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Dielectric and magnetic behavior of $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrites

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ABSTRACT

We have prepared $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ($x=0$, 0.5, 1, 1.5 and 2.0) W-type hexagonal ferrites by standard ceramic method. In this work, the structural, dielectric and magnetic properties have been studied of the prepared samples. The XRD analysis of the samples reveals single phase behavior sintered at 1400 °C for 6 h. The saturation magnetization (M_s) shows increasing behavior with the increasing concentration of Sr^{2+}. While the coercivity (H_c) decreases rapidly up to 409 G for $x=1.5$ and then increases for ($x>1.5$) due to the preference of Cd^{2+} for tetrahedral sites and the number of spin-down magnetic ions. The real and imaginary parts of the dielectric constant (ε',ε'') and dielectric loss tangent ($\tan\delta$) are determined in the frequency range 0.1–10^7 Hz. It is observed that both the real and imaginary parts of the dielectric constant and $\tan\delta$ decrease with the increasing concentration of Sr^{2+}, which is due to the contribution of Cd^{2+} ions in addition to Fe^{3+} and Fe^{2+} ions to interfacial polarization.

1. Introduction

Hexagonal ferrites have been playing a dominant role in the family of permanent magnet in the market due to their low costs, chemical stability and high frequency sustainability for several decades. A slight enhancement in hard magnetic properties having a great industrial importance, leads to many drastic changes in the properties of these compounds for specific applications [1]. With the extensive growth in the use of electrical and electronic devices in industrial, commercial, and military applications, electronic magnetic interference (EMI) has become a serious problem.

Electromagnetic (EM) materials, which usually consist of dielectric or magnetic fillers and polymers, are commonly used to minimize EMI [2]. Ferrites show significant magnetic losses at ferromagnetic resonance (FMR) position. Because of this they are the best material used as magnetic filler. As a ferrite absorber, they can be used for television ghosting, forged echoes in ships, radar signals, and electromagnetic wave leakage from various electric equipments [3]. Ferrites with spinel crystal structure can be applied in the high-frequency region of several hundreds MHz to several GHz [4]. While the other types of hexaferrites materials can be used across the whole GHz region [5]. The numerous ferrite chemical compositions and crystal structures make it possible to modify their magnetic and electrical properties, such as the saturation magnetization, the magneto-crystalline anisotropy, the permittivity, and the permeability. It has been also observed that the processing of ferrites and their composition for the development of suitable absorber is equally important. W-type hexagonal ferrites have an excellent feature as a microwave absorber material due to its soft magnetic character [6]. This soft magnetic character is the result of its formation at high sintering temperature greater than 1200 °C which implies a large grain size [7].

In this study, we have investigated the effect of Sr^{2+} substitutions for Cd^{2+} in Barium based $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrites. The results show that the coercivity decreases up to a few hundreds Oersted which is one of the necessary conditions for EM materials [8].

2. Experiment

Samples of $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ($x=0$, 0.5, 1.0, 1.5 and 2) were synthesized using conventional ceramic technique. Powders of $BaCO_3$, Fe_2O_3, CdO and $SrCO_3$ (99% pure Merk) were mixed homogenously in an agate mortar and pestle for about one and a half hour. The finally ground powder was cold pressed under the pressure of ~30 KN/mm^2 using Paul-Otto Weber hydraulic press in order to make pellets having diameter of 10 mm and thickness 1.5–1.7 mm. The pellets were pre sintered in air at 1270 °C for 20 h in digital electronic box furnace to reduce the surface energy of the powders [9–11]. All the samples were finally sintered at 1400 °C for 6 h followed by air quenching to improve the crystalline homogeneity of the material [12]. Brucker (D8 Focus) X-ray diffractometer equipped with Cu$K\alpha$ radiations was used to identify the phases developed in the samples.

Magnetic properties like saturation magnetization (M_s), remanent magnetization (M_r) and coercivity (H_c) were determined from M–H loops, taken on Lake Shore 7400, vibrating sample magnetometer with the applied magnetic field from 0 to ±8000 G at room temperature. The VSM was calibrated using Ni-standard sample. The complex permittivity i.e. ε' and ε'' was measured by using Alpha-N high resolution dielectric analyzer in the frequency range 1–10^7 Hz, at room temperature.

