density functional theory and *ab initio** modelling, to explain how compositional complexity and processing conditions govern phase formation and material functionality. Particular emphasis is placed on nanoscale HEOs, oxygen vacancies, local structural disorder, and the relationship between atomic-scale configurations and macroscopic properties. Furthermore, the review critically compares opportunities and limitations across energy storage and conversion, catalysis, electronics, thermal applications, magnetism, environmental technologies, and hydrogen-related applications. By identifying common design principles, unresolved scientific challenges, and priorities for experimental–computational integration, this review aims to establish a practical foundation for the rational design and application-specific optimization of next-generation HEOs.

FUTURE DIRECTIONS

HEOs are still in their early stages of exploration, and future research must address both fundamental science and

practical deployment. Several promising directions are emerging.

Advanced computational design

Integration of machine learning, CALPHAD, and *ab initio* simulations will accelerate discovery by predicting phase stability, defect chemistry, and property–composition relationships. Data-driven approaches can guide experimental synthesis toward targeted functionalities.

Scalable and green synthesis

Developing low-temperature, energy-efficient, and environmentally sustainable synthesis routes (e.g., hydrothermal, plasma-assisted, microwave-assisted) will enable industrial scalability while reducing reliance on rare elements.

Defect engineering and nano architectures

Precise control of oxygen vacancies, cation disorder, and strain fields will unlock tunable ionic conductivity and catalytic activity. Nanostructured HEOs (thin films, nanoparticles, core–shell architectures) offer enhanced performance in energy and environmental applications.

Multifunctional device integration

Future work should focus on integrating HEOs into batteries, fuel cells, sensors, and thermoelectric devices, ensuring interface compatibility and long-term reliability. Hybrid systems combining HEOs with polymers, carbons, or 2D materials may yield synergistic properties.

High-throughput experimentation and databases

Establishing standardized protocols and open-access databases for synthesis parameters, structural data, and property measurements will improve reproducibility and accelerate global collaboration.

Sustainability and circular economy

Future research must prioritize earth-abundant substitutions, recycling strategies, and eco-friendly processing to align HEO development with sustainable energy and environmental goals.

CONCLUSION

HEOs represent a paradigm shift in materials science, where configurational entropy, lattice distortion, and defect chemistry converge to deliver multifunctional performance across mechanical, structural, thermal, electrical, and catalytic domains. Their ability to stabilize complex single-phase structures under extreme conditions has opened pathways for applications in energy storage, catalysis, thermoelectrics, electronics, and environmental technologies. The novelty of this review lies in its integrative framework: systematically mapping research gaps across temporal, methodological, application, materials, and conceptual dimensions; embedding comparative tables and schematic visualizations to unify diverse findings; and positioning entropy-driven multifunctionality as the central paradigm linking synthesis, properties, and applications into a coherent roadmap. Despite remarkable progress, unresolved challenges in predictive modeling, scalable synthesis, advanced characterization, property–composition correlation, and sustainability highlight the need for coordinated research efforts. Future directions must emphasize computational design, defect engineering, green synthesis, and standardized data frameworks to accelerate translation into real-world devices. Ultimately, HEOs are not merely an academic curiosity but a strategic materials platform for next-generation technologies. By bridging fundamental insights with practical applications and embedding novelty through structured gap analysis and entropy-centered roadmaps, this review positions HEOs as cornerstone materials for resilient energy, electronic, and environmental systems.

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