Electromagnetic Properties of Co-Zr Substituted Ba-Sr Ferrite-Paraffin Wax Composite for EMC/EMI Applications

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Abstract

The electromagnetic parameters of synthesized M-type hexagonal ferrite composites, $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ (x=0.0, 0.2, 0.4, 0.6, 0.8, 1.0), are measured from 0.1-18 GHz. The ferrite composites exhibit dielectric behavior contrary to conventional magnetic nature of ferrite composites. The electron hopping between Fe^{2+} ions and Fe^{3+} ions enhances dielectric properties making it useful for EMI/EMC applications. The relationship is established between microstructure and microwave properties. The damped domain wall dispersion is observed along low frequency region. The various mechanisms behind variations of investigated parameters are discussed.

Introduction

Electromagnetic pollution poses serious problem due to rapid development in wireless communication. Ferrites are being incorporated as shielding/absorbing materials to circumvent electromagnetic interference (EMI) and electromagnetic compatibility (EMC) problem at microwave frequencies. The pre-requisite for this is the attenuation of microwave signal depending upon dielectric and magnetic losses. Domain wall resonance in hexagonal ferrites is present towards lower end of microwave frequency spectrum and ferromagnetic resonance (FMR) at higher microwave frequencies. However FMR peaks are suppressed and permeability is reduced at microwave frequencies [1-2]. The other option to enhance microwave attenuation is to increase dielectric losses through suitable substitution and sintering temperature.

Experimental

The ferrite powders, $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$, (x=0.0, 0.2, 0.4, 0.6, 0.8, 1.0) were synthesized by standard ceramic method and composites were produced from these ferrite powders by mixing them with melted paraffin wax. The volume fraction of the powders in composites was about 20%, with isotropic allocation of powder particles. Standard 7/3 coaxial line was used for the measurements. The complex permittivity and permeability of sample were determined using VNA HP 8720 from 0.1-18 GHz with measured reflection coefficients of the sample with two loads, a short circuit and line of known length with a short circuit. Nicholson-Ross method was adopted to calculate Complex permittivity and permeability [3].

Results and Discussion

The microstructure in figure 1 exhibits grain distribution in undoped and doped sample. The undoped sample 0.0 consists of non-magnetic voids as porosity and inter-grain connectivity is poor, causing the hindrance to the applied field. The substitution, in sample 0.4 and 0.8, causes in increase in grain size which further decreases porosity. Thus substitution enhances inter-grain connectivity which eases the applied field flow. Therefore substitution causes increase in density and decrease in porosity.

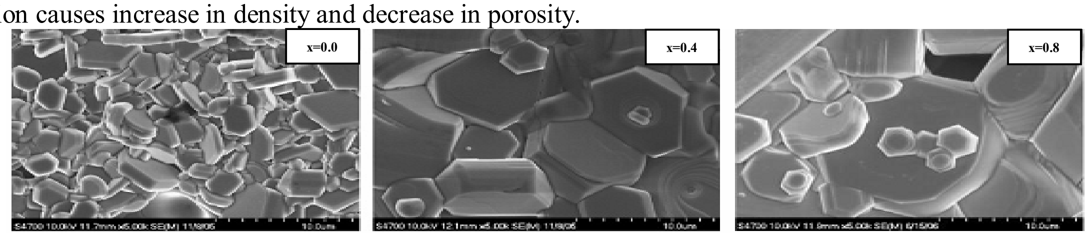

Fig. 1. Microstructure of ferrite samples (a) $Ba_{0.5}Sr_{0.5}Fe_{12}O_{19}$ (b) $Ba_{0.5}Sr_{0.5}Co_{0.4}Zr_{0.4}Fe_{11.2}O_{19}$ (c) $Ba_{0.5}Sr_{0.5}Co_{0.8}Zr_{0.8}Fe_{10.4}O_{19}$

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The dielectric constant in composites depends on space charge polarization and charge formation at grain boundary and additional interface, introduced by dielectric matrix. Composite materials, in which magnetic particles are coated with dielectric layers, introduce additional interfaces leading to interfacial polarization and this forms more polarization charges on the surface of the particles. This causes dielectric relaxation behavior more complex. It has been reported in literature [4, 5] that the properties of interfaces could have a dominant role in determining dielectric performance. The dielectric constant and dielectric loss (Figure 2 and 3) of all samples exhibits non-monotonic variation with substitution of Co^{2+} and Zr^{4+} ions. Table 1 shows non-linear variation of resistivity with substitution: It reflects two maxima at lower (sample 0.2) and higher substitution (sample 1.0) and high resistivity discourages polarization and conductivity. Thus both real and imaginary permittivity values are low in doped samples 0.2 and 1.0 while comparatively higher in other doped samples. All samples exhibit large value of permittivity and dielectric loss in spite of only 20 % volume fraction of ferrite. . It has been reported elsewhere that electron hopping between Fe^{2+} ions and Fe^{3+} ions leads to conductivity and dielectric polarization in ferrites [6].

