Journal of ELECTRONIC MATERIALS, Vol. 46, No. 2, 2017
DOI: 10.1007/s11664-016-4883-9

Magnetic and High-Frequency Dielectric Parameters of Divalent Ion-Substituted W-Type Hexagonal Ferrites

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Polycrystalline W-type hexagonal ferrites with chemical formulae $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (x = 0, 0.5, Me = Mn, Mg, Zn, Ni) have been prepared using sol–gel autocombustion. It has been reported in our earlier published work that all the samples exhibit a single-phase W-type hexagonal structure which was confirmed by x-ray diffraction (XRD) analysis. The values of bulk density lie in the range of 4.64–4.78 g/cm^3 for all the samples which are quite high as compared to those for other types of hexaferrites. It was also observed that Zn-substituted ferrite reflects the highest (14.7×10^7 Ω-cm) whereas Mn-substituted ferrite has the lowest (11.3×10^7 Ω-cm) values of direct current (DC) electrical resistivity. The observed values of saturation magnetization (M_s) are found to be in the range of 62.01–68.7 emu/g depending upon the type of cation substitution into the hexagonal lattice. All the samples exhibit a typical soft magnetic character with low values of coercivity (H_c) that are in the range of 26–85 Oe. These ferrites may be promising materials for microwave absorbers due to their higher saturation magnetization and low coercivities. Both the dielectric constant and tangent loss decrease with increasing frequency in the lower frequency region and become constant in the higher frequency region. The much lower dielectric constant obtained in this study makes the investigated ferrites very useful for high-frequency applications, i.e. dielectric resonators and for camouflaging military targets such as ships, tanks and aircrafts, etc.

Key words: W-type hexaferrites, divalent ion substitution, x-ray diffraction, M-H loops, dielectric parameters

INTRODUCTION

With the end of World War II, there came a great revival of interest in magnetism and the study thereof became more focused than it was previously. This renewed interest came mainly from the development of an entirely new class of magnetic materials, the ferrites that were developed, explained, and subsequently used for a variety of applications. These applications include permanent magnets, electrical and microwave devices, core materials, data storage and recording, plastoferrites, radar absorbing materials (RAM), microwave absorption, and magnetoelectric (ME) and multiferroic (MF) applications.[1] Among all ferrites, hexagonal ferrites exhibit a wide range of applications in various electronic devices such as electric power generators, automotive electronics, micro electromechanical systems (MEMS), rotors in small DC motors,

(Received December 22, 2015; accepted August 13, 2016;

microwave devices and ferrite cores.[2,3] The soft ferrites are mainly used as cores in special transformers or inductors for high-frequency applications. Some communication and electronic instruments have a need for broadband transformers having cores which demonstrate a similar behavior over a wide range of frequencies. In addition, soft ferrites are used in pulse transformers because the square pulse's Fourier components expand through broadband frequencies. Moreover, whenever the permeability of the core changes with the frequency, intense bending of the pulse shape takes place. Modern radio receivers are comprised of built-in ferrite antennas which consist of an uncomplicated assembly where a coil is wound around a ferrite rod. The antenna is responsive to both the electric and magnetic components of the incident electromagnetic waves, and by Faraday's law, the alternating flux in the ferrite rod induces an electromagnetic field (emf) in the coil. As a result, the ferrite core increases the permeability time of the area enclosed by the coil. Therefore, ferrites have significant microwave applications such as in communications and in radar circuits with frequencies of up to 10^{10} Hz.[4] X-, Z-, Y-, and W-type hexagonal ferrites belong to a well-known family of soft magnetic ferrites which might be used for operation in high-frequency applications (up to several GHz). It has been reported in many publications that good magnetic efficiency and performance in the GHz region and higher resonance frequencies are revealed by the above-mentioned hexagonal ferrites with planar magneto-crystalline anisotropy, as compared to spinel ferrites.[5,6]

