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Frequency, temperature and In^{3+} dependent electrical conduction in $NiFe_2O_4$ powder

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ABSTRACT

A series of polycrystalline spinel ferrites with the composition $NiIn_xFe_{2-x}O_4$ ($0 \le x \le 0.3$) were prepared by the solid state reaction to study the effect of In^{3+} ions substitution on their dc electrical resistivity and dielectric properties. The dc resistivity has been investigated as a function of temperature and composition. The indium ion increases the dc resistivity and activation energy of the system. A study of the dielectric properties of these mixed ferrites, as a function of composition, frequency and temperature, has been undertaken. The dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent ($\tan\delta$) all decreases with frequency as well as with the composition. The dielectric constant (ε') and dielectric loss tangent ($\tan\delta$) were increases with increasing temperature. AC conductivity increases with increase in applied frequency. The dielectric behavior of the present samples is attributed to the Maxwell–Wagner type interfacial polarization. The conduction mechanism in these ferrites is due to electron hopping between Fe^{2+} and Fe^{3+} ions on adjacent octahedral sites.

1. Introduction

Nickel and substituted nickel ferrite is one of the versatile and technologically important soft ferrite materials because of their typical ferromagnetic properties, low conductivity and thus lower eddy current losses, high electrochemical stability, catalytic behavior etc. [1–3]. The polycrystalline soft ferrites having high dc resistivity are used in making the core of the transformers and chokes, ferrites having extremely low dielectric loss and therefore very useful for microwave communication [4]. The dielectric properties of the polycrystalline soft ferrites are sensitive to the applied electric field frequency, preparative methods and preparation conditions, chemical compositions and doping of additives.

Study of the effect of temperature, composition and frequency on the dielectric behavior and dc resistivity offers much valuable information on the behavior of the localized electric charge carriers which can lead to a good explanation and understanding of the mechanism of electric conduction and dielectric polarization in ferrite systems [5]. Electrical properties for substituted $NiFe_2O_4$ have been studied by various researchers [6–8]. Our previous report deals with the structural and magnetic properties of In^{3+} substituted $NiFe_2O_4$ [9]. Though the electric properties of In^{3+} substituted Mg–Mn [10], Ni–Zn–Ti [11] and Mn–Zn [12] ferrites have been studied previously, but In^{3+} substituted $NiFe_2O_4$ ferrite system for their resistivity and dielectric properties has not been treated in the literature. The electrical conductivity and the magnetic properties of ferrites are governed by the Fe^{2+}–Fe^{3+} interaction (spin coupling of the 3d electrons) and also due to Ni^{2+} ions [13]. According to literature reports the resistivity of the parent spinel ferrite increases with the substitution of In^{3+} ions [10,11]. In view of this fact, we have investigated the electrical and dielectric properties of In^{3+} substituted $NiFe_2O_4$ in order to understand the conduction mechanism and the role of In^{3+} ions.

2. Experimental procedure

Ferrite samples of the system $NiIn_xFe_{2-x}O_4$ with x values ranging from 0.0 to 0.30 (in the step of $x = 0.05$), were prepared by the solid state reaction. The details of the experimental procedure were already discussed in our previous work [9]. The dc resistivity of the samples was measured using the two-probe method where silver paste is used as a contact material. The sample is firmly fixed between two electrodes to have a good surface contact. An auxiliary heater is used for heating of all investigated samples. The temperature was measured using a chromel–alumel thermocouple.

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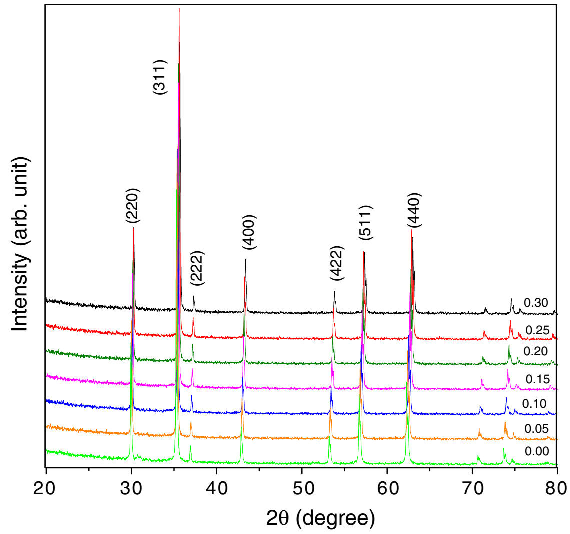

Fig. 1. X-ray diffraction patterns of $NiIn_xFe_{2-x}O_4$.

3. Results and discussion

3.1. Structural properties

The X-ray diffraction patterns of all the samples are given in Fig. 1. The X-ray diffraction patterns show the formation of single phase cubic spinel structure without any impurity peak. The lattice constant 'a' was calculated from the interplaner spacing d-values and the corresponding Miller indices (hkl) using the equation, $a = d\sqrt{(h^2 + k^2 + l^2)}$ [8]. It is observed that 'a' increases from 8.337 Å to 8.424 Å as the In^{3+} substitution increases. This can be related to the fact that the In^{3+} ion has ionic radius of 0.91 , which is larger than the Fe^{3+} (0.67). When the larger indium ions enter the lattice, the unit cell expands while preserving the overall cubic symmetry.

