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Sol–gel auto combustion processed soft Z-type hexa nanoferrites for microwave antenna miniaturization

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Abstract

Nanoparticles of Z-type hexaferrite, $Ba_3Co_2Fe_{24}O_{41}$ and $Ba_3Co_{1.9-x-y}Cu_xZn_yNi_{0.1}Fe_{24}O_{41}$ (where $x=0.4$, 0.2 and $y=0.5$, 0.7) were prepared by sol–gel auto combustion method. Electromagnetic properties of the prepared nanoparticles were realized in the frequency range from 1.6 GHz to 4 GHz for the design of miniaturized microwave antennas. Optimized synthesis conditions and the addition of selected dopants have been used to enhance the high frequency electromagnetic properties of low loss Co_2Z nanoferrites. The best results obtained were $\varepsilon'=3.07$, $\mu'=2.59$ at frequency 2 GHz and $\varepsilon'=2.33$, $\mu'=2.94$ at frequency 2.5 GHz for the composition $Ba_3Co_1Cu_{0.2}Zn_{0.7}Ni_{0.1}Fe_{24}O_{41}$. This is a significant improvement w.r.t. our previously reported results. Electromagnetic results are explained on the basis of relevant existing theories and models. Owing to these electromagnetic properties, in particular the controlled electric and magnetic losses close to zero in the frequency range from 1.6 GHz to 4 GHz, these nanoferrites could be useful to the miniaturization of antennas for mobile handheld wireless communication devices.

Keywords: B. Electron microscopy; C. Magnetic properties; D. Ferrites; Cavity perturbation

1. Introduction

Wireless and mobile communication technology has become indispensable part of our lives. Increased demand for compact and high bandwidth system has accelerated the research and development of miniaturized built-in antennas. Ferrites possessing both magnetic and dielectric properties, with permeability ($\mu' > 1$) and nearly equal permittivity ($\varepsilon' > 1$) and loss tangent ($\tan\delta$) < 0.01 can be potential candidates for antenna miniaturization [1]. Also, the magneto-dielectric materials with almost equal μ' and ε' are important for matching antenna impedance to free space, enhance antenna and RF device performance in GHz regions and enable antenna miniaturization [2]. In literature, Co_2Z nano hexaferrites ($Ba_3Co_2Fe_{24}O_{41}$) with excellent electromagnetic properties have been reported as the best suitable magneto-dielectric materials that can fulfill the present demand [3]. High ferromagnetic resonance frequency up to GHz region, high thermal stability and high permeability make these nanoferrites promising candidates for commercial use in high frequency applications such as telecommunications, radar systems and other microwave devices [4,5]. To understand the behavior of magnetic materials in a particular frequency region, it is important to investigate the frequency dependent electromagnetic properties including complex permittivity and complex permeability of these magnetic materials. These parameters play an important role in identifying the suitability of materials for a particular application. So far, hardly any publication has reported the low loss (close to zero) electromagnetic properties of sol–gel auto combustion processed Z-type nano hexaferrite in GHz frequency range upto 4 GHz. This manuscript, therefore, report on the interesting electromagnetic properties of substituted Co_2Z nano hexaferrite in the frequency range from 1.6 GHz to 4 GHz for antenna miniaturization. Divalent ion dopants Cu^{2+},

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Zn^{2+} and Ni^{2+} were simultaneously incorporated into the structure of Co_2Z nano hexaferrite in order to obtain low loss magnetic nanoparticles with enhanced electromagnetic properties. Ni^{2+} ions concentration has been fixed as per literature reports [6]. In literature, the partial substitution of Co^{2+} ions with Cu^{2+} ions has been proposed to reduce the Z-phase formation temperature and sintering temperature of Co_2Z nano hexaferrite [7]. Also, Zn^{2+} ions substitution in Co_2Z nano hexaferrite has been reported to enhance the magnetic properties [8]. In the present work our focus is to highlight the interesting electromagnetic properties of substituted soft Z-type nano hexaferrite. Controlled magnetic and electric losses close to zero have been achieved over the frequency range 1.6 GHz to 4.0 GHz.

