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EFFECT OF Zn SUBSTITUTION ON TELLURIUM-BASED $Sr_2Ni_{1-x}Zn_xTeO_6$ DOUBLE PEROVSKITE: EXPERIMENTAL AND FIRST PRINCIPLE DFT–LDA+U CALCULATION

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ABSTRACT

Double perovskite materials such as Sr_2NiTeO_6 shows great interest in the science community since 1940s due to their excellent properties and novel features. The Sr_2NiTeO_6 could provide advantages in current applications such as in electronic devices. The substitution of Zn may enhance the dielectric properties of this material since the small in ionic size distorted the structural orientation and thus effect the dielectric properties of the materials. In this study, a series of tellurium-based double perovskite, $Sr_2Ni_{1-x}Zn_xTeO_6$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) were prepared using conventional solid-state method in order to investigate their structural, optical and dielectric properties. The Rietveld refinement of X-ray diffraction data shows that all compounds were crystallised in monoclinic symmetry with the *I*2/*m* space group. Morphological scanning electron microscopy reported that the grain sizes decreased as the dopant increased, from ~1.63 to ~1.20 μm for x = 0.0 to 1.0, respectively. The UV-vis diffuse reflectance spectroscopy conducted for all samples found that the optical band gap energy increased from 3.54 to 4.07 eV and 3.71 to 4.14 eV for x = 0.0 to 1.0, by using cut-off wavelength and Tauc plot method, respectively. The dielectric permittivity values increased for the highest Zn-doped compound (x = 0.8), being ~1000 and ~60 in the low- and high-frequency range, respectively, correspond to the highest density value. All samples exhibited low dielectric loss (tan d ≤ 0.20) in the range of 10^4–10^5 Hz frequency. Impedance measurement revealed that grain resistance decreased with enhancement in Zn content in the Sr_2NiTeO_6 crystal lattice. The DFT calculation for *I*2/*m* monoclinic Sr_2NiTeO_6 double perovskite was performed via DFT–LDA+U to study the influence of self-corrected Hubbard U on structural optimization, optical and electronic properties. The strong Coulomb repulsion between electrons was corrected using Hubbard U, which ranged from 0 to 8 eV for Ni 3d states. The calculated results with U value of 8 eV showed a good agreement with previous experimental results. From the electronic properties, the optimized electronic band gap and density of states indicated that U values were significantly influenced the hybridization of O 2p and Ni 3d states at valence and conduction bands, respectively. The optical properties showed a significant change with varying U values. The DFT–LDA+U calculations on $Sr_2Ni_{1-x}Zn_xTeO_6$ (x = 0.00, 0.25, 0.50, 0.75 and 1.00) double perovskite compound were performed with U value for Ni 3d was set to 8 eV. The calculated lattice volume structure for all Zn concentration ratios were in good agreement with the experimental lattice volume. The dielectric permittivity of rich Zn-doped (x = 0.75) showed the highest value compared to other x. The absorption coefficient for all x presented a similar plot as experimental data. For electronic properties, all x exhibit semiconductor behavior, similar reported by previous experiment work with calculated energy band gap 1.723, 1.583, 1.462, 1.410 and 0.757 eV for x = 0.00, 0.25, 0.50, 0.75 and 1.00, respectively. The electronic density of states of $Sr_2Ni_{1-x}Zn_xTeO_6$ double perovskite compound revealed that Sr, Ni, Zn and O atoms highly contributed by the electronic states at Fermi level (E = 0 eV) with high degree of hybridization between the Sr/Ni/Zn and O 2p electrons.

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