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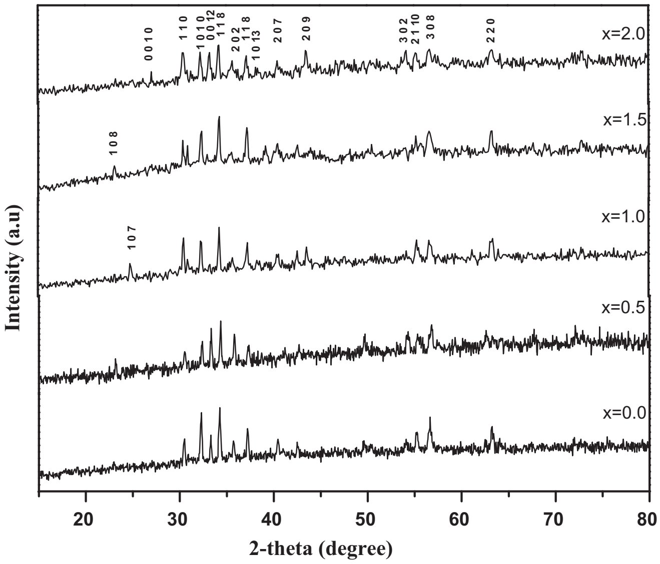

Fig. 1. X-ray diffraction patterns for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrites with $x = 0.0$, 0.5, 1.0, 1.5 and 2.0 concentration.

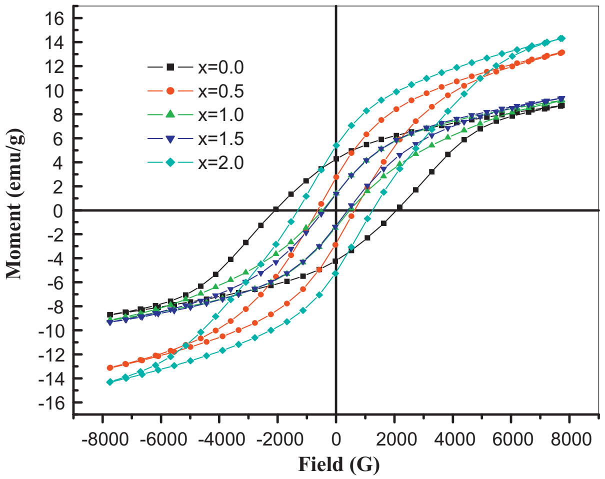

Fig. 2. The hysteresis loop of $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrites with $x = 0.0$–2.0 sample, at room temperature.

3. Results and discussion

The XRD patterns of $BaCd_{2-x}Sr_xFe_{16}O_{27}$ with $x = 0.0$, 0.5, 1, 1.5 and 2.0 are shown in Fig. 1. The PANalytical X'Pert High Score Software was used to identify the peaks obtained in the region $2\theta = 20$–$80°$. The results show that all the samples are single phase with W-type hexagonal structure. The observed phase was identified by using position and intensity of the X-ray diffraction lines of Lindquistite, $Pb_2MeFe_{16}O_{27}$, JCPDS Card No. 82-0041 [13]. At $x = 0.5$–2.0 two peaks at $2\theta = 40.502$, 49.594 of M-type phases of $BaFe_{12}O_{19}$ were also observed by using the JCPDS cards No.84-0757 which is quite in agreement with the literature [14]. It has been reported [10] that the cadmium containing W-type hexagonal ferrites prepared by the wet method were not single phase. In the present work at $x = 0.0$, the sample $BaCd_2Fe_{16}O_{27}$ which is prepared by the dry method, shows a well define W-type phases. At $x > 0.0$, Cd^{2+} was replaced by Sr^{2+} as a divalent element, which shows a possibility for the formation of M-type phases in $BaCd_{2-x}Sr_xFe_{16}O_{27}$ compounds. It is because of the comparable ionic radii (1.32 Å) of Sr^{2+} atoms with Ba^{2+} ionic radii (1.49 Å), due to this most of Sr^{2+} atoms may replace Ba^{2+}, rather to occupy interstices states, which produces few peaks of M-type structure [15].