Table 1. Resistivity variation of $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ (x=0.0-1.0)

x	0.0	0.2	0.4	0.6	0.8	1.0
Log(ρ)Ω.cm)	4.5	6.4	4.1	3.5	3.9	5.1

Researchers have reported formation of Fe^{2+} ions from Fe^{3+} ions at high temperature synthesis of ferrites [7-11]. The charge carriers are excited through thermal energy at high temperature and follow the changes in applied field, enhancing dielectric properties through polarization [12]. Thus large value of ε′ and ε″ in all samples, with just 20 % volume of ferrite, is due to formation of Fe^{2+} ions from Fe^{3+} ions at sintering temperature of 1250ºC. The real and imaginary permittivity of sample 0.0 undergoes pronounced dispersion and becomes weak with the substitution of Co and Zr ions. Sample 0.0 has maximum number of Fe^{3+} ions than doped samples, so large number of Fe^{2+} ions are produced from Fe^{3+} ions. This causes polarization enhancement and increase of both ε′ and ε″ in sample 0.0 along low frequency region. According to the Kramers-Kronig relations [13-14], larger ε″ corresponds to stronger frequency dispersion of the real part which is another reason for large ε′ in sample 0.0. The dielectric constant decreases with increase in frequency in all samples (except sample 0.2.) at low frequency region. This behavior is in agreement with Koops theory [15] based on Maxwell-Wagner model for non-homogenous dual layer dielectric structure. Thus layer of poor conducting grains is effective at low frequencies whilst good conductivity grains are effective at high frequencies. Therefore ε′ is higher at low frequencies and the fall of ε′ at higher frequencies is ascribed to the fact electronic exchange or hopping between Fe^{2+} and Fe^{3+} ions can not follow applied field.

The dielectric loss mechanism in ferrites depends on DC and AC conductivity or ion jumping and dipole relaxation according to

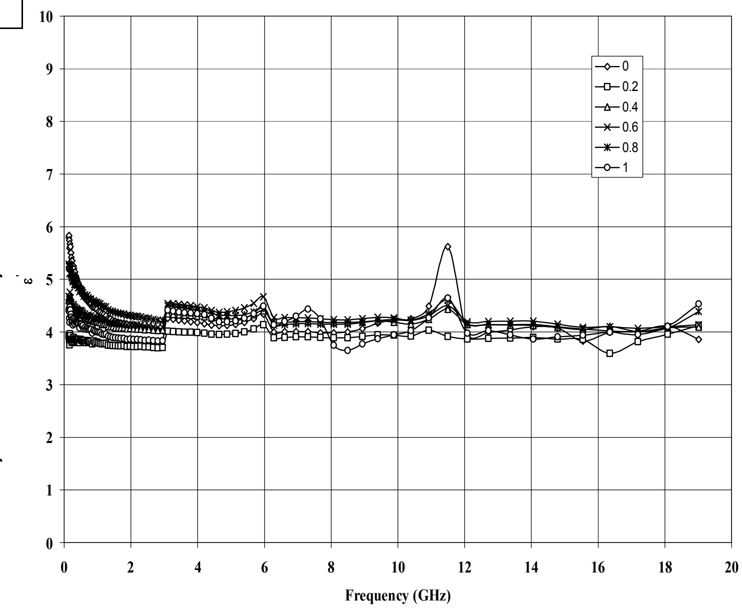

Figure 2. Dielectric Constant variation of $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ ferrite as a function of frequency and substitution (x=0.0-1.0).

