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Investigation of nickel effects on some physical properties of magnesium based ferrite

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ABSTRACT

$Mg_{0.6-x}Ni_xCo_{0.4}Fe_2O_4$ (x = 0 and x = 0.2) ferrites are elaborated under same conditions using the Pechini sol-gel technique. The obtained samples reveal a single phase without any detectable of secondary phase. But, the introduction of nickel in the parent ferrite system affects the grains shape, lattice parameters, volumetric mass density and the electrical resistivity. The X-ray diffraction patterns indicate that the investigated ferrites reveal a single phase without any detectable of secondary phase. Also, grains in the nickel doped compound have prismatic shape which indicates a better crystallization than in undoped one. From electrical measurements, a dramatically decrease of resistivity is observed at low temperature range. A decrease with 7 decades is observed in the temperature range 100 K-500 K. Both samples exhibit a semiconductor character in an important temperature range 80 K-700 K. The nickel effect on the electrical resistivity is well pronounced at higher temperature region (T > 150 K). Also, the nichel addition reduces the activation energy. Such variation is associated to the chemical composition, cristallite size, porosity, grain boundary and density effects. In a specific temperature range (ΔT), the ac-conductivity of the investigated materials becomes almost frequency independent. For x = 0 and x = 0.2, such range increases from ΔT = 100 K to ΔT = 220 K respectively. For x = 0.2 and for $f \leq 160$ kHz, such range is extended from ambient temperature to 700 K to be ΔT = 400 K. This behaviour may be important to make the material suitable for some desired applications. Impedance analysis confirms the contribution of grain boundary on transport properties and proves the presence of relaxation phenomenon in nickel-magnesium ferrite. The temperature dependence of the average normalised change parameter is in good agreement with the temperature dependence of the dc-resistivity and the ac-conductivity of the investigated samples.

1. Introduction

Ferrites nanomaterials are the subject of intense research in the aim to understand their physical properties and to enhance their performance for technological applications [1–9]. Due to both, surface and quantum effects, nanomaterials are considerably different of bulk ones [10]. Therefore, these effects affect extensively the physical properties of these materials specially electrical, magnetic and optical ones. Due to their properties, the nanosized particles are required as materials for several technological applications such as high frequency devices, high density recording, color imaging, magnetic refrigerators and ferrofluids [11,12]. Nano-ceramic materials are extensively employed in magnetic resonance imaging. Also, these materials replace radioactive one on delivering drugs to specific areas of the human body. Ferrites are the major materials constituting ceramic compounds. Specially, $CoFe_2O_4$, $MgFe_2O_4$, and $NiFe_2O_4$ are a very interesting ferrites used in different technological fields. Nickel based ferrites are the most used as soft magnets and low loss materials at high frequencies [13]. Some works have been consecrated to the preparation and to the physical properties of $NiFe_2O_4$ nano-ceramics synthetised by different routes. A literature investigation demonstrates that the

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structural, magnetic, electrical and optical properties of these materials depend essentially on the synthesis methods, the concentration and the nature of the substituted element, sintering temperature of the sample and its grain size. The wide use of these materials in several applications requires the improvement of many elaboration procedures such as: mechanical-alloy, hydrothermal, pulsed wire, ultrasonic, shockwave, freeze drying, co-precipitation, spray drying and sol–gel methods [14–24]. Recently, sol–gel methods are one of the best advantageous procedures for the preparation of nano-ferrite samples. Such method permits, in a reasonably short time and low temperature, the obtention of ultrafine particles. It is noted that synthesis's parameters, precursors, pressure, reaction time, and the temperature [25,26], of Ni-ferrite using sol–gel process affect intensely the physical properties of ferrites materials. In the literature [27–30], it is observed that the best possible properties of nickel ferrites compounds can be attained at a particular annealing temperature. It is found that structural and electrical conductivity of these materials are dopant element content dependent. Conduction process is affected by nickel concentration. Structural, electrical and magnetic behavior of Ni-ferrite and its useful application have been recently investigated [31]. Also, the nickel concentration effects, at room temperature, on the electrical properties of ferrites are studied [32]. Electrical resistivity and thermo-electric power investigations indicate the contribution of small polaron hopping (SPH) mechanism in the transport process [33]. It is well known that the distribution of the cations and the chosen site participate on determining the physical properties of ferrites materials.

Several research groups have carried out works on physical investigations of ferrites with doping by Ni, Mg, Zn, Mn, Ti and Cu [34–40]. We have recently investigated the Ni doping effects on the phase purity, the homogeneity, lattice structure, cell parameters and the electrical properties of $Zn_{1-x}Ni_xFe_2O_4$ ferrites prepared using the conventional solid state reaction method [40]. When Ni is introduced the conductivity of the material is enhanced. Such effect disappears at high temperature (T > 600 K). It is also found that activation energy, of transport process, is sensitive to both Ni content and temperature range. In the line with the previous work, we report in the present one an effort on understanding the structural and the electrical behavior of Ni-Mg-ferrites synthetised by sol-gel method. We intend to understand and to optimize the properties of low cost compounds, using inexpensive and easy elaboration process, useful for some desired technological applications.

