

Microstructure and magnetic behaviour of Ni-Zn-Co ferrites

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Ni-Zn-Co ferrites with the formula $\text{Ni}_{0.35}\text{Zn}_{0.65-x}\text{Co}_x\text{Fe}_2\text{O}_4$ have been prepared by classical ceramic method. X-ray diffraction patterns confirm single phase cubic spinel structure in all the samples. Saturation magnetisation, initial permeability and relative loss factor measurements are made on the samples. Micrographs are taken on sintered surfaces of the samples. The variations have been interpreted in terms of Co substitutions contributed to magnetic interactions between the cations and microstructural changes brought about by the sintering conditions.

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1. Introduction

Ferrites find their utility in many applications at frequencies ranging from few kilohertz to several megahertz. The properties of ferrites depend on sintering conditions and the amount of impurities present in or added to them. There have been several reports on ferrites [1-2] substituting with cations of different valences to improve the basic properties. The devices in power applications extensively use inductor and transformer cores made up of Mn-Zn and Ni-Zn ferrites [3]. When the operating frequencies are to be required below 1 MHz, Mn-Zn ferrites are preferred and beyond this frequency there is a switch over to Ni-Zn ferrites [4]. The cores for such applications require high magnetisation and low core losses. Improvements in magnetisation and other parameters responsible for decreasing core losses of these materials as a result of compositional and microstructural modifications would certainly help the devices to be more compact in application systems.

In our previous papers [5-7] we reported the properties of Ni-Zn ferrites, undoped and doped by Nb and Ti.

The present paper reports the variations of saturation magnetisation, initial permeability and relative loss factor of Ni-Zn ferrites substituted with ferromagnetic divalent cobalt and discusses the results on the basis of exchange interactions and microstructural contributions.

2. Experimental details

Polycrystalline Ni-Zn ferrites with the chemical formula $\text{Ni}_{0.35}\text{Zn}_{0.65-x}\text{Co}_x\text{Fe}_2\text{O}_4$ where x values ranging from 0.00 to 0.25 in steps of 0.05 have been prepared by conventional ceramic technique using the procedure described elsewhere [8]. Calcination and sintering of the samples were done at 975 °C and 1250 °C respectively for 4 hours in air atmosphere followed by furnace cooling. X-ray diffraction patterns confirm single phase cubic spinel structure in all the samples.

Characterization of the samples was done further by comparing the measured values of lattice constant and Curie temperature of the basic composition with the respective parameters of the same composition reported earlier and these are found to be in good agreement with each other [9]. Saturation magnetisation measurements on the samples are made by using a VSM with fields up to 15 kOe. Initial permeability and relative loss factor measurements are made by using LF 4192A Impedance Analyzer at 10 kHz and 1 MHz frequencies respectively. Micrographs of the samples are taken on sintered surfaces using a DMLM Leica Microscope.

3. Results and discussion

Room temperature hysteresis loops of all the samples are shown in Fig. 1. The loops areas are minimal with small coercivities indicating that the hysteresis loss is negligible in these ferrites. Fig. 2 shows variation of saturation magnetisation as a function of Co concentration. The saturation magnetisation has been found to increase significantly with substituent's concentration throughout.

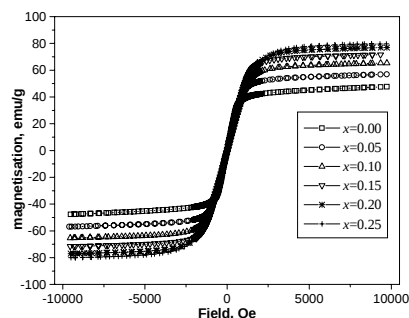


Fig. 1. Room temperature hysteresis loops of the system $\text{Ni}_{0.35}\text{Zn}_{0.65-x}\text{Co}_x\text{Fe}_2\text{O}_4$.

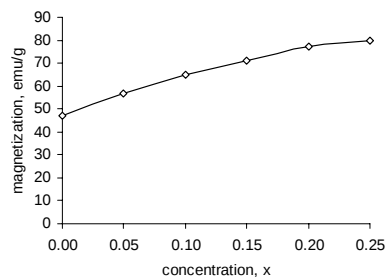


Fig. 2. Magnetisation with Co concentration in the system $Ni_{0.35}Zn_{0.65-x}Co_xFe_2O_4$

Since the basic composition contains 0.65 wt % zinc content and it is well known that all these Zn^{2+} ions occupy A-sites only, there are only 0.35 wt % Fe^{3+} ions likely to occupy A-sites for this composition and the remaining 1.65 wt % Fe^{3+} ions are expected to sit in B-sites along with 0.35 wt % Ni^{2+} ions [10]. The small quantity of Fe^{3+} ions present in A-sites is observed to be too small to maintain the predominant A-B exchange interaction alone among A- and B-sublattices and hence some degree of B-B exchange interaction is assumed to take place for this composition. As a result, the basic composition of the present system provides relatively low magnetisation value.