Cu-substituted W-type hexaferrites ($BaCo_{2-x}Cu_xFe_{16}O_{27}$ with $x = 0.4$, 0.6, 0.8, 1 and 1.2) were prepared by the standard ceramic technique.[7] The dielectric constant and the dielectric loss tangent were measured and the results showed that the dielectric constant for different contents was found to be in the range of 20–500 which is very high as compared to those observed in our work. Its value goes on increasing with the increase of temperature. This behavior could be discussed on the basis of the assumption that the mechanism of dielectric polarization is similar to the conduction.[8] In another study, results[9] show that the dielectric constant increases as sintering temperature increases–, whereas the dielectric constant and loss tangent decrease as frequency increases and as temperature decreases. These effects are attributed to the Maxwell–Wagner double-layer polarization. As the Mn substitution increased, samples showed abnormal dielectric behavior in the form of relaxation peaks which shifted towards higher frequencies; the dielectric constant and loss tangent also decrease gradually, reaching a minimum between $x = 0.02$ and 0.06.

The polycrystalline samples of the composition $Zn_{2-x}Co_xBaFe_{16}O_{27}$ were prepared by the usual ceramic technique with $x = 0.4$, 0.6, 0.8, 1, 1.2, 1.4 and 1.6.[10] The samples were sintered at 1300°C for 8 h. Besides other measurements, M-H loops for different substituted samples of a W-type hexagonal nature were measured as a function of different concentrations (x). It was observed that the saturation magnetization of all the samples was found to be in the range of 38–45 emu/g which is much lower as compared to our samples.

Furthermore, it has been observed that the processing and composition of ferrites are equally important for developing suitable absorbers. Microwave absorber material is an exceptional feature of W-type hexagonal ferrites because of its soft magnetic property.[11] In recent years, many researchers have attempted to alter the electrical, structural and magnetic properties of W-type hexaferrites by substituting their divalent ions, trivalent ions and binary mixture. W-type hexaferrites can be synthesized by a variety of techniques. Via the sol–gel technique, submicron particles at low temperature can be prepared. In addition, a very suitable method was recently found by which multi-component oxide materials can be prepared at relatively lower ranges of temperatures. The major advantages of the method are to reduce the product formation temperature with good control over the homogeneity and microstructures.

In this paper, we report the results of a systematic variation of the different divalent ions in $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ ($x = 0$, 0.5, Me = Mn, Mg, Zn, Ni) ferrites synthesized by a sol–gel autocombustion method. The effects of different cation substitutions in W-type hexagonal ferrites on the structural and magnetic behavior as well on the dielectric properties are discussed. Moreover, this work is a sequence of our already reported results[12] where the effects of different divalent ions on the structure, magnetic and microwave absorption parameters were discussed.

EXPERIMENTAL

W-type hexagonal ferrites with composition $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ ($x = 0$, 0.5, Me = Mn, Mg, Zn, Ni) have been prepared using the sol–gel autocombustion method. Aqueous solutions of nitrates of different metals with % purity described elsewhere[12] were used without any treatment. Citric acid (99.91%, Fisher Scientific, UK) was used as a chelating agent. During the synthesis, the aqueous solutions of the metallic salts and citric acid according to stoichiometric ratios were slowly added together in 100 ml of water. Then resultant sol was transformed into gel by using a hot plate magnetic stirrer, while applying constant heat at 80°C. Till the formation of gel the pH value of the sol was maintained at approximately 7–8 by using ammonia (35%, Fisher Scientific, UK). After 6–7 h, powdered samples were prepared by auto combustion of dried gel with an increase of the temperature up to 200°C while being on the hot plate. Finely ground powders were pressed into pellets with a Paul Otto Webber hydraulic press and 35 kN force was applied for about 1.30 min. Finally, single-phase W-type hexaferrites were obtained by simultaneously sintering the powders and pellets in a temperature-controlled furnace (Nabortherm, Germany) at a temperature of 1300°C for 5 h with a heating rate of about 3°C/min. X-ray diffraction (XRD) was used for structural analysis of powdered samples (with a scan step of 0.02°/5 s and at room temperature) equipped with a Co-K$_\alpha$ radiation source (λ = 1.79 Å) at 35 mA and 30 kV. An energy-dispersive x-ray spectroscope (EDXS, Model: FEI Quanta 200F) was used for examining the elemental composition of all the samples. For this experimental purpose, the powder sample was sputtered with a thin layer (4 nm) of Au–Pd (60:40) to discharge the negative charge present in the microscope. The magnetic measurements (M-H loops) for all the samples were taken using a commercial physical property measurement system (PPMS) Q6000 equipped with a vibrating sample magnetometer (VSM) from Quantum Design. The PPMS consists of two major components: a liquid helium dewar with a 9*T* (90000 Oe) longitudinal superconducting magnet and a temperature controller. The microwave measurements were carried out at room temperature for pellets of 7-mm radius and 2-mm thickness by an impedance analyzer (Model E4991A RF) in a frequency range of 0.5–1.5 GHz.