Small amount of pores in SEM image (Fig. 2) reveals that the sintering is done in a satisfactory manner. The enhancement of the grain growth is observed as effect of increasing indium content. Uniform grains are progressively increased from 2.5 µm (x = 0.0) to 8.1 µm (x = 0.3) with increasing In^{3+} content x and the ferrite sample exhibit an aggregated continuous grain growth.

3.2. DC resistivity

Fig. 3 shows the variations in dc resistivity with temperature. It is observed from Fig. 3 that for all the samples resistivity decreases with increasing temperature, indicating the semiconducting nature of the samples. The temperature dependence of resistivity found to follow the Arrhenius equation [10].

$$\rho = \rho_0^{-\left(\frac{E_g}{k_B T}\right)} \tag{1}$$

where ρ_0 is the pre-exponential factor with the dimensions of Ω-cm, k_B is the Boltzman constant (8.6173439×10^{-5} eV/K), Eg is the activation energy, and T is the absolute temperature.

The conduction mechanism of ferrites can be explained on the basis of the Verwey de Boer [14] mechanism that involves exchange of electrons between the ions of the same elements present in more than one valence state and distributed randomly over crystallographic lattice sites. A very small amount of Fe^{2+} and Ni^{3+} are formed during

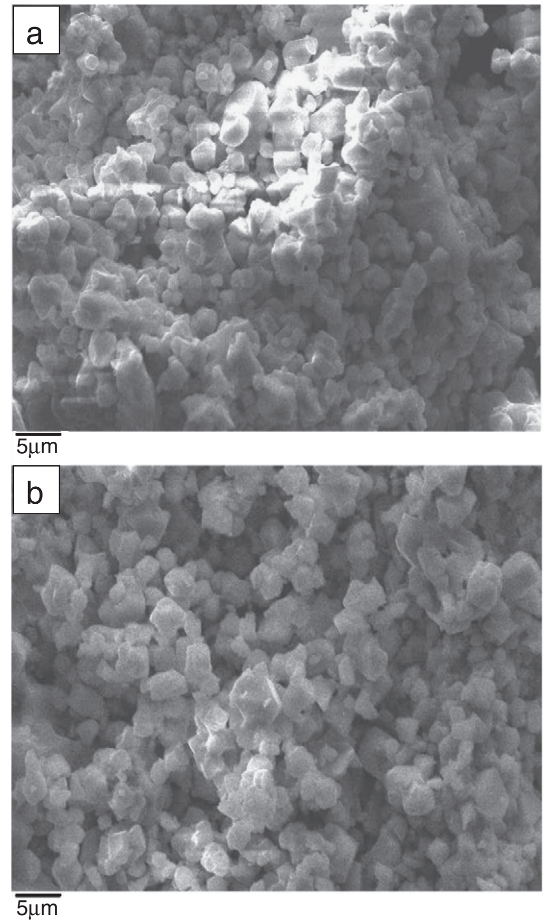

Fig. 2. Typical SEM for (a) x = 0.1 and (b) x = 0.2 of $NiIn_xFe_{2-x}O_4$.

the sintering process, and electron exchange is believed to be between the iron ions and nickel ions [15] which can be written as;

$$Ni^{2+} + Fe^{3+} \leftrightarrow Ni^{3+} + Fe^{2+} \tag{2}$$

The decrease in resistivity with increase in temperature is due to the increase in drift mobility of the charge carriers. Also conduction in ferrites is attributed to hopping of electrons from Fe^{3+} to Fe^{2+} at elevated temperatures [4]. For spinel ferrite samples usually three

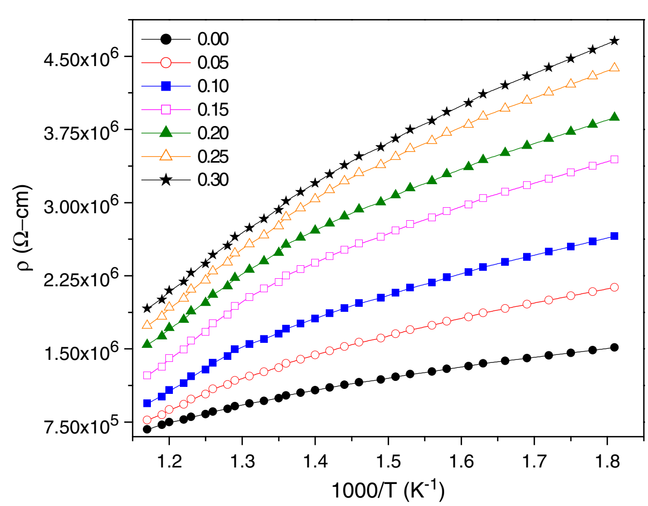

Fig. 3. Variations in dc resistivity with temperature.