The M–H loops for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ with $x = 0.0$, 0.5, 1, 1.5 and 2.0. W-type hexagonal ferrites are shown in Fig. 2. The values of the saturation magnetization (M_s), remanent magnetization (M_r) and coercivity (H_c) are presented in Table 1.

The results show that the saturation magnetization (M_s) has a maximum value at $x = 2.0$ and has a minimum value at $x = 0.0$ as shown in Fig. 3. Saturation magnetization (M_s) has the minimum value at $x = 0$ because W-type structure is not developed completely i.e. tetrahedral sites remain unoccupied by the Cd^{2+}, because Cd^{2+} ions have the preference for tetrahedral sites. As this preference of Cd^{2+} for tetrahedral sites decrease the number of spin-down magnetic ions thus increase the overall saturation magnetization [14]. The remanence (M_r) decreases with the increasing substitution of Sr^{2+} but increases maximum at $x = 2.0$ for maximum substitution of Sr^{2+} as shown in Fig. 4. The coercivity (H_c) has a maximum value 2083 G at $x = 0.0$, decreases at $x = 1.5$ (~409 G) and then shows increasing trend for $x > 1.5$, as presented in Table 1 and shown in Fig. 5. In W-type phase substitution of Sr^{2+} as a divalent element, most of these elements occupy tetrahedral sites in the unit cell, resulting in decrease of coercivity and increase in saturation magnetization. At $x = 2$ maximum value of coercivity obtained was due to presence of Sr^{2+} because Sr^{2+} has an ionic radius of 0.132 nm which is comparable to Ba^{2+} (0.149 nm). Hence Sr^{2+} ions will prefer to go on oxygen sites just like Ba^{2+} ions [15].

Figs. 6(a) and (b) and 7 show the complex permittivity and loss tangent in the frequency range from 1 to 10^7 Hz. It can be seen from the results that the content of Sr^{2+} ions has a significant effect on the value of real and imaginary part of permittivity with increasing frequency. The results show that at $x = 0$ complex dielectric constant ε' and ε'' has maximum values at very low frequency ~0.1 Hz, as shown in Fig. 6(a) and (b). At $x = 0$ which contains only Cd^{2+} as

Table 1
The values of magnetic properties like, saturation magnetization, M_s; remanent magnetization, M_r; and coercivity, H_c for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ with $x = 0.0$–2.0 W-type hexagonal ferrites sintered at 1400 °C.

Composition (x)	Magnetization (M_s) (emu/g)	Retentivity (M_r) (emu/g)	Coercivity (H_c) (Oe)
0.0	8.715	4.23	2083.9
0.5	13.145	2.747	636.12
1.0	9.16	1.371	468.96
1.5	9.33	1.323	409.47
2.0	14.311	5.249	1278.4

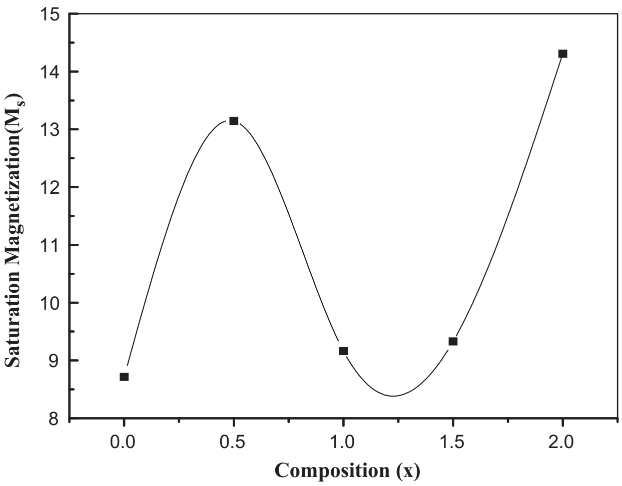

Fig. 3. Dependence of the saturation magnetization M_s on the concentration for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ferrites.

