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Structural, magnetic and dielectric properties of magnesium doped nickel ferrite nanoparticles

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ABSTRACT

Magnesia doped nickel ferrite nanopowders ($Ni_{1-x}Mg_xFe_2O_4$, x = 0, 0.1, 0.3, 0.5, 0.7) have been synthesized by co-precipitation method and all samples were annealed at 900 °C. The structural, morphological and magnetic properties of the products were characterized by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), field emission scanning electron microscopy (FESEM), energy-dispersive X-ray (EDX) spectroscopy and vibrating sample magnetometer (VSM). Also microwave dielectric properties were measured at frequencies 1 GHz–12 GHz. XRD analysis indicates that all samples have spinel structure and there is not any extra phase in all samples. The increase in magnesia concentration caused to variation in the average crystallite size and lattice constant. Doping magnesia in nickel ferrite gave rise to a decrease in the saturation magnetization whereas doping led to increase in coercivity field. The parameters like dielectric constant, dielectric loss and ac conductivity of the nanoparticles samples are studied in the frequency range from 1 to 12 GHz. All these parameters show, size dependent variations. Complex dielectric permittivity of samples was decreased by increasing in frequency.

1. Introduction

Spinel ferrite nanoparticles are one of the most important nanomaterials because of their significant electronic, optical, electrical, magnetic properties which are different from their bulk structure [1–7]. Molecular formula of magnetic spinel ferrites is (M^{2+}) $[Fe^{3+}]$ $O^{2-}{}_4$, where M^{2+} is divalent cation. M^{2+} and Fe^{3+} occupy the tetrahedral (A) and octahedral (B) interstitial sites of the fcc lattice formed by O^{2-} ions in normal spinel ferrites, respectively [8]. Nickel ferrite ($NiFe_2O_4$) has an inverse spinel structure in the bulk state [9]. Single crystalline nickel ferrite ($NiFe_2O_4$) which has an inverse spinel structure is reported to show a mixed spinel structure when grain size is decreased to nanometer range and the magnetic moment at low temperatures is lower than the value of the bulk material [10]. Chinnasamy et al. [11] found a mixed spinel structure for nickel ferrite when the grain size is reduced to a few nanometers. Due to the cation redistribution or change in the coordination of the tetrahedral and octahedral sites, the structure of $NiFe_2O_4$ deviates from an inverse spinel structure to a mixed spinel structure [10].

In this structure octahedral site was occupied by Ni^{2+} ions and Fe^{3+} ions will occupy both the tetrahedral and the octahedral sites. Nickel ferrite powders have been investigated for many applications, including ferrofluids, catalysts, microwave devices, gas sensors, and magnetic materials [1–7]. The structural and magnetic properties of spinel ferrites depend on the magnetic interaction and cation distribution in the two tetrahedral (A) and octahedral (B) lattice sites.

Insulating materials can be heated by applying high frequency electromagnetic energy. The physical origin of this heating conversion depends on the ability of the electric field to induce polarization of charges within the heated product. Polarization phenomena are the local reorganization of linked and free charges. The interaction between a dipole and an electric or magnetic field is clearly interpreted by quantum theory. For electric fields the coupling is weaker and there is such demultiplication of quantum levels that they are very close to each other. Because of the weak coupling between dipole and electric field, there are no quantified orientations and study of the interaction between a dipole in an electric field gives more information about the surroundings of the dipole than about then dipole itself. Dipoles are, moreover,

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associated with chemical bonds, and any motion of the dipole induces a correlative motion of molecular bonds, whereas motion of a magnetic moment is totally independent of any molecular motion. This polarization cannot follow the extremely rapid reversals of the electric field and induce heating of the irradiated media. The interaction between electromagnetic waves and matter is described by the two complex dielectric permittivity (ε_r) and the magnetic permeability (μ_r). The electric components of electromagnetic waves can produce currents of free charges (electric conduction that can be by electronic or ionic origin). It can also induce local reorganization of linked charges (dipole moments). The local reorganization of linked and free charges is the physical origin of polarization phenomena. The storage of electromagnetic energy within the irradiated medium and the thermal conversion in relation to the frequency of the electromagnetic stimulation appear as the two main points of polarization phenomena induced by the interaction between electromagnetic waves and dielectric media. The wave–matter interactions are expressed by the complex formulation of the dielectric permittivity as described by Eq. (1):

$$\varepsilon_r = \varepsilon' + i\varepsilon'' \quad (1)$$

where ε_r is the dielectric permittivity of a medium, ε' and ε'' are the real and imaginary parts of the complex dielectric permittivity. The storage of electromagnetic energy is shown by the real part, whereas the thermal conversion is attributed to the imaginary part. The dielectric properties of nickel ferrite could be varied by the addition of small traces of cobalt, manganese and copper, and also by bringing deviation from stoichiometry [12].

There are various chemical and physical methods to prepare nanoparticles. Some of the main methods include sol–gel methods [13], sonochemical technique [14], hydrothermal methods [15], microwave processing approaches [16], co-precipitation [17], etc. Among these methods, co-precipitation method attracts much more attention to preparing ferrite nanoparticles. In this work, we investigated the synthesis of the nanostructured nickel ferrite nanopowders by Co-precipitation. Also, the variation of the structural, magnetic and dielectric permittivity of pure and doped nickel ferrite powder is investigated.

