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Synthesis, electrical and magnetic properties of sodium borosilicate glasses containing Co-ferrites nanoparticles

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ABSTRACT

Co-ferrites nanoparticles that have been prepared by the co-precipitation method were added to sodium borosilicate ($Na_2O–B_2O_3–SiO_2$) glass matrix by the solid solution method and they were characterized using X-ray diffraction (XRD), transmission electron microscopy (TEM), Fourier transform infrared (FTIR) and magnetization measurements. (XRD) revealed the formation of the Co-ferrite magnetic crystalline phase embedded in an amorphous matrix in all the samples. The investigated samples by (TEM) showed the formation of the cobalt ferrite nanoparticles with a spherical shape and highly monodispersed with an average size about 13 nm. IR data revealed that the BO_3 and BO_4 are the main structural units of these samples network. IR spectra of the investigated samples showed the characteristic vibration bands of Co-ferrite. Composition and frequency dependent dielectric properties of the prepared samples were measured at room temperature in the frequency range 100–100 kHz. The conductivity was found to increase with increasing cobalt ferrite content. The variations of conductivity and dielectric properties with frequency and composition were discussed. Magnetic hysteresis loops were traced at room temperature using VSM and values of saturation magnetization M_S and coercive field H_C were determined. The obtained results revealed that a ferrimagnetic behavior were observed and as Co-ferrite concentration increases the values of M_S and H_C increase from 2.84 to 8.79 (emu/g) and from 88.4 to 736.3 Oe, respectively.

1. Introduction

Due to their unusual physical properties, Magnetic nanoparticle materials have been the focus of a number of researchers when compared to their bulk form. They can be used in different technological applications, such as electronic devices, transformer cores, magnetic devices, switching devices, recording tapes, permanent magnets, hard disc recording media, flexible recording media, active components of Ferro-fluids, magnetic drug delivery, catalysis, magnetic refrigeration, detoxification of biological fluids, magnetically controlled transport of anti-cancer drugs, magnetic resonance imaging (MRI), and contrast enhancement and magnetic cell separation [1].

As Spinel ferrites possess high resistivity, they have a large variety of their structural, electronic, electrical, magnetic and catalytic properties, and also as they possess a negligible eddy current losses [2–4]. Cobalt ferrite $CoFe_2O_4$ is one of the most significant magnetic materials which can be widely used in electronic technologies, especially on magnetic and magneto-optical recording media as a result of their high magnetocrystalline anisotropy, moderate saturation magnetization, mechanical hardness, and high coercivity and chemical stability [5–7]. Magnetic nanoparticles are expected to improve the properties of the materials in which they are embedded. Incorporation of the magnetic nanoparticles into suitable host matrices gives a class of composite materials that have different applications in photonic devices such as light waveguides and optical switches [8]. Among these host matrices, glasses are more attracted matrices due to the fact that they can serve as a source for nano-structured systems with good optical nonlinear properties [9]. Also, due to the fact that they are easy to fabricate, they are inexpensive; besides that the composition of the glass can be well designed and tuned according to the needs of the encompassed photonic components [10]. Compared to SiO_2 glasses, borosilicate glasses based on the Na_2O-B_2O_3-SiO_2 system play a significant role in various applications, ranging from chemically and thermal resistant technical glass to optical, sealing and nuclear waste [11].

The aim of this work is to synthesis and studies the properties of sodium borosilicate glasses that contain Co-ferrites nanoparticles, as to the best of our knowledge this is the first time to Synthesis sodium borosilicate glasses with high cobalt ferrite content and to study their structure, electrical and magnetic properties in regard to possible

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application.

2. Experimental technique

Cobalt ferrite $CoFe_2O_4$ nanoparticle was prepared by the chemical co-precipitation method [12]. The glass sample 10 SiO_2-40 B_2O_3-50 Na_2O was prepared by the melt quenching technique. Then, the product glass sample was grained in a ball mill for 2 h. The powders of the glass and the Cobalt ferrite $CoFe_2O_4$ nanoparticle were mixed and milled for 2 h according to the following compositions (100-x) (10 SiO_2-50 Na_2O-40 B_2O_3): x $CoFe_2O_4$, where (x=10, 20, 30, 40, 50 wt%). The produced fine powders were pressed into disc-shaped pellets (18 mm in diameter and 2–3 mm in thickness) at an isotactic pressure of 5 t. The pelletized samples were finally sintered at 250 °C in the air for 2 h.

The density (ρ_g) of the glasses was determined at room temperature using Archimedes' principle with toluene (density=0.867 g/ml at 20 °C) as buoyant liquid [13].

The connected porosity was calculated using the imbibition method. The samples are immersed in water until they are fully saturated. The sample is weighed before and after the imbibition, and the density of the water is known, then the difference in weight is $\rho\ V_P$, and the pore volume V_P can be calculated. The bulk volume V is measured using Vernier callipers, and assuming that the sample is cylindrical. V_P and V can then be used to calculate the *connected porosity*. The time required for saturation depends upon the sample permeability.

The reproducibility of the density data obtained was no worse than ± 0.01 g/cm^{-3}.

X-ray diffraction (XRD) patterns of the prepared samples under study were recorded with (BRUKUR D8ADVANCE), with Cu-Kα radiations.

The microstructure of the samples under study and in particular the crystalline grains formed as a result of nanocrystallization or massive crystallization, were observed by Field Transmission Electron Microscopy (JEOL 2010 (DV 300W1)) set-up.