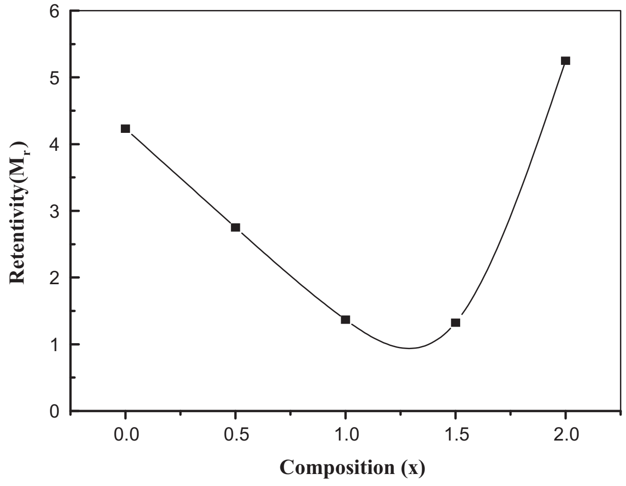

Fig. 4. Dependence of the remanent magnetization, M_r on the Sr^{2+} concentrations for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ferrites.

divalent element, real and imaginary parts both have maximum values, while with the substitution of Sr^{2+} atoms for Cd^{2+}, complex permittivity ε' and ε'' decreases (~10.396–1.168). The values of complex permittivity "ε" for each composition decrease further with the increase of frequency up to 10^5 Hz and become constant at higher frequency as shown in Fig. 6(a) and (b), which is in the agreement with the results for the W-type hexagonal ferrites [2]. This can be attributed to the fact that the dielectric properties of polycrystalline ferrite arise due to the interfacial polarization. The interfacial polarization results from the heterogenous structure of ferrites comprising low-conductivity grains separated by higher-resistivity grain boundaries as proposed by Koops [16]. The decrease in dielectric constant for sample $BaCd_{2-x}Sr_xFe_{16}O_{27}$ with increasing frequency is due to the fact that the polarization decreases with increasing frequency and then reaches to constant values similar as in Shaikh et al. [17]. The higher values of ε' and ε'' (for $x = 0$) may be due to the significant contributions of Cd^{2+} ions in addition to Fe^{3+} and Fe^{2+} ions to interfacial polarization. The smaller values of complex permittivity for $x > 0$ indicate that the major ion ($Cd^{2+}Sr^{2+}$) constituents in $BaCd_{2-x}Sr_xFe_{16}O_{27}$ contribute less effectively to cause interfacial polarization as compared to Fe^{3+} and Fe^{2+} ions [18].

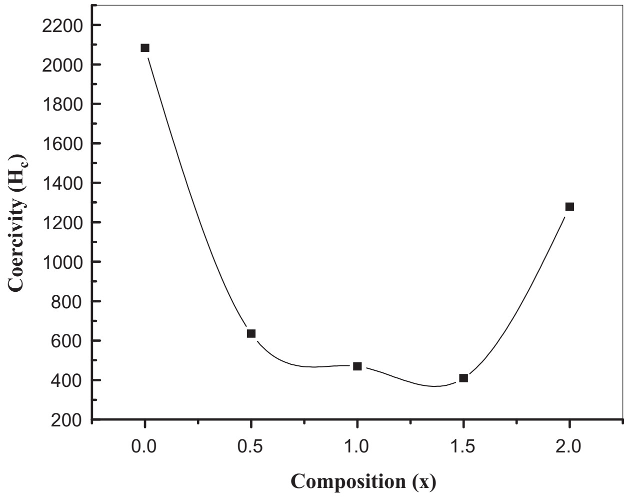

Fig. 5. Plot of coercive force, H_c on the Sr^{2+} concentrations for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ferrites.

