Tailoring Thermoelectric Performance of Stabilized High-Entropy Perovskite Ceramics through Nb^{5+} Substitution

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ABSTRACT

Novel Stablized high-entropy perovskite oxides, $Sr_{0.25}Ba_{0.25}Ca_{0.25}La_{0.25}Ti_{1-x}Nb_xO_3$ with varying Nb^{5+} concentrations were synthesised using the conventional solid-state route and sintered using field-assisted sintering technology at a temperature range from 1200 - 1350 °C. Novel compositions with a high configuration entropy were selected based on the theoretical predictions, considering tolerance factor, polarizability, electronegativity, configuration entropy, and global instability index G*II*-value (using "Structure prediction diagnostic software" SPUDS). The XRD analysis confirmed the formation of a single-phase simple cubic perovskite structure, having a space group of Pm-3*m.* The SEM and EDX analysis confirmed the formation of dense microstructures with a relative density higher than 95 % and phase purity of the sintered samples. The FTIR analysis confirmed the formation of a single phase and metal oxides bonding in the structures. The UV-visible spectra measured in the diffuse reflectance mode indicated a gradual increase in the optical band gap value of the samples with increasing Nb^{5+} concentration. The increase in the configuration entropy of the samples had a significant impact on the thermoelectric performance of the samples. The samples possessed a high negative Seebeck coefficient (S), confirming their n-type semiconducting behaviour. In addition, the samples exhibited a low thermal diffusivity < 1 mm^2/s, a low thermal conductivity K < 3.8 $Wm^{-1}K^{-1}$ and a moderate electrical conductivity of σ = 1.26 S/cm for $x = 0.05$. These parameter favours thermoelectric performance, leading to an increased ZT value. The present work offers a compositional design approach for perovskite oxides to reduce their intrinsic thermal conductivity and enhance their thermoelectric performance.

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