

Structural and dielectric properties of yttrium substituted nickel ferrites



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ABSTRACT

The influence of Y^{3+} ions on structural and dielectric properties of nickel ferrites ($NiFe_{2-x}Y_xO_4$, where $0 \leq x \leq 0.3$) has been studied. The as-synthesized samples, prepared by the co-precipitation method, were analyzed by XRD and FTIR which suggested that Y^{3+} ions were incorporated into the crystal lattice for all the samples. However, the XRD analysis of the sintered samples showed that secondary phases appear in the samples with $x > 0.07$. The samples have densities greater than 90% TD and the SEM images showed that the grain size decreases with the addition of yttrium. Dielectric properties measured from 150 to 25 °C in the frequency range of 100 Hz–1 MHz showed that the addition of yttrium slightly increases the ac conductivity and decreases the $\tan \delta$ therefore making the materials better suited for the use in microwave devices.

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1. Introduction

Spinel ferrites, having a general formula AFe_2O_4 , where A annotates a metal ion, are very important materials for many technological applications. They have been widely used due to their excellent magnetic properties in magnetic resonance imaging enhancement, high-density magnetic recording, magnetic fluids, hyperthermal cancer treatment, etc. [1,2]. Polycrystalline spinel ferrites, having low ac electrical conductivity and consequently low eddy currents and dielectric losses, are particularly well suited for use in high-frequency applications. In addition spinel ferrites have also been used in catalysis, decomposition of CO_2 , drug delivery, gas sensing technology, etc. [3–6].

The properties of ferrites are heavily dependent on their microstructure, composition and ion distribution (as the spinel structure is very open), which in turn are very sensitive to the selected preparation process [7,8]. Therefore, by tailoring the composition and the structure of the spinel, it is possible to improve the properties required for specific applications. Because of this possibility to tailor the properties of ferrites, they have been synthesized by various methods, including solid state reaction [9], hydrothermal [10], sol–gel [11], sol–gel autocombustion [12], co-precipitation [13], citrate precursor technique [14], thermal plasma [7] and pulsed wire discharge [15]. Furthermore, precisely

selected processing conditions (such as sintering method, temperature and time) [16,17], and controlled substitution of either A or Fe ions with different cations are also important for desirable performance of ferrite devices [18,19].

$NiFe_2O_4$ is a soft ferrite with an inverse spinel structure and high saturation magnetization, electrical resistivity and chemical stability. Their electrical, dielectric or magnetic properties can be even improved by substitution of Ni^{2+} with divalent cations, most commonly Zn^{2+} [14,20–22], or by substitution of Fe^{3+} with trivalent cations such as Er^{3+} [23], Gd^{3+} [24], Tb^{3+} [25], Y^{3+} [26,27], Al^{3+} [27,37]. The substitution can induce changes in crystallization process, lattice constant, cation distribution and thus changes of the conduction mechanism, dielectric and magnetic behavior. The effect of nonmagnetic Y^{3+} ions on structure and dielectric properties of yttrium substituted nickel ferrites is still not completely defined. One of the reasons can be the fact that some of the researchers have concentrated on measuring the dc conductivity, even though the application of the material is in the high-frequency range. Thus, the aim of the present study is to investigate the influence of substitution of Fe^{3+} with Y^{3+} ions on the ac electrical and dielectric properties of nickel ferrites. Therefore, temperature dependences of electrical and dielectric properties of $NiFe_2O_4$ ceramics substituted with varying amounts of yttrium were measured in alternating fields. Additionally, explanation of the mechanisms governing the electrical and dielectric properties was suggested based on the structural studies, electrical and dielectric measurements, i.e. ac conductivity, dielectric constant and loss tangent.

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2. Experimental

Yttrium substituted nickel ferrite nanopowders, $\text{NiFe}_{2-x}\text{Y}_x\text{O}_4$, where $x = 0, 0.03, 0.05, 0.07, 0.15, 0.2$ and 0.3 , have been synthesized by the co-precipitation method from nitrate salts of nickel (Fluka, $\geq 97\%$), iron (Merck, 99%) and yttrium (Merck, Art. 12516). The nitrates were dissolved in distilled water and heated to 80°C and pH was adjusted to $\text{pH} = 13$ with NaOH. After one hour of intense stirring at 80°C the particles were removed from the dispersion by centrifugation and subsequently washed with distilled water until $\text{pH} = 7$. The resulting powder was dried, uniaxially pressed under 250 MPa of pressure and the pellets were finally annealed at 600°C for 1 h and sintered at 1250°C for 4 h.