The dielectric constant for all the samples was calculated using the following formula;

$$\varepsilon = C_p d/\varepsilon_o A \tag{1}$$

where C_p is the capacitance of the pellet in farad units, d is the thickness of the pellet in meters, A is the cross-sectional area of the flat surface of the pellet and ε_o is the permittivity of free space.

The dielectric tangent loss factor (tan δ) was calculated using the following formula;

$$\tan\delta = 1/2\pi f R_p C_p \tag{2}$$

where δ is the loss angle, f is the frequency, R_p is the equivalent parallel resistance and C_p is the equivalent parallel capacitance of the sample.

RESULTS AND DISCUSSION

Phase Determination

Single-phase samples are necessary for a particular application and play a major role for better performance of a device. Therefore, it is necessary to eliminate the impurities from the investigated sample and to get a single-phase sample after employing different synthesis conditions. In order to confirm whether the samples are single-phase W-type hexagonal ferrites or contain some other impurities, XRD measurements were carried out for all the samples and the results are reported in our previously published manuscript.[12] It was reported there that the diffraction peaks for all the samples greatly match with the standard patterns for a W-type hexagonal structure and no sample contains an impurity phase which confirms that all the samples are single-phase. In addition, variation of cell parameters such as lattice parameters (*a, c and c/a* ratios) and cell volume (*V*) was described there[12] as a function of different divalent ions substituted into the hexagonal lattice of W-type hexagonal ferrites.

Physical Properties

The corresponding values of bulk density (d_B), x-ray density (d_x) and porosity (P) as a function of divalent ions are given in Table I. It is obvious that the value of x-ray density is highest for Zn-substituted hexaferrite whereas that for Mn-substituted hexaferrite is lowest because the atomic weight of Zn (65.38) is higher and that of Mn (24.3) is lower than Co ions (58.9). The values of bulk density lie in the range of 4.64 g/cm^3–4.78 g/cm^3 for all samples which is quite high as compared to other hexaferrites of similar structure.[13] Usually, a higher sintering temperature is known to increase the density of ferrite samples. In general, higher-density ferrite samples are known to increase permeability and, hence, microwave absorption.[14] Moreover, the values of bulk density for all the samples are slightly lower than those for x-ray density due to the presence of pores in the material created during sintering. The (%) porosity of all the samples was also calculated; the values are given in Table I. It is

Table I. Effects of different divalent ion substitution on bulk density (d_B), x-ray density (d_x), porosity (P), resistivity (ρ), charge carrier concentration (n) and mobility (μ_d) for $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (Me = 0, Mn, Mg, Zn, Ni) hexaferrites

Me	$d_B \pm 0.01$ (g/cm^3)	d_x (g/cm^3)	P (%)	$\rho \times 10^7 \pm 0.02$ (Ω-cm)	$n \times 10^{22}$ (cm^{-3})	$\mu_d \times 10^{-12}$ (cm^2/V s)
0	4.743	5.282	10.2	12.2	2.92	1.75
Mn	4.778	5.221	8.48	11.3	2.96	1.87
Mg	4.696	5.281	11.08	13.7	2.93	1.56
Zn	4.649	5.428	14.35	14.7	2.87	1.48
Ni	4.706	5.420	13.17	12.9	2.90	1.67

evident that the calculated porosity lies in the range of 8–14% which is quite low due to higher values of bulk density as discussed above.

