Magnetization and initial permeability studies of Nd^{3+} substituted Zn–Mg ferrite system

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Abstract. Polycrystalline ferrites, $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_{2-y}\text{Nd}_y\text{O}_4$ ($x = 0.00, 0.20, 0.40, 0.60, 0.80$ and 1.00 ; $y = 0.00, 0.05$ and 0.10), were prepared by standard ceramic method. The compositions, on characterization by X-ray diffraction, showed formation of single phase cubic spinels. Magnetic study of the compositions showed increase in magnetic moment, n_B , with Zn^{2+} concentration up to $x = 0.4$ and a decrease thereafter. This was attributed to the existence of canted spin. The compositions for $x = 0.8$ and 1.0 showed paramagnetic behaviour at and above room temperature. The substitution of Nd^{3+} ion caused reduction in the magnetic moment and Curie temperatures. Substituted Nd^{3+} ion showed its occupancy on octahedral (B) site. The dependence of the initial permeability was studied in the temperature range 298 K–700 K. This μ_i – T curve reflects the positive temperature coefficient of initial permeability for the compositions $x \leq 0.4$ and $y = 0.00$. On substitution of Nd^{3+} ion (i.e. $y = 0.05$ and 0.10), the μ_i – T curves flatten, showing almost temperature independence of initial permeability. This is explained by positive contribution to the anisotropy constant, K_1 , by substituted rare-earth, Nd^{3+} ion.

Keywords. Polycrystalline ferrite; rare-earth; magnetic moment; initial permeability; anisotropy constant.

1. Introduction

Polycrystalline spinel ferrites have wide ranging applications in various technical fields due to their interesting electrical and magnetic properties. It is well known that the intrinsic properties of ferrites depend on their chemical composition, preparative conditions and substitutions (Rezlescu and Rezlescu 1997). By introduction of a relatively small amount of foreign ion, important modifications in structural and magnetic properties can be obtained (Rezlescu *et al* 1992, 1993). The intrinsic properties of Zn^{2+} and Mg^{2+} containing ferrites have been investigated (Suryavanshi *et al* 1991; Bhosale *et al* 1995; Ki Chul Han *et al* 1995). Effect of rare-earth (Sm, Gd, La, Nd, Dy) substitution on electrical properties of Cu–Zn ferrites have been extensively studied (Sattar and Shokrofy 1997). Rezlescu and Rezlescu (1997) had reported the effect of rare-earth (Yb, Er, Dy, Tb, Gd, Sm, Ce) substitution on magnetization and initial permeability in Ni–Zn ferrite system. In the present communication we are reporting the results concerning the influence of Nd^{3+} substitution on magnetization and initial permeability in Zn–Mg ferrite system.

2. Experimental

Polycrystalline ferrites $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_{2-y}\text{Nd}_y\text{O}_4$ ($x = 0.00, 0.20, 0.40, 0.60, 0.80$ and 1.00 ; $y = 0.00, 0.05$ and 0.10),

were prepared by a standard ceramic technique, using AR grade oxides, ZnO, MgO, Fe_2O_3 and Nd_2O_3 . The oxides were mixed in proper weight proportion and milled with acetone base. The samples were presintered at 700°C and then cooled in the furnace. The sintering temperature was maintained at 1000°C for 24 h and then slowly cooled at the rate of 80°C/h to room temperature. In order to confirm completion of solid state reaction, X-ray diffraction of powdered product samples was done on a X-ray diffractometer, Philips Make, PW 1710, with $\text{CuK}\alpha$ radiation. Saturation magnetization of the pelletized samples was measured by using high field B–H loop tracer, as described elsewhere (Radhakrishnamurthy *et al* 1971). Initial permeability of the samples in toroidal shape, with inner diameter 10 mm and outer diameter 20 mm was calculated from inductance measurement on LCR-Q meter HP 4192, in the temperature range 298 K–700 K, keeping the frequency of applied field constant at 1 kHz.