X-ray diffraction patterns of the as-synthesized powders and the sintered pellets were obtained in the 2θ range of $20\text{--}80^\circ$ on the Bruker D8 Advanced diffractometer with Sol-X detector and $\text{Cu K}\alpha$ radiation. FTIR analysis was performed on the Thermo-Nicolet Nexus 670 instrument with the DTGS detector. Sample was diluted with KBr and placed in 1–5 cm (IR) UVIRSiL cell and the measurement has been performed at 298 K with a resolution of 2 cm^{-1} . SEM fracture surface images were obtained using a JEOL JSM6460LV scanning electron microscope at different magnifications and acceleration of 20 kV . Prior to SEM imaging, the samples were sputtered with gold to make them conductive.

Electrical and dielectric properties, i.e. ac conductivity, dielectric constant and $\text{tg } \delta$, were calculated from capacitance and resistance of the sintered samples measured by LCR-8101 1 MHz Precision LCR Meter, Gwinstek in the frequency range from 100 Hz to 1 MHz at temperatures ranging from 150 to 25°C in steps of 5°C . Prior to the measurements, the wires of the sample holder were connected to the silver electrodes that were applied to the sides of the samples and the whole sample holder was placed inside the furnace. After reaching the maximum temperature, the sample capacity and resistance were measured while the sample was cooling down gradually.

3. Results and discussion

X-ray diffraction pattern of the as-synthesized powder with the highest yttrium content ($\text{NiFe}_{1.7}\text{Y}_{0.3}\text{O}_4$) is shown in Fig. 1. All the peaks can be ascribed to the spinel phase and no evidence of secondary phases can be seen. The incorporation of Y^{3+} ions into the crystal lattice of nickel ferrite nanopowders can be corroborated by the FTIR spectra of the as-synthesized samples (Fig. 2). A slight shift of the peaks corresponding to M–O at tetrahedral and

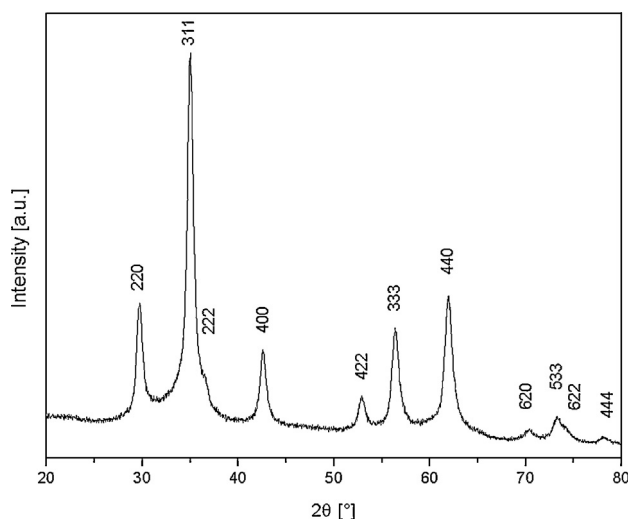


Fig. 1. XRD pattern of $\text{NiFe}_{1.7}\text{Y}_{0.3}\text{O}_4$ nanopowder.

