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Effect of γ-rays irradiation on the structural, magnetic, and electrical properties of Mg–Cu–Zn and Ni–Cu–Zn ferrites

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ABSTRACT

Nanoparticles of $Ni_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ and $Mg_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$, have been synthesized by citrate precursor method. Then some of the prepared samples have been irradiated by γ-rays of ^{60}Co radioactive source at room temperature with doses of 1 Mrad and 2 Mrad, at a dose rate of 0.1 Mrad/h to study the effect of γ-rays irradiation on some structural, magnetic and electrical properties of the samples. The X-ray diffraction analysis (XRD), transmission electron microscopy, Fourier transform infrared spectroscopy and vibrating sample magnetometer measurements have been used to investigate the samples. The XRD results show that the irradiation has caused a decrease in the crystallite size and the measured density and an increase in the porosity, specific surface area, and microstrain in the case of Ni–Cu–Zn ferrite whereas in the case of Mg–Cu–Zn ferrite the reverse trend has been noticed. The lattice constant of the investigated samples has been increased with the increase of irradiation due to the conversion of Fe^{3+} (0.67 Å) to Fe^{2+} (0.76 Å). The magnetization results show an increase in saturation and remnant magnetizations for the two prepared ferrites after γ-rays irradiation. The main reason of this behavior is most probably due to the redistribution of the cations between A and B sites. The cation distribution has been proposed such that the values of theoretical and experimental magnetic moment are identical and increase as the magnetization increases. Moreover, a theoretical estimation of the lattice constant has been calculated on the basis of the proposed cation distribution for each sample and compared with the corresponding experimental values obtained by XRD analysis; where they have been found in a good agreement with each other. This can be considered as another confirmation of the validity of the cation distribution. Moreover, the cation distribution is thought to play an important role in increasing the values of dc conductivity of all samples with increasing the irradiation dose. The frequency dependence of ac conductivity, dielectric constant and dielectric loss of all samples have been studied. The Cole–Cole plots of (Z'' vs. Z') give different two overlapping incomplete semi-circles depending upon the electrical parameters. Also, The Cole–Cole plots of (M'' vs. M') insure that the electric stiffness is the dominant property of the investigated samples.

1. Introduction

Recently, magnetic nanoparticle materials are studied intensively due to their novel physicochemical properties. Among these materials, ferrites are considered a group of technologically important materials that are used in the fabrication of magnetic, electronic and microwave devices. They have gained technological importance by virtue of their high resistivity and negligible eddy current losses [1]. With the rapid development of mobile communication and information technology, small, inexpensive, and high performance electronic devices are in high demand [2]. In this regard, Ni–Cu–Zn [3–6] and Mg–Cu–Zn ferrites [7–10] are employed dominantly as magnetic materials for multilayer chip inductors (MLCIs) because of their suitable electromagnetic properties at high frequencies and their low sintering temperature besides that they are economical and easy to synthesize [11]. MLCIs have recently been developed as a surface mounting device (SMD) for miniaturization of electronic devices. They are used in the high-frequency circuitry applications and they are important components for the electronic products, such as cellular phone, notebook computer, video camera, etc. [12]. MLCIs are manufactured using thin sheets made of soft ferrite on which coil patterns are printed with metallic silver paste. By arranging these sheets in multiple layers, a spiral-shaped internal electrode pattern is produced. The multilayer technique allows the coil to be formed without the need to wind wire on a core, facilitating both miniaturization and mass production. This multilayer ceramic metal composite should be co-fired below 950 °C to suppress

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interfacial diffusion of Ag metal in to the ceramic as the melting point of Ag is 961 °C [13]. For this reason, Ni–Cu–Zn and Mg–Cu–Zn ferrites are considered as promising candidates to be used in this application because they could be sintered below 950 °C particularly if they were prepared by the chemical wet method.

The recent increase in the development of measuring devices and data acquisition modules in nuclear reactors and accelerators raises the problem of the irradiation effects on these electronic components. Even the satellites and spacecrafts can be exposed to cosmic radiation of rather high dose. The radiation may cause excitation, ionization of atoms, or disturbance in the structure of matter [14]. Gamma rays are regarded as one of different kinds of radiations which may produce defects in the materials by affecting the structural, morphological and physical properties. Regarding the change in the structural properties of ferrites when irradiated by gamma rays, it has been reported [15] that it may be due to the breaking of ferrimagnetic ordering, surface state pinning, cation inversion, etc.

Accordingly, the aim of the present work is to prepare nanoparticles of Ni–Cu–Zn and Mg–Cu–Zn ferrites with the following chemical formulae: $Ni_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ and $Mg_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ respectively; and to perform a comparison study of the effect of γ-rays irradiation with different doses (1 Mrad and 2 Mrad) on some structural, magnetic and electrical properties of these two compositions of ferrites which may represent an industrial importance as mentioned above.