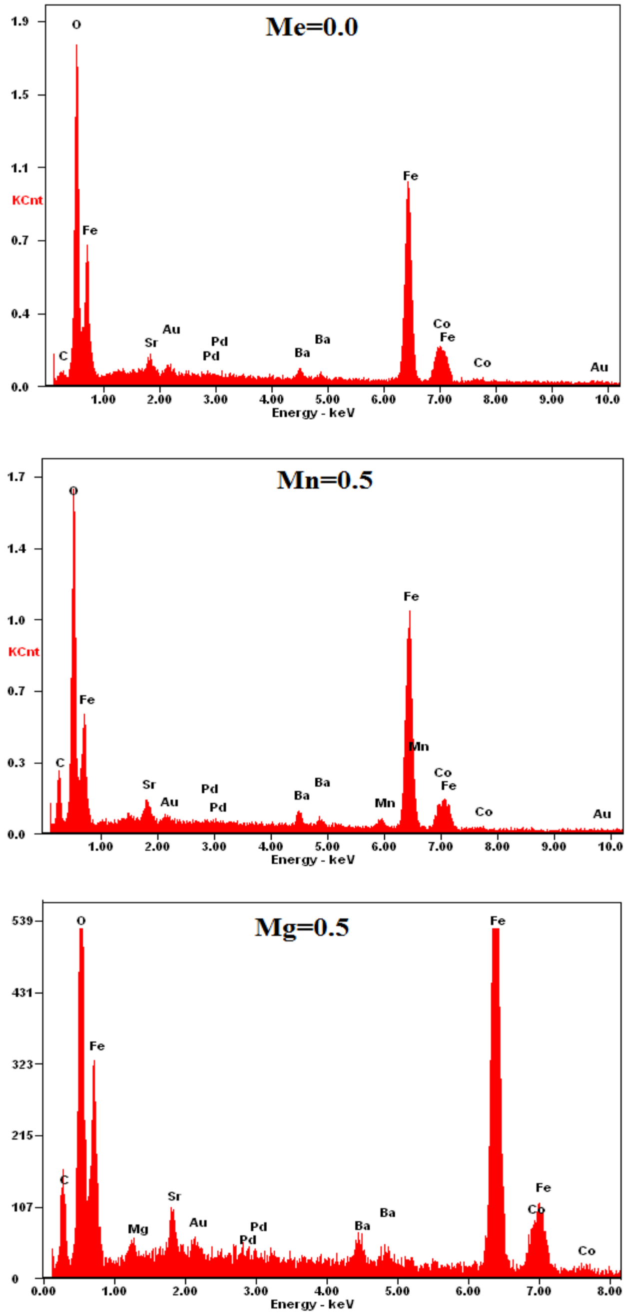

Fig. 1. EDX spectra for some selected $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (x = 0.5) hexaferrite samples.

EDX Analysis

The elemental composition of the investigated hexaferrites in the present study is calculated by using energy-dispersive x-ray spectroscopy (EDX) because this technique is employed to confirm the presence of dissolved constituent elements in the samples. Figure 1 shows the EDX spectra for some selected samples of the investigated hexaferrite series. The calculated values of the constituent elements of different samples are in close conformity with their nominal compositions which confirms that all substituted divalent ions replace cobalt ions in the hexagonal lattice. Furthermore, the EDX spectra for all the samples confirm that no impurity elements other than dissolved reactants are present in the samples, which is further confirmation of our claim that the samples are single-phase. The calculated elemental compositions of all the investigated samples are summarized in Table II.

Electrical Measurements

The room-temperature DC electrical resistivity was measured by a two-probe method because of the high values of resistivity of the investigated ferrites, and the values are given in Table I as a function of substituted divalent ions. The value of resistivity lies in the range of (11.3×10^7–14.7×10^7) Ω-cm for all the samples. It is also clear that Zn-substituted ferrite reflects the highest, whereas Mn-substituted ferrite has the lowest values of resistivity. It is well known that porosity has a significant effect on the electrical resistivity of ferrite materials and both are directly related to each other. Zn-substituted samples have a large number of pores which, in turn, hinders the motion of charge carriers from one site to the other, resulting in an increase of the electrical resistivity.[15] This makes these materials suitable for microwave devices and high-frequency applications as highly resistive materials are the requirements of these applications.[16] Similarly, an Mn-substituted sample has the lowest value of

Table II. Elemental composition of $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (Me = 0.0, 0.5) as determined by EDXS analysis