The measurements of IR transmission spectra were recorded at room temperature by the KBr disc method using an FTIR spectrophotometer type Tensor 27, Bruker over the wave number 4000–200 cm^{-1}.

The electric measurements of dielectric constant, ε', dielectric loss, ε'', and ac conductivity, σ_{ac} were carried out at room temperature using (Hioki 3520 LCR HiTester Meter Bridge) in the frequency range of 100–100 kHz (see more details in [14]).

Room temperature magnetic measurements were carried out using the Lake-Shore vibrating sample magnetometer (VSM) model 7410. Hysteresis loops were traced in a magnetizing range from −25 to 25 kG and values of saturation magnetization M_S and coercive field H_C were determined.

3. Results and discussion

3.1. X-ray diffraction

Fig. 1 shows the XRD patterns of Cobalt ferrite $CoFe_2O_4$ nanoparticle prepared by the chemical co-precipitation method and the samples 10 SiO_2-40 B_2O_3-50 Na_2O: X ($CoFe_2O_4$) with (x=0,10,20,30,40,50 wt %) after calcinations at 250 °C for 2 h. The vitreous phase coexists with the crystalline phase (there are remarked glass diffraction patterns (x=0) which are over posed with the peaks from the crystalline phase of $CoFe_2O_4$ nanoparticle). Comparing the XRD pattern in Fig. 1 with the standard data, Joint Code for Powder Diffraction Standards (JCPDS PDF card No. 22-1086), the formation of cobalt ferrite nanoparticles was confirmed. The X-ray powder diffraction pattern of the material also proved its highly crystalline cubic spinel structure. No additional peaks are found ensuring the phase purity and typical amorphous structure for the as-prepared sample (x=0). It can also be seen that the

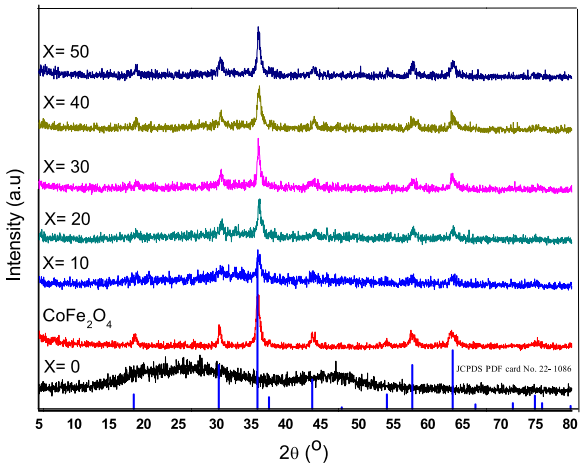

Fig. 1. X-ray diffraction patterns for 10 SiO_2-40 B_2O_3-50 Na_2O: X ($CoFe_2O_4$) samples after calcinations at 250 °C for 2 h.

intensity of the peaks increased with the increasing $CoFe_2O_4$ content. The diffraction peaks are broad because of the nanometer size of the crystallite. The crystallite size 'D' of the samples has been estimated from the broadening of XRD peaks using the Scherrer equation (Pawan et al., 2010).

$$D = K\lambda/\beta\cos\theta \quad (1)$$

where k is a constant equal to 0.9, λ is the wavelength of the X-ray (CuKα radiation), θ is the diffraction angle, and β is the full width half maximum (FWHM). The average values of D, listed in Table 1.

3.2. Transmission electron microscope

Fig. 2**a** shows the micrograph for **(**$CoFe_2O_4$) samples and 10 SiO_2-40 B_2O_3-50 Na_2O: X ($CoFe_2O_4$). As seen in Fig. 2**a**, one can observe a large number of monodispersed nano-sized particles in the glass matrix for samples have (0≤ x≤50). The micrograph show uniform grains with a nearly similar spherical shape and size in each sample. The average values of particles size is around 13 nm for sample (0≤ x≤50). The size of these nanoparticles is in agreement with the result of XRD listed in Table 1. Fig. 2**b** shows the electron diffraction pattern for the cubic spinel structure of ($CoFe_2O_4$) sample [15].

3.3. Density measurements

The density was theoretically calculated for the base glass according to the following equation: $\rho_{calc.} = \sum_i x_i\rho_i$, where x_i and ρ_i are the molar fraction and density of each component, respectively. Theoretically, calculated density (2.38 g/cm^3) is higher than the experimentally obtained value (2.22 g/cm^3**)**. This may be due to the structural changes and the formation of BO_4 units that has higher molecular volume than BO_3 unit and the bores exist in the glass structure, as the theoretical model does not take into account these considerations.

The $CoFe_2O_4$ nanoparticles have a diameter of tenth nanometers as shown in the figures of the transmission electron microscope. So, $CoFe_2O_4$ nanoparticles can be easily fitted into the bores that exist in the glass structure so it does not affect the molecular volume and leads to a decrease in the porosity from (24.8) to (15.7). So, the increase in the density with increasing $CoFe_2O_4$ content could be attributed only to an increase of the average molecular weight of oxide ions in the glass due to that, both CoO and Fe_2O_3 have a relative higher molecular mass.

Density responds to variations in glass composition sensitively in technological practice. Density of glass, in general, is explained in terms of a competition between the masses and size of the various structural groups present in glass. Accordingly, the density is related to how tightly the ions and ionic groups are packed together in the

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