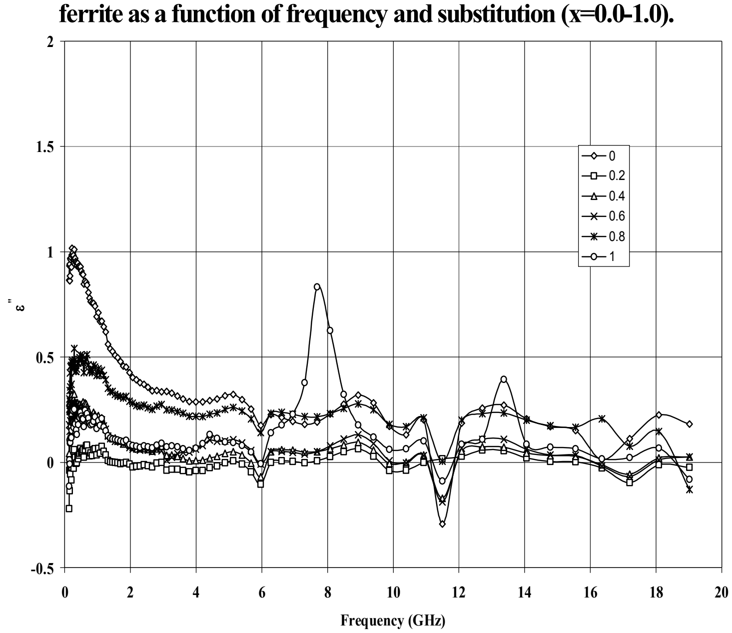

Figure 3. Dielectric Loss variation of $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ ferrite as a function of frequency and substitution (x=0.0-1.0).

expression, $\varepsilon'' = [(\sigma_{DC}/\omega\varepsilon_o) + \varepsilon''_{AC}]$ [16-17].It implies the inverse nature of conductivity w.r.t. frequency; all samples (except 0.2) in figure 3 follow this equation. Thus loss factor increases at low frequency region. Sample 0.2 exhibits lowest ε′ and ε″ (Figure 2 and 3) almost along entire frequency region: It has high resistivity among all samples (Table 1). The high resistivity manifests for polarization reduction and is desirable to suppress eddy currents for high frequency applications. The negative values of imaginary permittivity and permeability in the low frequency region are attributed to drift in network analyzer.

As explained earlier and in our previous investigation on bulk Ba-Sr ferrites [18] porosity decreases with substitution of Co^{2+} and Zr^{4+} ions, increasing polarization (ε′ and ε″) due to ease of field flow. On the other side resistivity varies nonlinearly (Table 1) with substitution two maxima at lower and higher substitution: High resistivity discourages polarization and conductivity. The highest density in samples 1.0 [16] causes increase in ε′ and ε″ while it has high resistivity than sample 0.8, decreasing ε′ and ε″. The competition between two factors results in larger ε′ in sample 0.8 than sample 1.0. Thus reciprocal nature of resistivity and polarization causes non-linear variation of ε′ and ε″ with substitution.

Permeability increases with substitution of Co and Zr ions as shown in figure 4. It is attributed to the fact that substitution causes reduction in grain boundaries (Figure 1) accompanied by increase in grain size [19]. The grain enlargement reduces the resistance to domain wall motion, thereby enhancing permeability. The sample 0.0 and 0.2 have nearly similar μ″ from 1.3-3.1 GHz while sample 0.8 and 1.0 from 2.1-3.9 GHz. Sample 0.2 shows minimum μ″ ranging from 0.0003 -0.009 from 1.8-3.6 GHz while next to it is sample 1.0 with values 0.001-0.009 from 1.4-3.9 GHz. The damped domain wall dispersion is present in the vicinity of 1 GHz (Figure 5) in undoped sample 0.0 as well as doped samples. The damping is related as: In a material whichis not magnetically homogeneous, it is plausible that the resonance frequency is, in general, different in different elementary volumes (even when no external static field is applied). The polycrystalline sample in external static field would, of course, experience different effective orientations of the crystalline magnetic anisotropy fields relative to the applied field. The crystallite shape may have an analogous effect and in each case the result will be a broadening of the single resonance into a band determined by the spread of the effective internal fields, so giving apparent damping of assumed single resonance. The damped dispersion shifts to right with the substitution of Co and Zr ions. Snoeks limit is not valid here, presumably associated with uncertainty which is further related to effect of VNA drift on real permeability.

