

High Entropy Oxide Ceramic by Pressure Assisted Sintering of Multi-Component Rare-Earth Oxides

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High-entropy oxides (HEOs) with fluorite structure exhibit tunable bandgaps, high transparency, and multi-wavelength emissions, making them attractive for optoelectronic applications. Their single Wyckoff cation site ensures stable, homogeneous cation distribution, while high-temperature stability arising from random cation arrangement drives their rapid development. Unlike conventional oxides, the configurational entropy in HEOs provides additional stabilization, enabling property tuning beyond the limits of traditional material design. This work addresses challenges in synthesizing fluorite-structured HEOs with controlled properties by investigating the effects of cation substitution and sintering methods. HEOs with the composition $\text{CeO}_{2-\delta}(\text{RE}, \text{La}, \text{Sm}, \text{Y})_2\text{O}_3$ [RE = Pr, Dy, Gd] were synthesized via reactive sintering, revealing that replacing Pr with Gd or Dy increased the bandgap (from 1.8 eV to 3.13–3.18 eV), and enhanced luminescent properties. Translucent, highly dense HEOs $\text{CeO}_{2-\delta}(\text{RE}, \text{La}, \text{Sm}, \text{Y})_2\text{O}_3$ [RE = Dy, Gd] with single-phase bixbyite structure were achieved at 1600°C with relative density >99% and translucence in the visible range, exhibiting strong multi-wavelength emissions and near-cold white light under UV excitation. Hot isostatic pressing led to phase separation and bandgap reduction. Spark plasma sintering (SPS) of $\text{CeO}_{2-\delta}(\text{Dy}, \text{La}, \text{Sm}, \text{Y})_2\text{O}_3$ HEOs induced reversible cubic-to-monoclinic phase transitions, influenced by cerium oxidation state and oxygen vacancies, further affecting bandgap energy. Overall, this study not only demonstrates how composition and processing routes dictate optical performance, but also establishes design strategies for achieving translucency, near-theoretical density, tunable bandgaps, and white-light emission. These advancements position fluorite-structured HEOs as promising candidates for next-generation phosphors in optoelectronic and lighting technologies.