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Preparation and investigation of dc conductivity and relative permeability of epoxy/Li–Ni–Zn ferrite composites

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ABSTRACT

Ferrite nanoparticles – having the compositions $Li_{(x/2)}(Ni_{0.5}Zn_{0.5})_{(1-x)}Fe_{(2+x/2)}O_4$ ($x=0$, 0.2, 0.3) – have been prepared by the co-precipitation method. The prepared powders have been divided into groups and sintered at different temperatures (373 K, 1074 K and 1473 K). X-Ray diffraction analysis (XRD) for all samples has confirmed the formation of the desired ferrites with crystallite sizes within the nanoscale (<100 nm). The dc conductivity, the relative permeability and the magnetization of the ferrite samples have been investigated and according to the results, the sample $Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}$ $Fe_{2.15}O_4$ sintered at 1473 K has been chosen to prepare the composites. The particle size of this sample has been recalculated by using JEOL JEM-100SX transmission electron microscope and it has been found about 64.7 nm. Then, a pure epoxy sample and four pristine epoxy resin /$Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}$ $Fe_{2.15}O_4$ composites have been prepared using different ferrite contents (20%, 30%, 40%, and 50%) wt.%. These samples have been characterized by Fourier transform infrared (FTIR) spectroscopy and their dc conductivity, relative permeability and magnetization have also been investigated. The obtained results indicate that the investigated composites may be promising candidates for practical applications such as EMI suppressor and high frequency applications.

1. Introduction

Ferrites are very important class of magnetic materials from application point of view and are extensively used as permanent magnets in market because of their low price, high magnetic performance, and have attracted attention over years. The very high specific resistance and remarkable flexibility in the magnetic properties makes the ferrite ideal choice material for telecommunications and microwave applications [1]. Ni–Zn ferrite has been used in various electronic devices and electromagnetic applications that require a high magnetic permeability, such as inductors and electromagnetic wave absorbers. However, current interest is to make nanosized Ni–Zn ferrite particles in order to reduce energy losses associated with bulk powders.

Furthermore, some electronic applications require these materials to be pressed into larger shapes with near theoretical density, which is difficult to obtain if the particles have a wide size distribution. Various preparation techniques have been used by researchers for the synthesis of fine particles of ferrites, which exhibit novel properties when compared to their properties in bulk. Preparation condition serves as an effective means to control the particle size. Ultrafine ferrite particles can be prepared by simple chemical co-precipitation method [2].

Among various ferrites, lithium and metal substituted lithium ferrites also have attractive structural and magnetic features that render them as a potential material for various technological applications such as, lithium batteries, recording heads, transformer cores, and noise filters. The structural and magnetic properties of these ferrite systems with respect to different sintering temperature conditions have been reported by many workers [3].

With the rapid development of electronics industry coming into microwave frequency range, the problems of electromagnetic interference (EMI) are becoming more and more important in our daily life. Therefore, the demand for the microwave absorbers in this frequency range is increased to solve the EMI problems. Such electromagnetic materials generally fall into two categories: dielectric composites with conductive fillers and magnetic composites. As far as thickness and working frequency bandwidth are concerned, magnetic composites have obvious advantages. The magnetic fillers used in such composites are ferrite materials, such as spinel ferrites and hexaferrites [4].

Composite materials are one of the main types of the engineering materials, next to metals and alloys, ceramics and polymeric materials. They are made at least of two separate types

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of substances each with its own characteristics, one of them is called matrix while the second is the filler. In composite materials these two phases are immiscible and are separated by boundary interface layer. Such a wide range of constituent materials gives materials scientists' very big field of possibilities to create new composites and new generations of composites. By proper innovative material selection and design, it is possible to change properties of products depending on requirements and future applications. Composite materials find practical applications in many domains of industry for instance in civil engineering, machine building, sport and leisure industry, automotive industry, aerospace industry etc. [5].

Epoxy resin is a promising polymer based on its high electrical resistivity, low dielectric constant, and ease of processing. The mechanical and chemical properties of an epoxy can be tailored to meet the required values for specific applications. A ferrite-based epoxy nano composite thin film with flexibility, light weight, and the ability to be easily manipulated into various shapes may be highly desirable and useful in practical applications [6]. Also it is worth mentioning in this introduction that there are many types of epoxy resin, among them (DGEBA) is the most important resin used by industry [7].

According to our knowledge, a limited number of researches investigating epoxy/Ni–Zn ferrite composites were reported [8–12]. However, epoxy/Li–Ni–Zn ferrite composites was not reported so we would expect that they may be promising candidates for important practical applications. Therefore, the aim of the present work is to prepare such composites and investigate their structural, electrical and magnetic properties and for comparison purposes, a study of the electrical and magnetic properties of all ferrite samples also will be reported besides the epoxy-ferrite composite results.