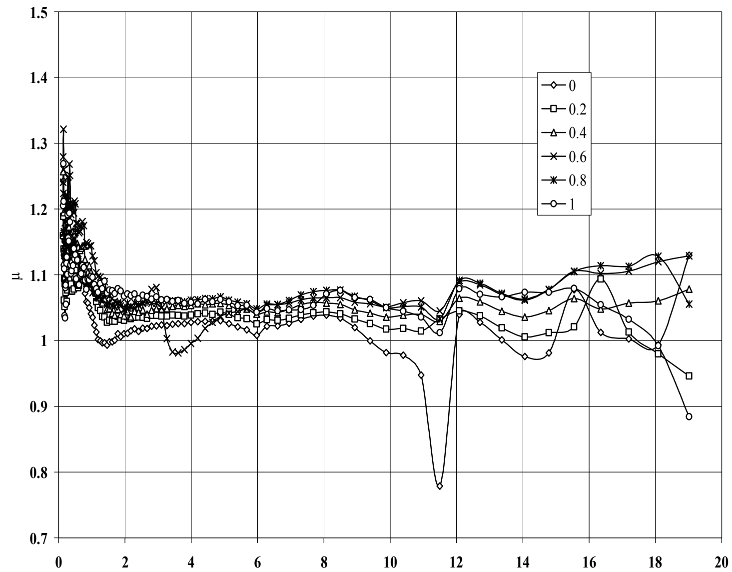

Figure 4. Permeability variation of $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ ferrite as a function of frequency and substitution (x=0.0 -1.0).

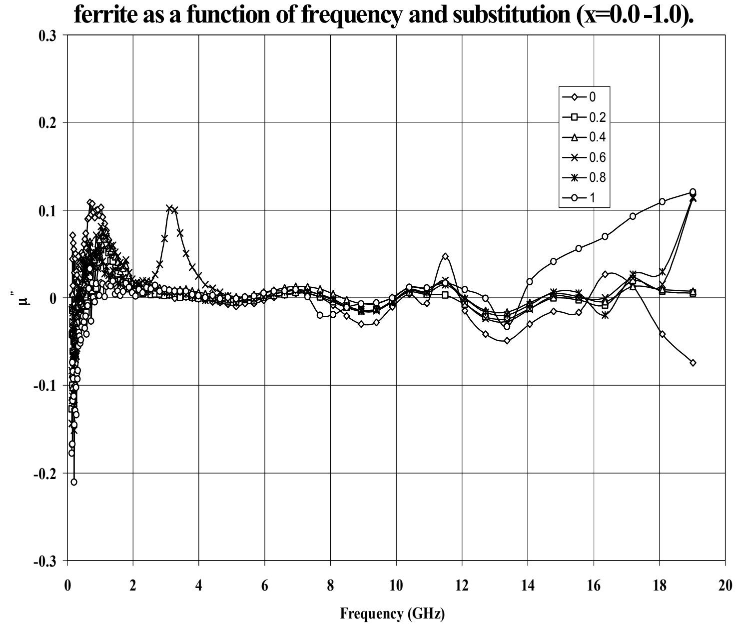

Figure 5. Magnetic loss variation of $Ba_{0.5}Sr_{0.5}Co_xZr_xFe_{(12-2x)}O_{19}$ ferrite as a function of frequency and substitution (x=0.0 -1.0).

Low μ′ and μ″ are observed in all samples in comparison to ε′ and ε″ along entire frequency region. The ferrite particles are encapsulated with paraffin wax which creates discontinuity between them. Hence the flow of magnetic flux is hindered leading to lowering of μ′ and μ″.

Conclusions

1. The prime finding of this investigation is related with enhanced dielectric properties in comparison to magnetic properties, magnetic ferrites behave as dielectric materials. Therefore ferromagnetic ferrite composites exhibit dielectric behavior at investigated region which is in contrary to earlier investigations which stress on improving magnetic properties. These enhanced dielectric properties open the possibility for their applications of EMC/EMI applications.
2. Microstructure influence complex permittivity and complex permeability. Therefore, a relationship is present among microstructural and dynamic properties.
3. High temperature (1250ºC) causes enhancement of electron hopping between Fe^{2+} ions and Fe^{3+} ions. This accompanied by porosity and resistivity variation are the decisive factors for variation of dielectric properties.
4. High resistivity, very low magnetic loss in sample 0.2 from 1.0-3.6 GHz make it useful to counter eddy currents for high frequency applications.
5. Sample 0.0, 0.2, 0.4, 0.8 and 1.0 become frequency and substitution independent from 2-5.4 GHz. These samples can be useful for application requiring constant permeability in the investigated frequency region.
6. We have obtained good electromagnetic properties with only 20% fraction volume of ferrite powder. This is also important from cost factor in the market.

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