	Values determined from EDXS analysis				
Sample (theoretical values)	**Ba (mol)**	**Sr (mol)**	**Co (mol)**	**Me (mol)**	**Fe (mol)**
$Ba_{0.5}Sr_{0.5}Co_2Fe_{16}O_{27}$	0.48	0.47	1.48	–	15.94
$Ba_{0.5}Sr_{0.5}Co_{1.5}Mn_{0.5}Fe_{16}O_{27}$	0.47	0.49	1.49	0.49	15.98
$Ba_{0.5}Sr_{0.5}Co_{1.5}Mg_{0.5}Fe_{16}O_{27}$	0.51	0.48	1.51	0.48	15.96
$Ba_{0.5}Sr_{0.5}Co_{1.5}Zn_{0.5}Fe_{16}O_{27}$	0.50	0.46	1.49	0.51	15.99
$Ba_{0.5}Sr_{0.5}Co_{1.5}Ni_{0.5}Fe_{16}O_{27}$	0.49	0.51	1.48	0.49	15.93

electrical resistivity because there will be a small number of pores to hinder the motion of charge carriers. Table I also exhibits the variation of mobility for all the samples and shows inverse behavior contrary to electrical resistivity as both parameters are inversely related to each other.

Magnetic Measurements

The 'S'-shaped M-H loops for all $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (Me = 0, 0.5 and Mn, Mg, Zn, Ni) hexaferrite samples are shown in Fig. 2 and the values of coercivity (H_c) and saturation magnetization (M_s) obtained from these loops are listed in Table III. It is obvious that all the samples exhibit a typical soft magnetic character with low coercivity in the range of 26–85 Oe. The coercivity H_c slightly varies after different divalent ions are substituted for Co^{2+} , and, also, the Zn-substituted sample reflects the lowest value of coercivity as compared to other substituted samples. This behavior can be ascribed on the basis of the aspect ratio (a/c)[12] and, in that case, the coercivity could be expressed as follows;[17]

$$H_c = 0.48 \times \left[\frac{K_1}{M_s} - N_d M_s\right] \tag{3}$$

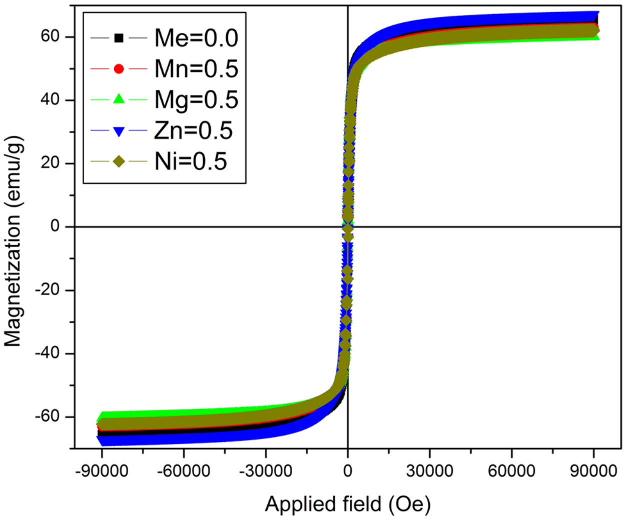

Fig. 2. M-H loops for all substituted $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ hexaferrite samples.

where C is a constant, K_1 is the magnetocrystalline anisotropy constant, M_s is the saturation magnetization and N_d is the demagnetizing coefficient that relates to the shape anisotropy of hexaferrites. The aspect ratio could increase the demagnetizing factor and thus decrease H_c and vice versa.[18] It can be observed that a Zn-substituted sample has the highest aspect ratio value which results in the lowest value of coercivity. The lowest value of coercivity is favorable for EM materials, to enhance the microwave absorption and is in good agreement to the requirement of W-type hexagonal ferrites[19] which behave like soft ferrites. This is because coercivity is much smaller as compared to a well-known hard magnet M-type barium hexaferrite, with a value of coercivity in the range of a few thousand oersteds (Oe) to 6700 Oe. The observed magnetic parameters for these samples are not encouraging for magnetic recording applications, but are more promising for microwave absorption and high-frequency applications due to their higher saturation magnetization, anisotropy and low coercivities.