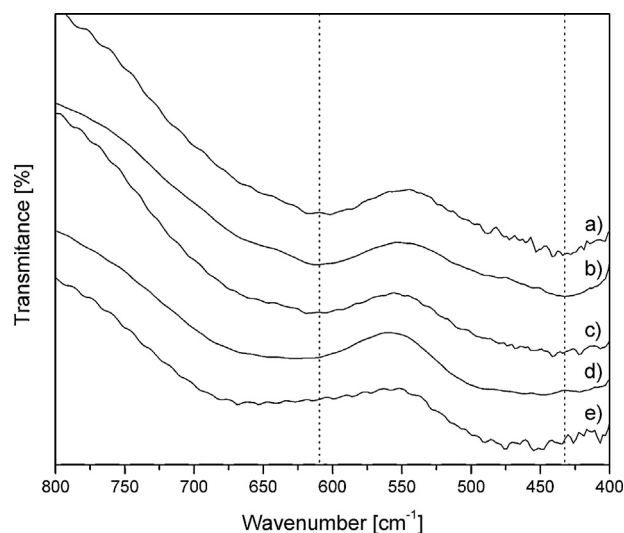


Fig. 2. FTIR spectra of the as-synthesized nanopowders: (a) NiFe_2O_4 , (b) $\text{NiFe}_{1.95}\text{Y}_{0.05}\text{O}_4$, (c) $\text{NiFe}_{1.85}\text{Y}_{0.15}\text{O}_4$, (d) $\text{NiFe}_{1.8}\text{Y}_{0.2}\text{O}_4$ and (e) $\text{NiFe}_{1.7}\text{Y}_{0.3}\text{O}_4$.

octahedral positions toward higher wavenumbers can be observed with the increase of Y^{3+} content. This can be explained by the increased bond length and strain that was caused by the incorporation of the Y^{3+} ions into the crystal lattice.

Fig. 3 shows the XRD data of the samples after sintering at 1250°C for 4 h. For the samples where $x \leq 0.07$ all the peaks match those at the ICDD card no. 10-0325. However, for the samples where $x \geq 0.15$, new peaks begin to appear at $2\theta = 32.4^\circ$ and 33.1° indicating the formation of the secondary phases. These peaks could be attributed to two different iron yttrium oxides – FeYO_3 (ICDD card no. 39-1489) and $\text{Fe}_5\text{Y}_3\text{O}_{12}$ (ICDD card no. 43-0507) and their intensities increase with the addition of yttrium. Similar results were published in a paper by Ishaque et al. [26] where the first indication of the secondary phase appears in the sample $\text{NiFe}_{1.84}\text{Y}_{0.16}\text{O}_4$. The lattice parameter increases with the addition of Y^{3+} ions. However, the rate of increase is reduced after the first appearance of the secondary phases, i.e. for $x \geq 0.15$, as can be seen in the inset of Fig. 3.

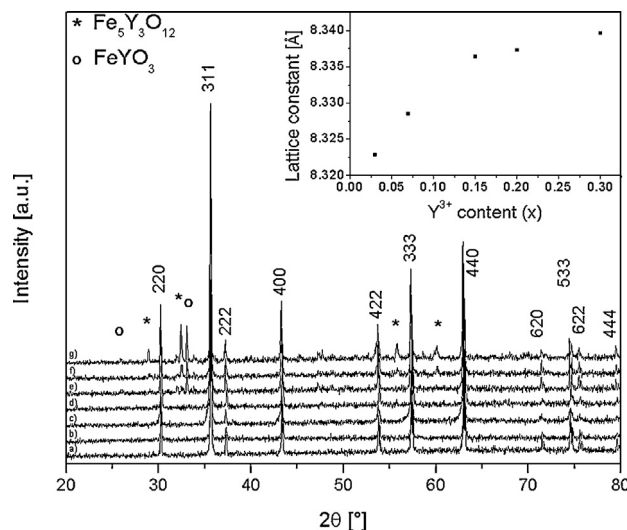


Fig. 3. XRD patterns of samples sintered at 1250°C for 4 h: (a) NiFe_2O_4 , (b) $\text{NiFe}_{1.97}\text{Y}_{0.03}\text{O}_4$, (c) $\text{NiFe}_{1.95}\text{Y}_{0.05}\text{O}_4$, (d) $\text{NiFe}_{1.93}\text{Y}_{0.07}\text{O}_4$, (e) $\text{NiFe}_{1.85}\text{Y}_{0.15}\text{O}_4$, (f) $\text{NiFe}_{1.8}\text{Y}_{0.2}\text{O}_4$ and (g) $\text{NiFe}_{1.7}\text{Y}_{0.3}\text{O}_4$. The inset shows the lattice constant plotted versus Y^{3+} content.

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