3. Results and discussion

X-ray diffractograms of the powered compositions reveal formation of the single phase, cubic spinel ferrite, showing well defined reflections, without any ambiguity. Figure 1 depicts the typical diffractograms for the compositions $x = 0.20$; $y = 0.00, 0.05$ and 0.10 , respectively. Variation of lattice constant ' a ' as indicated in figure 2, shows linear relationship with Zn^{2+} concentration (x), obeying Vegard's law. This increase in the

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Figure 1. Typical X-ray diffractograms for the compositions $x = 0.20$; $y = 0.00, 0.05$ and 0.10 .

lattice constants with Zn^{2+} concentration may be due to large ionic radius of Zn^{2+} ion, which when substituted, occupies tetrahedral (A) site and displaces smaller Fe^{3+} ion from tetrahedral (A) site to octahedral (B) site (Joshi *et al* 1991). This suggests strong occupancy of Zn^{2+} ion on A-site. The substitution of Nd^{3+} ion results in a slight reduction in the lattice constant. Similar results have been reported by Rezlescu and Rezlescu (1997) in rare-earth substituted Ni-Zn ferrite system suggesting occupancy of rare-earth ion on octahedral (B) site.

Saturation magnetization, M_s , magnetic moment, n_B , and Curie temperature, T_C are determined and presented in table 1. The results are in close agreement with the reported values for MgFe_2O_4 ferrite (Karche *et al* 1997). As indicated in table 1, the saturation magnetization (in emu/g) and magnetic moment, n_B , increase with Zn^{2+}

Figure 2. Variation in lattice constant 'a' against Zn^{2+} concentration.

Table 1. Magnetization, Curie temperature and initial permeability data for the composition $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_{2-y}\text{Nd}_y\text{O}_4$.

x	y	Saturation magnetization (M_s) (emu/g)	Magnetic moment (n_B)	Curie temperature (T_C) (K)	Initial permeability (μ_i at 300 K)
0.00	0.00	29.565	1.06	675	5.60
0.20	0.00	32.289	1.21	558	32.29
0.40	0.00	54.461	2.11	455	112.89
0.60	0.00	12.800	0.515	383	3.96
0.80	0.00	—	—	—	—
1.00	0.00	—	—	—	—
0.00	0.05	25.350	0.927	630	5.49
0.20	0.05	30.780	1.170	505	23.64
0.40	0.05	48.678	1.923	410	67.46
0.60	0.05	8.746	0.358	340	2.36
0.80	0.05	—	—	—	—
1.00	0.05	—	—	—	—
0.00	0.10	21.171	0.794	623	4.60
0.20	0.10	28.560	1.107	494	22.11
0.40	0.10	44.093	1.775	400	42.59
0.60	0.10	5.333	0.223	332	2.29
0.80	0.10	—	—	—	—
1.00	0.10	—	—	—	—

concentration showing a maximum at $x = 0.4$ and a decrease thereafter. This suggests the existence of Neel's two sub-lattice model for the compositions $x \leq 0.4$ and for compositions $x > 0.4$, the existence of non-collinear spin interaction. The composition for $x \geq 0.8$ show paramagnetic behaviour at and above room temperature. On inspection of table 1, it has been found that the substitution of Nd^{3+} ion results in the reduction of magnetic moment, n_B . This may be due to substitution of Nd^{3+} ion, having low magnetic moment at room

temperature, for Fe^{3+} ion, resulting in the reduction of net magnetization at B-site. This supports the occupancy of Nd^{3+} ion on octahedral (B) site. Curie temperatures, T_C , which depend on relative weighted magnetic interaction (Vasambekar *et al* 1998), as represented in table 1, shows a decreasing trend with increasing Zn^{2+} concentration. This can be due to Zn^{2+} ion having zero magnetic moment at room temperature, occupying tetrahedral (A) site and hence resulting in a decreased interaction (J_{AB}). The substitution of Nd^{3+} ion (table 1) results in decrease of the Curie temperature. This may be due to dilution in the Fe-Fe interaction on octahedral (B) site, because the Nd-Fe and Nd-Nd interactions are smaller than Fe-Fe interaction.