2. Materials and methods

2.1. Preparation process

$Ni_{1-x}Mg_xFe_2O_4$ (x = 0, 0.1, 0.3, 0.5, 0.7) nanoparticles were prepared by using a co-precipitation method. All the reagents used in the experiments were supplied by Chem-Lab. Iron chloride hexahydrate ($FeCl_3.6H_2O$), nickel chloride hexahydrate ($NiCl_2.6H_2O$), magnesium chloride anh ($MgCl_2$) and sodium hydroxide (NaOH) were used as starting materials. Salts dissolved in 50 ml double distilled water. We have used 0.1 M solution from nickel and magnesium and 0.2 M iron chloride. Salt solution was added to sodium hydroxide solution (2 M) drop wise till the PH reached close to 13. The reaction was carried out at 85 °C for 2 h with vigorous mixing. Then the precipitated product was centrifuged at 3200 rpm and the products were washed with deionized water for several times. Then powders dried at 70 °C for 24 h. After drying samples were annealed at 900 °C.

2.2. Characterization

The samples were characterized by X-ray diffractometer (XRD) using a PW 1800 (Philips, Netherland) and Cu-Kα radiation (λ = 0.154 nm). The average crystallite size D was obtained by using a Scherrer formula from the line broadening of the XRD peaks corresponding to all planes of the spinel structure. Fourier transform infrared spectroscopy (FTIR, Nicolet Magna, IR560, USA) studies were carried out on the samples to examine the compositional characteristics. Quantitative elemental analysis of the samples was done by the EDX fluorescence analysis and a field emission SEM (FESEM, Mira 3-XMU) was used for microstructure investigation of surfaces. The room temperature magnetic characterization was done using a vibrating sample magnetometer (VSM, Meghnatis Daghigh Kavir Co, Iran). Magnetic parameters such as saturation magnetization (M_s), remanence magnetization (M_r) and coercivity (H_c), initial magnetic susceptibility (χ_i) were calculated from these measurements. The dielectric permittivity was measured in the microwave frequency range from 1 GHz to 12 GHz using the Agilent Dielectric Probe Kit85070B. Measurements of dielectric parameters such as dielectric constant (ε'), dielectric loss (ε''), loss tangent (tanδ) were done at room temperature.

3. Results and discussion

3.1. Microstructure

The Mg doped nickel ferrite nanoparticles samples were characterized using X-ray diffraction (XRD) in the 2θ range of 20–70°. XRD patterns of ferrite samples with different compositions annealed at 900 °C are indicated in Fig. 1. All samples have a cubic spinel structure and there are not any extra peaks from other phases which confirmed phase purity of all samples.

The average crystallite size (D) calculated from Sherrer's formula [18].

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (2)$$

where D is the crystallite size, λ is the wavelength of X-ray radiation (λ = 0.154 nm), β is the full width at half maximum of the peaks and θ is the angle of diffraction. The diffraction peaks appeared at Bragg angles 2θ ~30.2°, 35.5°, 43.2°, 53.1°, 57.1°and 62.7° corresponding to (220), (311), (400), (422), (511) and (440) planes respectively that confirm the formation of cubic spinel structure [JCPDS, Card No.10-325]. The crystallite size was altered in all samples which are shown in Table 1. It is clear that Mg doping at first give rise to increase the crystallite size. Then it decreases with further increase in the doping concentration, which is revealed that the crystallite size treatment is a kind of comparison between the driving force for

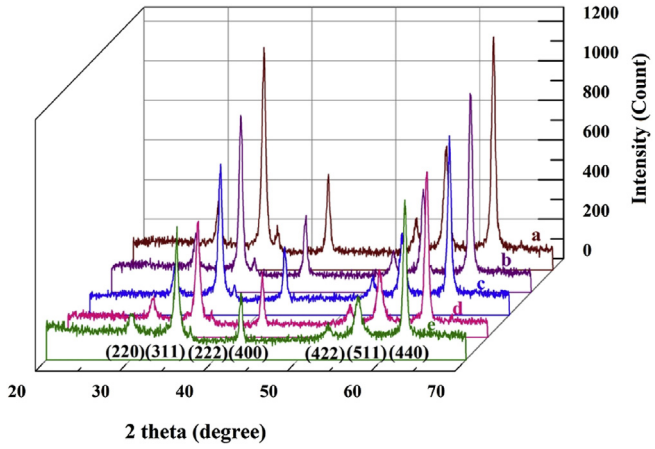

Fig. 1. XRD patterns of a) $NiFe_2O_4$, b) $Ni_{0.9}Mg_{0.1}Fe_2O_4$, c) $Ni_{0.7}Mg_{0.3}Fe_2O_4$, d) $Ni_{0.5}Mg_{0.5}Fe_2O_4$, e) $Ni_{0.3}Mg_{0.7}Fe_2O_4$ nanoparticles, which annealed at 900 °C.