World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:19, No:04, 2025

Super-Exchange Coupling in Oxygen Rich Rare-Earth Based $Sm_2MnRuO_{6+}\delta$ Double Perovskite

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Abstract : A rare-earth-based $Sm_2MnRuO_{6+}\delta$ (SMRO) double perovskite was prepared using a high-temperature solid-state reaction. The structural, morphological, chemical, thermodynamic, and magnetic properties were measured with X-ray diffraction (XRD), energy dispersive spectroscopy (EDS), X-ray photoemission spectroscopy (XPS), and vibrating sample magnetometer (VSM), respectively. The XRD revealed a tetragonal structure belonging to the I4/mmm space group, number 139, with linear Mn−O−Ru bonds. Replacing the well-studied alkaline earth metal with a rare-earth element increased the Mn-O bond length difference between the shorter equatorial (Mn-Oab) and the axial (Mn-Oc) bonds by approximately 6.3%. The elemental composition showed an O-rich double perovskite with a Ru deficit, which encourages the formation of a Ru^{6+} (d^2) state. XPS spectra of Sm-3d, Ru-3d, and Mn-2p revealed the coexistence of a double oxidation state for each cation; Sm^{2+}, Sm^{3+}, Ru^{3+}, Ru^{6+}, Mn^{2+} , and Mn^{3+}, in varying proportions. Entropy studies showed drastic ordering of spins at low temperatures (up to 12.4 K), whilst increasing temperatures above this point resulted in a drastic increase of disorder of the spins (up to 43.26 K), beyond which a constant slope of entropy is observed. Magnetic measurements revealed two magnetic ground states at TN = 12.4 K and TC = 43.3 K ordering antiferromagnetically (AFM) and ferromagnetically (FM), respectively. Kneller fit further showed that the materials become completely paramagnetic at TB = 88.1 K, (the blocking temperature). The existence of ferromagnetic (FM) super-exchange coupling in this work originating from Mn^{3+} ($t^3_2 e^1$)−O−Ru^{3+} ($t^5_2 e^0$) and Mn^{2+} ($t^3_2 e^2$)−O−Ru^{6+} ($t^2_2 e^0$) which plays an important role in suppressing the Mn/Ru−O−Mn/Ru antiferromagnetic (AFM) interactions.

Keywords : solid-state reaction, super-exchange coupling, ferromagnetic, Kneller’s law, entropy
Conference Title : ICMME 2025 : International Conference on Metallurgical and Materials Engineering
Conference Location : Cape Town, South Africa
Conference Dates : April 10-11, 2025