The saturation magnetization (M_s) obtained from M-H loops for all the samples was also calculated by the law of approach to saturation[20] and depicted in Fig. 3 as a function of divalent ions. Saturation magnetization was found to be highest (68.7 emu/g) for the Zn-substituted sample, and lowest for Mg-substituted hexaferrite (Table III). This variation of M_s for substituted ferrite can be explained on the basis of site occupancy of the substituted ions. Zn^{2+} ions show a marked preference for tetrahedral A-sites, owing to the presence of (non-magnetic) Zn ions, the magnetization of the tetrahedral lattice will be smaller as compared to pure ferrite, and since Co^{2+} ions have the larger moment ($3\mu_B$), the M_s for a Zn-substituted ferrite would be expected to increase with increasing Zn contents in the hexagonal lattice. This confirms the remarkable fact that substitution of magnetic ions by non-magnetic ions in a ferrimagnetic material is known to cause an increase in the saturation magnetization.[21] Dimri et al.[22] reported that Mg^{2+} -substituted ferrites exhibit the lowest values of saturation magnetization. It may be expected that substitution of non-magnetic ions (even at the smallest concentration) on the octahedral sites of the ferrites causes a

Table III. Effects of different divalent ion substitution on coercivity (H_c), saturation magnetization (M_s) and magnetic moment (n_B) for all $Ba_{0.5}Sr_{0.5}Co_{2-x}Me_xFe_{16}O_{27}$ (Me = 0.0, Mn, Mg, Zn, Ni) hexaferrites

Me	H_c (Oe)	M_s (emu/g)	n_B (μ_B)
0	58.09	67.47	18.80
Mn	72.53	64.64	17.98
Mg	50.90	62.01	17.08
Zn	26.99	68.70	19.18
Ni	84.90	64.01	17.83