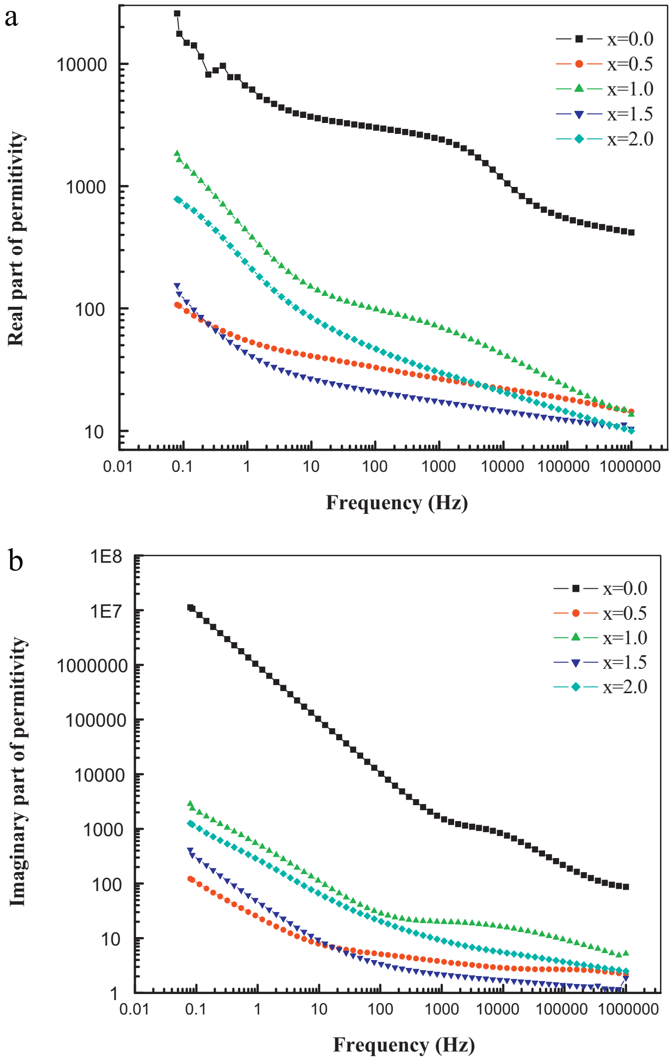

Fig. 6. (a) The real part of the complex permittivity (ε') vs. from 0.01 to 10^7 Hz for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrite with concentration $x = 0.0–2.0$. (b) The imaginary part of the complex permittivity (ε'') vs. from 0.01 to 10^7 Hz for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrite with concentration $x = 0.0–2.0$.

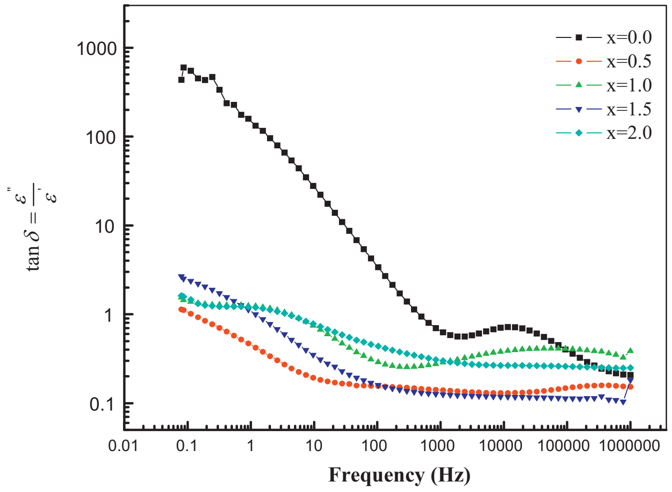

Fig. 7. Frequency dependence of loss of tangent ($\tan\delta$) at room temperature for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrite with concentration $x = 0.0–2.0$.

Fig. 7 shows a plot of the dielectric loss tangent tan δ vs. frequency for $BaCd_{2-x}Sr_xFe_{16}O_{27}$ W-type hexagonal ferrites. It can be seen that for all the samples it decreases continuously with increasing frequency, which is due to the fact that since ε'' decrease resulted in the decreasing behavior for all the concentrations in $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ferrites.

4. Conclusions

The substitution of Sr^{2+} for Cd^{2+} ions in $BaCd_{2-x}Sr_xFe_{16}O_{27}$ ferrites shows an improvement in the formation of W-type phase sintering temperature at 1400 °C prepared by standard ceramic method. The results exhibit that coercivity H_c, significantly decreases up to a few hundreds Oersted by the substitution of Sr^{2+} ions for Cd^{2+} which is almost in agreement with the requirement of W-type hexagonal ferrites.

The substitution of Sr^{2+} atoms effectively decreases the real and imaginary part of complex permittivity (ε', ε'') and tangent loss, confer that these ferrites are suitable for high frequency application such as microwave absorber.

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