2. Experimental procedure

2.1. Preparation of the materials

The $Mg_{0.6-x}Ni_xCo_{0.4}Fe_2O_4$ (x = 0 and x = 0.2) samples are elaborated under same conditions using the Pechini sol-gel technique [14]. The $Ni(NO_3)_2{\cdot}6H_2O$, $Mg(NO_3)_2{\cdot}6H_2O$, $Cu(NO_3)_2{\cdot}3H_2O$ and $Fe(NO_3)_3{\cdot}9H_2O$ precursors are used with a stoichiometric amounts. At first, the metal nitrates are dissolved in distilled water to obtain a mixed solution. Successively, after the complete dissolution of the nitrates, a citric acid is introduced with controlled amount and dissolved with stirring. The molar ratio was fixed as 1:1 of nitrates to citric acid. A small quantity of ammonia was added to the solution to adjust the pH value at about 7.

The obtained solution is heated on hot plate under regular stirring to 363 K. Then, the ethylene glycol, used as a polymerization agent, is added. Heating and stirring are maintained as far as obtaining the gel after about 4 h. Formerly, the obtained gel is dried at 523 K to obtain a frothy dry which will be grounded in a mortar, then dried again at 873 K for 12 h in air. After the heat treatment process, a powder is obtained. Finally, such powder is pressed into pellets and sintered, at 1073 K for 24 h and at 1323 K for 48 h, in order to obtain the desired crystalline phase.

2.2. Measurements

The "PANalytical X'Pert Pro" diffractometer with filtered (Ni filter) Cu radiation is used to collect the powder X-ray diffraction (XRD) data. A (2θ) range 15°–80° with a step size of 0.0167° and a counting time of 18s per step are used. To obtain the instrumental resolution function, standard *Si* powder is employed. To analyse the morphology of the investigated samples, the scanning electron microscopy (SEM; Philips XL30 microscope) is used under an accelerating voltage of 20 kV. To conduct electrical measurements, the sample should be prepared previously. At first, a thin silver film is deposited on the both side of the pellet through a circular mask of 6 mm of diameter. Consequently, a configuration of plate capacitor is obtained. The latter is used to measure both the conductance and the capacitance as a function of frequency and temperature. The investigated sample is attached in a cryostat to vary the temperature from 77 K to 700 K. The electrical measurements are conducted under vacuum and in dark with an Agilent 4294 analyzer using a signal amplitude of 20 mV [41].

3. Results and discussion

3.1. Structural properties

Fig. 1 shows the X-ray diffraction patterns of $Mg_{0.6-x}Ni_xCo_{0.4}Fe_2O_4$ (x = 0 and x = 0.2) samples. The observed results indicate that the investigated ferrites reveal a single phase without any detection of secondary phase. As shown in Fig. 1, the diffraction peaks are indexed using "*X'Pert HighScore Plus*" software with respect to the cubic spinel type structure with the space group $Fd\bar{3}m$. The XRD data and equation (1) [42,43] are used to calculate the lattice constant *a*.

$$a = \frac{\lambda}{2}\frac{\sqrt{h^2+l^2+k^2}}{sin\theta} \quad (1)$$

where λ is the wavelength and *h k l* are the corresponding Miller indices. The obtained lattice constant are *a* = 8.351 Å and

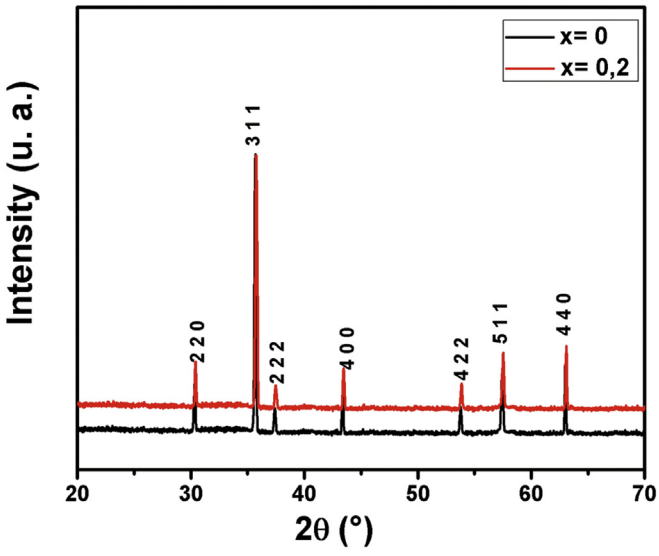

Fig. 1. XRD patterns of $Mg_{0.6-x}Ni_xCo_{0.4}Fe_2O_4$ (x = 0 and x = 0.2) samples.