Initial permeability of the compositions were calculated by inductance measurement using (Patil *et al* 1991)

$$\mu_i = \frac{L}{0.0046 N^2 h \log(d_2/d_1)}, \quad (1)$$

where the symbols have their usual meanings. Initial permeability values measured at 300°C and presented in table 1, show an increasing trend with Zn^{2+} concentration, showing a maximum for $x = 0.4$ and a fall off thereafter. This may be attributed to the fact that $\mu_i \propto M_s^2$ (Karche *et al* 1997).

In order to study the temperature dependence of initial permeability, μ_i , the permeability was determined and plotted as a function of temperature (μ_i - T curves) as shown in figures 3-5. The μ_i - T curves for the compositions $x \leq 0.40$; $y = 0.00$ as depicted in figure 3 show increase in initial permeability with temperature, giving positive temperature coefficient of initial per-

Figure 3. Dependence of initial permeability μ_i against temperature, T , for the compositions $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_2\text{O}_4$.

meability, $(\mu_i/\Delta T)$ and rise to the peak value near Curie temperature, giving sharp drops at Curie temperature, T_C . This sharp drop in μ_i supports the single phase formation of the compositions (Karche *et al* 1997). The compositions for $x = 0.6$; $y = 0.00$ show a decrease in μ_i with temperature. Initial permeability which depends upon the grain size, porosity, and chemical composition (Globus 1973; Stoppls 1980), has a measured contribution from domain wall motion, but a smaller effect due to spin rotation (Globus and Duplex 1968; Globus *et al* 1971). Permeability due to domain wall motion (Baca *et al* 1985) is given by

$$\mu_i - 1 = \frac{3\pi M_s^2 D}{4\gamma}, \quad (2)$$

where γ is domain wall energy, which is found to be proportional to anisotropy constant. Thus (Guyot and Globus 1972),

$$\mu_i = M_s^2 D / K_1 l,$$

where M_s is saturation magnetization, D the average grain size and K_1 the magnetocrystalline anisotropy constant. The peaks in μ_i - T curves observed for the compositions $x \leq 0.4$; $y = 0.00$ near Curie temperature are due to zero crossing for magnetocrystalline anisotropy constant K_1 (Roess 1970).

Figure 4. Dependence of initial permeability μ_i against temperature, T , for the compositions $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_{1.95}\text{Nd}_{0.05}\text{O}_4$.

Figure 5. Dependence of initial permeability μ_i against temperature, T , for the compositions $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_{1.90}\text{Nd}_{0.10}\text{O}_4$.

An interesting result was obtained concerning the temperature dependence of initial permeability (μ_i - T curves), by Nd^{3+} substitution (figures 4 and 5). On inspection of figures 4 and 5 it was found that, μ_i - T curves flatten for the compositions $x \leq 0.20$; $y = 0.05$ and 0.10 , whereas for compositions $x = 0.40$; $y = 0.05$ and 0.10 , the initial permeability shows an almost temperature independent behaviour. This may be due to a positive contribution of Nd^{3+} to the anisotropy constant, K_1 (Rezlescu and Rezlescu 1997) owing to its $4f$ orbitals being shielded by $5s$ and $5p$ orbitals (Moeller 1973). The curve for $x = 0.6$ is similar to that of unsubstituted compositions.

4. Conclusion

Variation of lattice parameter, ' a ' obeys Vegard's law, showing an increasing trend with Zn^{2+} concentration. Saturation magnetization and magnetic moment increase up to $x = 0.4$ and then decrease thereafter. The substitution of Nd^{3+} ion causes reduction in saturation magnetization and magnetic moment. The compositions for $x \geq 0.80$ show paramagnetic behaviour at and above

room temperature. The substituted Nd^{3+} ion resides on octahedral (B) site. The dependence of initial permeability with temperature (μ_i - T curve) gives positive temperature coefficient up to $x \leq 0.40$ for unsubstituted ferrites. Due to rare-earth [Nd^{3+} ion] substitution, the μ_i - T curves are flatter almost showing temperature independence for the compositions $x \leq 0.40$; $y = 0.05$ and 0.10 . This may be attributed to positive contribution of anisotropy constant by Nd^{3+} ion.

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