2. Experimental

2.1. Preparation of ferrite nanoparticles

Ferrite nanoparticles with three compositions $Li_{(x/2)}(Ni_{0.5}Zn_{0.5})_{(1-x)}Fe_{(2+x/2)}O_4$; where $x=(0, 0.2, 0.3)$ have been prepared by the chemical co-precipitation method. The starting precursors were $LiCl\cdot 2H_2O$, $NiCl_2\cdot 6H_2O$, $ZnCl_2$, $FeCl_3$ and NaOH. The details of the method of preparation had been previously published [13]. The powder products of prepared compositions have been divided into two groups the first group has been sintered at 373 K (i.e.100 °C) for 4 h and the second group has been sintered at higher temperatures; where the sample $Ni_{0.5}Zn_{0.5}Fe_2O_4$ has been sintered at 1073 K (i.e. 800 °C) for 4 h and the two other samples $[Li_{0.1}(Ni_{0.5}Zn_{0.5})_{0.8}Fe_{2.1}O_4]$ and $[Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}Fe_{2.15}O_4]$ have been sintered at 1473 K (i.e. 1200 °C) for 4 h to improve crystallinity without introducing undesirable phases. Some amounts of the powders have been pressed into disk-shaped pellets for the electrical measurements.

2.2. Structural characterization by XRD

The structure and crystallite size of the ferrite samples have been investigated by X-Ray diffraction (XRD) using a BRUKER-D8 instrument (CuKα as a target and the incident wavelength $\lambda=1.5406$ Å radiation). The crystallite size D_s of the sample has been estimated from the full-width at half maximum (FWHM) of the strongest reflection of the plane (311) using the X-ray diffractometer software program, based on Scherrer equation at the Central Metallurgical Research and Development Institute (El-Tebbeen, Helwan, Egypt). By using the interplanar distances (d) calculated from Bragg's law, the lattice parameter (a) of all samples has been calculated by

$$a = d\sqrt{h^2 + k^2 + l^2}.$$

The X-ray density (theoretical density ρ_x) of the samples has been calculated according to the following equation:

$$\rho_x = \frac{8M}{Na^3}(\mathrm{g/cm^3})$$

where M is the molecular weight of the sample; N is Avogadro's number, a^3 is the volume of the lattice unit and 8 stands for the number of chemical formulas in a unit cell.

The experimental density (ρ_m) of the disk shaped samples has been determined by using the relation:

$$\rho_m = \frac{\text{mass}}{\text{volume}}\left(\mathrm{g/cm^3}\right)$$

where the mass has been determined by using a digital balance (OHAUS B100). The porosity (P) of those samples has been calculated using the following relation:

$$P = 1 - \frac{\rho_m}{\rho_x}$$

2.3. Transmission electron microscopy (TEM)

The particle size of the ferrite nanosample chosen for preparing the composites $[Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}Fe_{2.15}O_4]$ sintered at 1473k (i.e.1200 °C) has been recalculated by using JEOL JEM-100SX transmission electron microscope. To take the TEM images the powder sample has been dispersed in distilled water and has been exposed to ultra sonic waves for 15 min then a drop of the solution has been placed on a copper supported carbon grid and left to dry for a few minutes.

2.4. Preparation of pure epoxy sample and epoxy/$Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}Fe_{2.15}O_4$ composites

For preparing the pure epoxy sample, 1 g of pristine epoxy resin (diglycidyl ether of bisphenol A (DGEBA)) with an equivalent weight (EEW) of 170 g/mol, has been added to a proper amount of tetrahydrofurane (THF) to reduce the resin viscosity then 0.6 g of polyoxypropylene diamine (Jeffamine D-400) has been added too and all were placed in a glass dish treated by dichloro di-methylsilane. The sample has been heated in an evacuated oven at 353 K (i.e. 80 °C) for 2 h then at 393 K (i.e. 120 °C) for 3 h.

Four samples of pristine epoxy/$Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}Fe_{2.15}O_4$ composites have been prepared with different ferrite weight percentages (wt. %): (20%, 30%, 40%, and 50%).

2.5. The detailed method of preparing of the composites

For example to prepare pristine epoxy ferrite composites using 20% wt.% of ferrite, the following steps have been followed: 0.2 g of the prepared ferrite, 0.5 g of epoxy have been dispersed in THF (5 ml) using ultrasonic technique for 15 min, followed by mechanical stirring for 15 min until uniform dispersion has been achieved. The resulting mixture has been then mixed with the stoichiometric amount of Jeffamine (0.3 g). The sample has been heated in an evacuated oven at 353 K (i.e. 80 °C) for 2 h then at 393 K (i.e. 120 °C) for 3 h.

The four samples of pristine epoxy/$Li_{0.15}(Ni_{0.5}Zn_{0.5})_{0.7}Fe_{2.15}O_4$ composites and the pure epoxy have been cut into circular disk-shaped pellets for electrical measurements and toroids for magnetic measurements.

The pristine epoxy sample and the epoxy ferrite composites have been characterized by Fourier transform infrared (FTIR)