Since the ferromagnetic Co^{2+} ions with magnetic moments of $3 \mu_B$ have marked preference for B-sites [11], substitution of cobalt ions in the basic composition forces some Fe^{3+} ions to migrate into A-sites to fill the vacant Zn^{2+} sites as a result of substitutions and thereby yields higher saturation magnetisations due to either strengthening of A-B exchange interaction or weakening of B-B exchange interaction with each step of substitution. The observed higher values of magnetisation (Fig. 2) are in accordance with the above predictions.

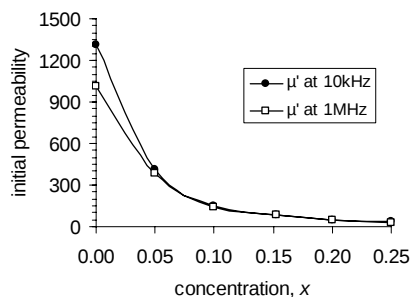


Fig. 3. Variation of initial permeability with Co concentration in the system $Ni_{0.35}Zn_{0.65-x}Co_xFe_2O_4$

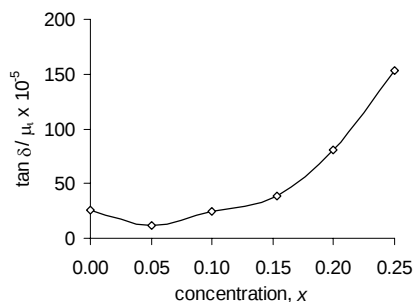
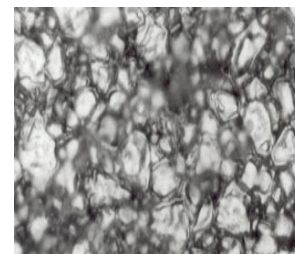
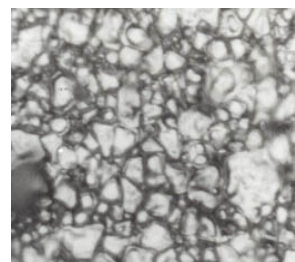


Fig. 4. Relative loss factor ($\tan \delta / \mu$) with Co concentration in the system $Ni_{0.35}Zn_{0.65-x}Co_xFe_2O_4$

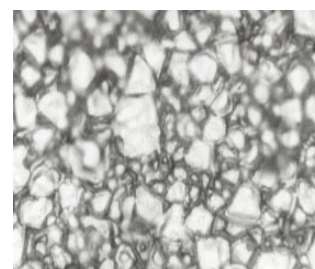
Variations of initial permeability and relative loss factor with Co concentration are shown in figs 3 and 4 respectively. The permeability is observed to decrease steeply with the substitution of cobalt. The permeability is dependent on compositional and microstructural modifications of the samples. Since, permeability is directly proportional to saturation magnetization in the case of contributions from spins rotations and proportional to the square of the saturation magnetization in the case of contributions from domain wall motions [12] and the present samples displayed higher values of magnetisation with each step of substitution, the permeability is also expected to increase with cobalt substitution. But, contrary to our expectations decreasing trend is observed with the increase in cobalt substitution. Since the permeability also depends largely on microstructure [13], to further investigate this behaviour micrographs of the sintered surfaces of the samples are examined.



a) $D=5 \mu m$; $x=0.05$



b) $D=4 \mu m$; $x=0.10$



c) $D=5 \mu m$; $x=0.15$



d) $D=3 \mu m$; $x=0.20$

Fig. 5. Micrographs of sintered surfaces of $Ni_{0.35}Zn_{0.65-x}Co_xFe_2O_4$; error = $\pm 0.5 \mu m$.

As can be seen in the micrographs (Fig. 5), the microstructures are not well characterized with specific observations such as grain size, intra- or inter-granular pores to determine the steep decrease in permeability. Thus, the variations in permeability must be governed by some other mechanism which could be magnetic anisotropy and magnetostriction contributions due to the presence of cobalt. The reports revealing that cobalt ions are known to produce large values of anisotropy constant and magnetostriction [14], which have inverse relation with the permeability [15], support the arguments made above.

The variation of relative loss factor, $\tan \delta/\mu_i$, is minimal up to $x=0.10$ Co concentration and beyond this value it increases steeply. The $\tan \delta/\mu_i$ refers loss component and represents various losses in the material. Though there is an increase in the loss component with cobalt concentration, the materials as a whole exhibited rather low losses making them useful for applications. Despite the steep decrease in permeability, the losses have not gone up. This could be due to the small hysteresis loss component as observed from the narrow hysteresis loops which keeps the relative loss factor low for lower concentrations of cobalt.

4. Conclusions

The results got on the system under investigation suggest that the cobalt substitutions against zinc in Ni-Zn ferrite system enhances saturation magnetisation, but this increase in magnetisation could not prevent the system from displaying reduced initial permeabilities. The increase in magnetisation due to the effect of replacement of non magnetic zinc ions by ferromagnetic cobalt ions with $3 \mu_B$, while the decrease in permeability is explained as due to the anisotropy and magnetostriction contributions of cobalt ions in these materials. The low values of relative loss factor are understood as consequences of rather low hysteresis losses. This fact makes these materials compatible for use in applications.

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