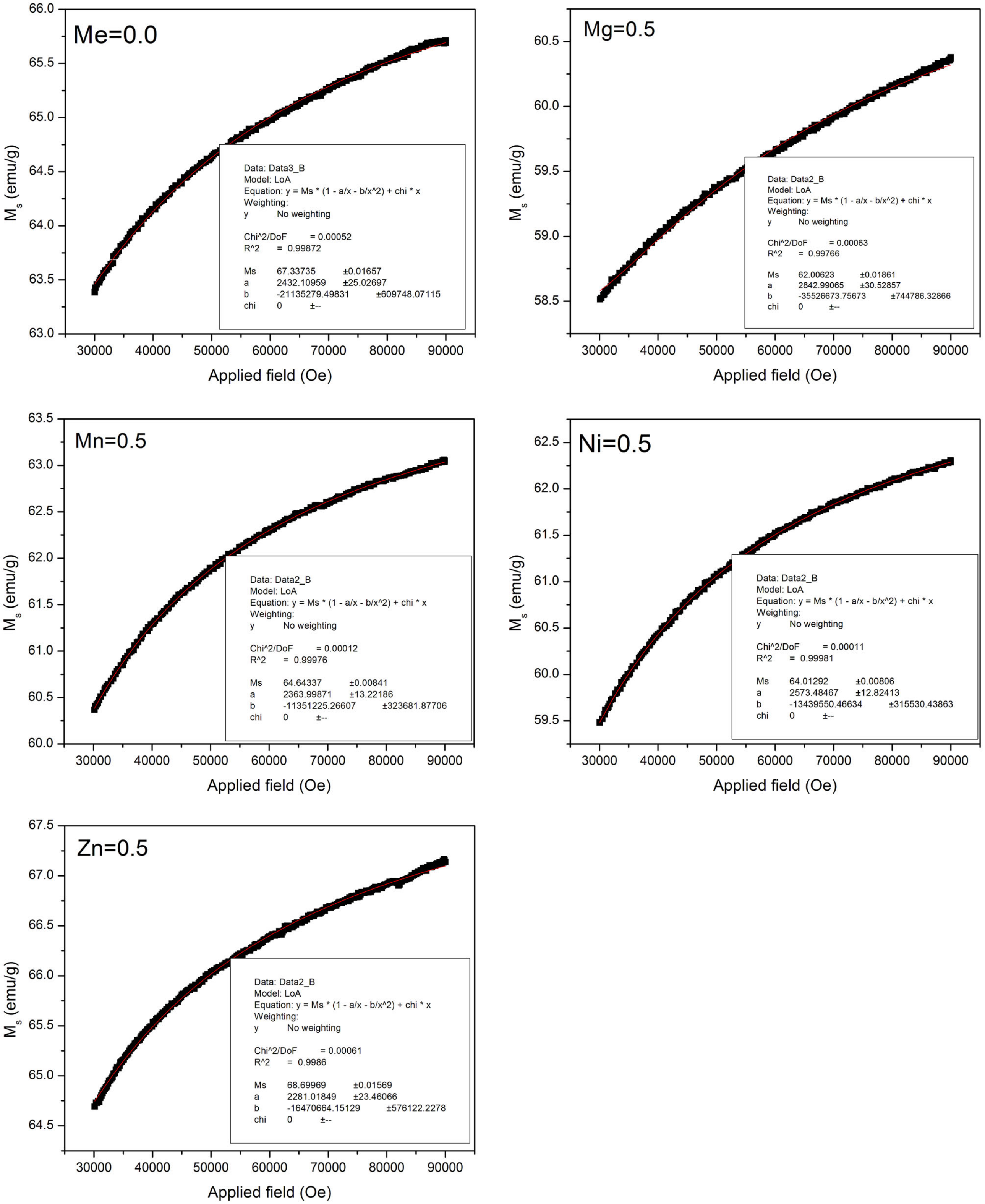

Fig. 3. Fitted curves for M_s calculated by the law of approach to saturation for all the samples.

decrease in the saturation magnetization.[21] The results show that Ni substitution causes a small reduction in the saturation magnetization of Co^{2} hexaferrites. This is because both Ni^{2+} and Co^{2+} ions prefer to occupy the octahedral B-site and have a greater preference energy for these sites than Fe^{3+}. Therefore, Ni^{2+} and Co^{2+} occupy octahedral sites where the spin direction is the same as the whole magnetic moment.[21] In addition, as Ni^{2+} has a smaller value of magnetic moment if compared with Co^{2+}, the substitution of Ni for Co causes a decrease in the saturation magnetization. Similar behavior is observed when Mn ions are substituted in a hexagonal lattice. The magnetic moment for each sample was also calculated by using the following formula[21] and the values are in good agreement with the values of M_s (Table III) for all the samples.

$$n_B(\mu_B) = \frac{M \times M_s}{5585}. \quad (4)$$

Dielectric Measurements

Dielectric constant is also known as relative permittivity and is the factor by which the electric field between the charges is decreased relative to vacuum.[23]

The loss of power in a dielectric caused by the loss of energy in the form of heat generated by an electric field is known as dielectric loss.[24] Dielectric loss quantifies a dielectric material's inherent dissipation of electromagnetic energy (e.g. heat).

The variation of the dielectric constant and the dielectric loss factor (tan δ) as a function of frequency are depicted in Figs. 4 and 5, respectively. Both dielectric constant and tan δ decrease with increasing frequency in the low-frequency region and become constant in the high-frequency region. This is a normal behavior for ferrites. The Maxwell–Wagner model explains it well for interfacial polarization.[25] This model predicts the dielectric structure of a ferrite material which is supposed to be made up of two layers. The first layer consisting of a larger number of conducting grains is separated by the second layer which consists of highly insulating grain boundaries. Two mechanisms, polarization and conduction, in ferrites are similar which is due to the electron hopping between Fe^{2+} and Fe^{3+} ions. The polarization is inversely proportional to the frequency as it decreases with increasing frequency. With the increase in frequency of an externally applied electric field the electron hopping between Fe^{3+} and Fe^{2+} ions lags behind and cannot follow the alternating field. The higher values of dielectric constant at low frequencies are due to the voids, dislocations, piling up and other defects present in the samples.[26] It has been reported that a higher dielectric constant causes a decrease of the penetration depth of the electromagnetic waves by lifting up the skin effect. The aim of the present study is to make the investigated ferrites suitable for high-frequency applications by lowering the values of the dielectric constant. Hence, lower dielectric constants of the investigated ferrites deserve their applications at high frequencies. In addition, dielectric tangent loss is a significant part of the total core loss in ferrites which corresponds to the dissipation of energy in the dielectric medium. Hence, low dielectric losses are required for low core losses.[27]

The dielectric loss was determined using Eq. 2. Dielectric tangent loss is a significant part of the total core loss in ferrites.[28] Hence, low dielectric losses are required for low core losses. The dielectric loss as a function of frequency for all the compositions is depicted in Fig. 5. The behavior of dielectric