2. Experimental

Ni–Cu–Zn and Mg–Cu–Zn ferrites with compositional formulae $Ni_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ and $Mg_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$, respectively have been synthesized by citrate precursor auto combustion method. Ni $(NO_3)_2 \cdot 6H_2O$, Mg $(NO_3)_2 \cdot 6H_2O$, Zn $(NO_3)_2 \cdot 6H_2O$, Cu $(NO_3)_2 \cdot 3H_2O$, Fe $(NO_3)_3 \cdot 9H_2O$ and $C_6H_8O_7$ have been used as starting materials. All chemicals used are of high grade purity. Stoichiometric amounts of metal nitrates and citric acid - which is used as a fuel – have been dissolved separately in distilled water keeping the molar ratio of the citric acid to all the metal nitrates is 1:1. The mixture of aqueous solutions of citric acid and metal nitrates has been heated to 50 °C with constant stirring for about 1 h and has been neutralized to pH 7 by adding ammonia solution drop-wise until a kind of clear solution has been formed. The neutralized solution has been evaporated to dryness by heating on a hot plate at 400 °C until a very viscous gel has been finally formed. The gel then has ignited, undergoing a strong auto combustion process with the evolution of large amount of gases, which have started at the hottest zone of the beaker and propagated from the bottom to the top, giving rise to a dark gray product with a structure similar to a branched tree. The formation stages of one of the investigated samples are shown in Fig. 1(a)–(h). The resulting products of all the samples have been grinded thoroughly in an agate mortar then sintered at 600 °C for 3 h. Each sample of the $Ni_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ and $Mg_{0.35}Cu_{0.15}Zn_{0.5}Fe_2O_4$ has been divided into three groups which have been labeled as [A_o, A_1 and A_2] and [B_o, B_1 and B_2], respectively. The samples [A_o, B_o] represents the unirradiated samples, while the samples [A_1, B_1] and [A_2, B_2] represent the irradiated by γ-rays of ^{60}Co radioactive source at room temperature with a total exposure dose 1 Mrad and 2 Mrad respectively at a dose rate 0.1 Mrad/h [by using an irradiation cell in the radiation technology unit, Nuclear Research Center, Atomic Energy Authority, Cairo, Egypt]. The unirradiated and irradiated samples have been characterized by X-ray diffraction (XRD) BRUKER-D8 using CuK_α radiation with wavelength $\lambda = 1.5406$ Å. Also, the particle sizes of some of the investigated samples have been checked out using a transmission electron microscope (TEM) JEOL JEM-1230. The magnetization measurements have been carried out for all samples at room temperature up to a maximum field of 20 kG, by using vibrating sample magnetometer (VSM), model Lake Shore 7410. The dc conductivity of disc-shaped pellets of the samples has been measured by using the two probe method in which silver paste has been used as a contact material, while the dielectric measurements have been measured by using impedance analyzer (Hioki Model IM3570).

3. Results and discussion

3.1. Structural properties

The XRD patterns of the unirradiated and irradiated samples of the Ni–Cu–Zn and Mg–Cu–Zn ferrites are shown in Fig. 2(a) and (b), respectively. All the XRD patterns confirm the formation of FCC structure of the samples without any undesired second phases. The average crystallite size 'D_{XRD}', lattice constant 'a', measured density 'ρ_m', X-ray density 'ρ_x', porosity 'P', specific surface area 'S', and microstrain 'η' of the investigated samples are displayed in Table 1. The XRD results show a rather reverse effect of irradiation dose on the crystallite sizes of the Ni–Cu–Zn and Mg–Cu–Zn ferrite samples. In the case of Ni–Cu–Zn ferrites, the crystallite sizes decrease with increasing irradiation dose presenting i.e. the irradiation has increased the disordered grain boundary regions at the expense of ordered parts of the crystallites (grains). On contrast, the crystallite sizes of Mg–Cu–Zn ferrite samples increase showing the role of irradiation energy in increasing the stacking of the unit cells and consequently increasing the well ordered grain sizes. Also, it can be noticed a slight increase of the lattice constant of the two investigated ferrites with increasing irradiation dose due to the dominant effect of γ-rays interaction with the outer electrons of iron in the lattice, where the ferric ions Fe^{3+} with ionic radius (0.67 Å) is converted to ferrous Fe^{2+} with larger ionic radius (0.76 Å) as reported frequently in literature [16–18]. The X-ray density ρ_x of all samples changes with an inverse trend i.e. decreasing with increasing irradiation which is logically accepted since the X-ray density is inversely proportional to the lattice volume. Also, it can be seen that the X-ray density ρ_x of the Ni–Cu–Zn ferrite samples are higher than the samples of the Mg–Cu–Zn ferrites because the atomic weight of Ni (58.71 u) is higher than that of Mg (24.312 u). The measured density behaves as the crystallite size where in the case of the Ni–Cu–Zn ferrite samples it decreases with increasing irradiation dose while it increases in the case of Mg–Cu–Zn ferrites, such observation have been reported by M.M. Eltabey et al. [2]. The porosity and specific surface area of the samples show a reverse trend compared to that of the measured density and crystallite size as expected from their definitions. Moreover, the XRD results show that the microstrain of Ni–Cu–Zn ferrite samples which have been estimated by using Williumson-Hall equation [19] increases, whereas in Mg–Cu–Zn ferrites, it decreases with increasing irradiation dose. It is well known that microstrain arises due to imperfections within the crystalline lattice, including vacancies, dislocations, stacking faults, and others. So, in the present case of investigated ferrites, it seems that the microstrain becomes larger when the disordered regions of grain boundaries become larger and the well ordered grain sizes become smaller.

The TEM images have been used just to estimate and check up the average particle sizes of some of the investigated samples. The TEM images of the unirradiated (A_o, B_o) and irradiated (A_2, B_2) samples are shown in Fig. 3(a)–(d), respectively. It can be seen that the samples have both irregular and spherical-shaped agglomerated particles. The estimated average particle sizes of the samples are displayed in Table 1. It is clear that the average particle sizes