

## Effect of iron substitution by nickel on crystal structures, optical, and magnetic properties in double perovskite series $\text{Sr}_2\text{Fe}_{1-x}\text{Ni}_x\text{TeO}_6$ with $x = 0, 0.25, 0.50, 0.75,$ and $1$

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The double perovskite series with the formula  $\text{Sr}_2\text{Fe}_{1-x}\text{Ni}_x\text{TeO}_6$  ( $x = 0, 0.25, 0.50, 0.75,$  and  $1$ ) has been synthesized in polycrystalline form by a conventional solid-state reaction process by heating to 1300 K in air. Their crystal structures were probed by means of X-ray diffraction at room temperature. Rietveld analysis revealed that all the compositions crystallize in monoclinic space group  $I2/m$ . The double perovskite structures contain two alternating types of octahedra  $(\text{Fe}/\text{Ni})_{2d}\text{O}_6$  and  $(\text{Te})_{2a}\text{O}_6$ , tilted in the system  $(a^-a^-c^0)$ , as suggested by Glazer. Furthermore, the refinement has shown a complex cation distribution over the octahedra sites, where Fe, Ni, and Te atoms were found to occupy both possible two-fold sites in the structure;  $2d$   $(\frac{1}{2}, \frac{1}{2}, 0)$  and  $2a$   $(0, 0, 0)$  sites. The successful refinement of the cation distribution results in the following crystallography formulas:  $\text{Sr}_2$   $[\text{Fe}_{0.84}\text{Te}_{0.16}]2d$   $[\text{Te}_{0.87}\text{Fe}_{0.13}]2a$   $\text{O}_6$ ,  $\text{Sr}_2$   $[\text{Fe}_{0.71660}\text{Ni}_{0.18534}\text{Te}_{0.09806}]2d$   $[\text{Te}_{0.90194}\text{Fe}_{0.0334}\text{Ni}_{0.06466}]2a$   $\text{O}_6$ ,  $\text{Sr}_2$   $[\text{Fe}_{0.46612}\text{Ni}_{0.49537}\text{Te}_{0.03851}]2d$   $[\text{Te}_{0.96149}\text{Fe}_{0.03388}\text{Ni}_{0.00463}]2a$   $\text{O}_6$ ,  $\text{Sr}_2$   $[\text{Fe}_{0.24981}\text{Ni}_{0.71479}\text{Te}_{0.03540}]2d$   $[\text{Te}_{0.96460}\text{Fe}_{0.00019}\text{Ni}_{0.03521}]2a$   $\text{O}_6$  for  $x = 0, 0.25, 0.50, 0.75,$  respectively. The calculated tolerance factor suggest that the room-temperature structures of all compounds should not be cubic and that the distortion of the octahedra increases with increasing nickel. This proposition, was confirmed by Raman spectroscopy, which reveals an increase in the number of Raman modes as Fe is substituted by Ni. Variable temperature magnetic susceptibility data show obvious antiferromagnetic transitions below 40 (1) K (38 (1) K, 31(1) K, 25 (1) K, 20(1) K, and 35(1) K for the compounds  $x = 0, 0.25, 0.50, 0.75,$  and  $1,$  respectively). Interestingly, this reduction in  $T_N$  is getting strong when  $\text{Fe}^{3+}$  and  $\text{Ni}^{2+}$  ions are together in the structure, as well as when  $\text{Ni}^{2+}$  is increasing, which indicates a strong AFM interaction. This fact was explained based on the rules of Goodenough-Kanamori, which announced that the antiferromagnetic behaviour is dominant when multiple types of interactions are present in the systems [1]. This confirms that  $\text{Fe}^{3+}$  ( $3d^5$ ) ions are in HS state with  $t_{2g}$  and  $e_g$  half-filled, and  $\text{Ni}^{2+}$  ( $3d^8$ ) ions are in HS state with  $t_{2g}$  saturated of electrons and  $e_g$  half-filled. A divergence between FC and ZFC curves for all compositions has been observed, confirming the presence of spin-glass behaviour. The optical characteristics of the series were determined by diffuse reflectance UV-Vis spectroscopy. The data were analysis using the Tauc method and by Derivation of Absorption Spectra Fitting (DASF) techniques to derive optical band gaps [2].

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[2]. Souris D, Tahan Z E, (2015). *Appl. Phys.* **B 119**, 273–279.

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