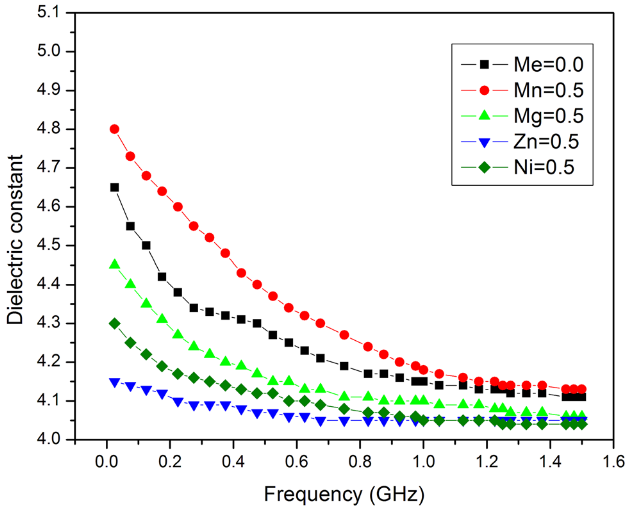

Fig. 4. Dielectric constant as a function of frequency for all substituted hexafferites.

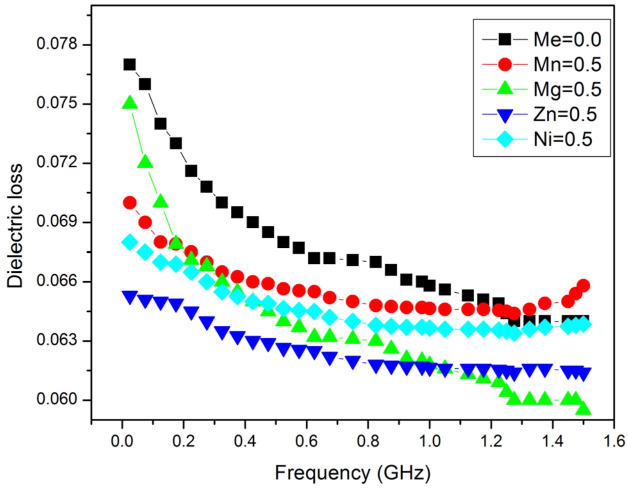

Fig. 5. Dielectric loss as a function of frequency for all substituted hexaferrites.

loss is similar to the real part of the dielectric constant as a function of frequency. An increase in electric polarization is caused by the increase in hopping electrons in a local displacement in the direction of the extent electric field which thus enhances dielectric loss. Hudson[29] has reported that the dielectric loss in ferrite is generally related to the conductivity and the material with higher conductivity exhibits high losses and vice versa.

CONCLUSIONS

Single-phase W-type hexagonal ferrites with substitution of different divalent ions were successfully synthesized by sol–gel auto combustion. The EDX spectra for all the samples show that the calculated values of the constituent elements of different samples are in close conformity with their nominal compositions which confirms that all substituted samples have no impurity elements. The value of resistivity lies in the range of (11.3×10^7–14.7×10^7) Ω-cm for all the samples and Zn-substituted sample exhibits the highest value which is due to this sample having a larger number of pores as described in physical measurements. Saturation magnetization was found to be highest (68.7 emu/g) for Zn-substituted sample, and lowest for Mg-substituted hexaferrite (62.01 emu/g). Moreover, the Zn-substituted sample reflects the lowest value of coercivity as compared to other substituted samples which can be ascribed on the basis of the aspect ratio (a/c). Both dielectric constant and tan δ decrease with increasing frequency in the low-frequency region and become constant in the high-frequency region and this behavior is in accordance with the Maxwell–Wagner model for interfacial polarization. The highest values of electrical resistivity and saturation magnetization and lowest values of coercivity, dielectric constant and dielectric loss for Zn-substituted samples obtained in this study make the investigated ferrites very useful for high-frequency applications where low eddy current losses are desirable.

ACKNOWLEDGEMENT

One of the authors (Mukhtar Ahmad) greatly acknowledges the financial assistance from the Higher Education Commission (HEC) of Pakistan under the Grant No. 21-686/SRGP/R&D/HEC/2014.

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