2. Experimental procedure

Nanoparticles of $Ba_3Co_2Fe_{24}O_{41}$ (Z) and $Ba_3Co_{1.9-x-y}Cu_xZn_yNi_{0.1}Fe_{24}O_{41}$ (where $x=0.4$, 0.2 and $y=0.5$, 0.7) labeled as Z1, Z2 were prepared via sol–gel auto combustion method [9]. The following raw materials of high purity were used: $Fe(NO_3)_3 \cdot 9H_2O$, $Ba(NO_3)_2$, $Co(NO_3)_2 \cdot 6H_2O$, $Zn(NO_3)_2 \cdot 6H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$, $Ni(NO_3)_2 \cdot 6H_2O$, anhydrous citric acid and ethylene glycol. According to the chemical composition all the nitrates were dissolved in 40 ml distilled water in proper stoichiometric proportions with stirring to get a clear solution. Then the aqueous solution of citric acid was mixed with the metal nitrate solution. The solution was allowed to mix with constant stirring for 30 min. The ammonia solution was then slowly added into the solution to adjust the pH at 7. The mixed solution was then evaporated on a hot plate at 80 °C with continuous stirring. Ethylene glycol was then added to the solution at 80 °C. The solution was slowly evaporated until a viscous gel was formed. With continuous heating at 150 °C for 4 h the viscous gel began bubbled up. As the temperature increased the gel started to swell with the evolution of a large amount of gases and was finally burnt with glowing flints. The brown colored fluffy powders so obtained were then calcinated in a furnace at 1200 °C for 5 h at a heating rate of 300 °C/h and was subsequently cooled with the same rate to room temperature. X-ray diffraction measurements were taken by an X-ray diffractometer (XPERT-PRO) in the range of $2\theta=20$–$80°$ using Cu$K\alpha$ radiation of wavelength 1.54 Å. The microstructures of the samples were examined using a scanning electron microscope (SEM: QUANTA 250 FFID 9393). The M–H measurements at room temperature have been carried out using Vibrating Sample Magnetometer of Microsense. Microwave properties of the prepared nanoparticles were investigated by resonance based cavity perturbation method using ZVA 50 vector network analyzer as explained in our previous studies [10,11]. The complex permittivity and complex permeability of the sample can be calculated from the variation of resonant frequency and quality factor of the metal cavity.

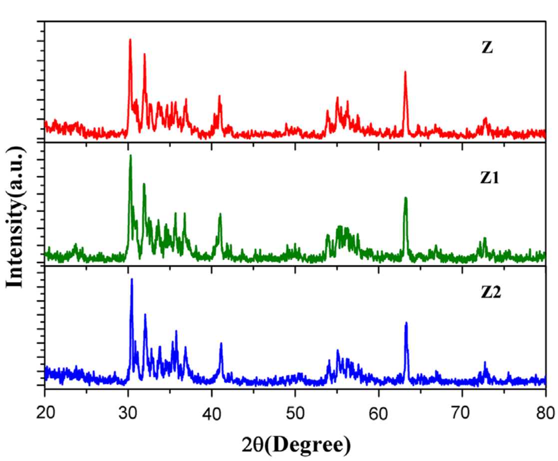

Fig. 1. XRD patterns of Co_2Z-nano hexaferrites.

3. Results and discussion

3.1. Structural study

The typical X-ray diffraction patterns of $Ba_3Co_2Fe_{24}O_{41}$ and $Ba_3Co_{1.9-x-y}Cu_xZn_yNi_{0.1}Fe_{24}O_{41}$($x$=0.4, 0.2 and y=0.5, 0.7) samples leveled as Z, Z1and Z2 are shown in Fig. 1. The peaks clearly indicate the formation of pure Z-phase according to JCPDS file 19-97 [12] and there was no evidence of the appreciable secondary phase. The broadening of diffraction lines is due to the small crystalline size of the prepared samples. The average crystallite size was determined from the XRD patterns of the samples according to the Scherrer formula

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (1)$$

where D is the crystallite diameter, β is the full width at half maximum (FWHM), θ is the Bragg's angle of diffraction in degrees, k is the shape factor (taken as 0.9) and λ is the X-ray wavelength (1.54 Å). The average particle size calculated from XRD for Co_2Z nano hexaferrites was found to be in the range of 30–45 nm. The SEM micrographs shown in Fig. 2(a, b and c) are the surface images of Z, Z1 and Z2 nano hexaferrites calcinated at 1200 °C for 5 h. All the samples show a well-densified microstructure of the prepared nano hexaferrites. The grains are uniformly and densely distributed over the surface of the samples.

3.2. Magnetic study

Fig. 3 shows the magnetic hysteresis loops of the prepared nanoparticles measured at room temperature with a maximum field upto 20 kOe. The hysteresis curves of all the prepared samples exhibit typical characteristics of soft ferrites with low coercivity. The saturation magnetization M_s was determined by extrapolating the M versus $1/H$ curves to $1/H=0$. The resulting M_s values for the prepared samples labeled as Z, Z1 and Z2 are 44.84 emu/g, 44.65 emu/g and 50.43 emu/g, respectively. The substitution of Co^{2+} ions by Zn^{2+} ions results in an increase of M_s due to the distribution of Zn^{2+} ions among the tetrahedral A-sites of the structure which enhances net molecular magnetic moment of the crystal. According to Neel's Model [13], in A–B exchange interaction magnetic moments on A-site are antiparallel to the magnetic moments on B-site and the net magnetic