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Magnetic Properties of $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$

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ABSTRACT

Ni-Cd-Zn ferrites have been processed through conventional ceramic method. The diamagnetic contribution of cadmium has been investigated by substituting it in place of nickel. Observed variations in saturation magnetization, Curie temperature and initial permeability of the ferrites have been explained on the basis of superexchange interactions among tetra and octahedral sites of spinel lattice and by considering the anisotropy dependent parameters.

Introduction:

The fields of application of Ni-Zn bulk ferrites in electronic components for high frequency devices are continuously increasing. The limiting frequency of operation of these ferrites is found to be 100 MHz. Still there is a need to shift its frequency of operation to be applicable up to microwave frequencies for which an improvement in saturation magnetization and dc resistivity is inevitable. Selection of proper impurity, optimization of processing conditions and control over grain size are the necessary parameters in order to improve saturation magnetization and resistivity which in turn enhance the frequency of operation. Improvement in resistivity is possible by controlling grain growth and such small grains would provide more grain boundary area, act as barrier for electron flow. Enhancement in saturation magnetization can be expected by substituting a suitable diamagnetic or paramagnetic impurity occupying tetrahedral site and a ferromagnetic impurity having preference for octahedral site of the spinel lattice. Though a little quantity of impurity can remarkably influence the electrical and magnetic properties of ferrites, the kind of dopant and its concentration in ferrite system could be decided by the nature of applications intended to design.

An improvement in saturation magnetization is noticed [1] in $Ni_{1-x}Zn_xFe_2O_4$ till x = 0.5 with increasing zinc concentration in the spinel lattice. However, an appreciable value of initial permeability is not seen particularly at this composition. A compromise between these two parameters is considerably observed for $Ni_{0.65}Zn_{0.35}Fe_2O_4$. Suitable diamagnetic impurity having preference for tetrahedral sites similar to zinc is needed to enhance magnetization and initial permeability together. None other than cadmium ions tend to occupy tetrahedral sites due to its electronic configuration with free 5s5p orbitals and able to form covalent bonds with oxygen ions. Hence cadmium ions have been substituted in place of nickel ions in $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$ exactly till x = 0.2 so as to pronounce the diamagnetic content left with x = 0.5 without disturbing zinc concentration as described above.

Experimental details:

Samples having the general composition $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$ (x varies from 0.00 to 0.20 in steps of 0.04) were processed through conventional ceramic method. The starting materials were analytical reagent grade nickel, zinc, cadmium and iron oxides. Suitable proportions of these oxides were thoroughly ground in the presence of acetone into fine homogeneous powder. The resulting mixture was calcined in air for 4 h at 950°C and was further ground for another 2 h to obtain in the form of pellets by adding a small quantity of 15 % polyvinyl alcohol as binder. The pellets were sintered in air for 2 h at 1250°C and subsequently cooled to room temperature. In order to remove any oxide layer formed on the surface of the pellets, necessary grinding of the surface of the pellets was done. The observed single phase structure in basic Ni-Zn ferrite from X-ray diffraction pattern confirmed the procedure adopted for the processing of the series was correct one. X-ray diffraction patterns of all the samples were recorded using Rigaku Miniflex X-ray diffractometer with Cu-Kα radiation (λ = 1.5406 Å). Saturation magnetization for all the samples was measured using Ponderometer method [2]. Curie temperature was determined using Soohoo method [3]. Inductance values were measured using

HP4192A LF Impedance analyzer at the small voltage of 1mV at a frequency of 1 kHz, from which the initial permeability was calculated with the equation $\mu_i = \frac{L}{L_0}$ where $L_0 = 0.4606\, N^2 h \log\left(\frac{OD}{ID}\right)$ μH [4] is the air core inductance and N is the number of turns.

Results and Discussion:

Figure 1 gives the X-ray diffraction patterns of Ni-Cd-Zn ferrites indicating the spinel structure. Accurate estimation of lattice constant is done from X-ray diffraction patterns of Ni-Cd-Zn ferrites using Nelson-Riley function [5], provided the lattice constant for the basic Ni-Zn ferrite as 8.3830 Å which is in agreement with that of the reported [6]. Gradual increase in lattice constant is observed with increasing cadmium concentration (fig.2) following Vegard's law [7]. Increase in lattice constant is attributed to larger ionic radius of Cd^{2+} ion (0.84 Å) [8] compared to Ni^{2+} ions (0.69 Å) which suggests the entry of cadmium into the spinel lattice throughout the series by distorting the lattice. As cadmium ions are replacing nickel ions, one can expect the occupancy of cadmium towards octahedral site which is the usual preferential site for nickel in bulk Ni-Zn ferrites. As it is reported [9] that cadmium ions prefers to occupy tetrahedral sites, the vacancy created by the removal of nickel by cadmium ions at octahedral site will be filled by the migration of iron ions from tetrahedral to octahedral sites and hence resulting in increased lattice constant.

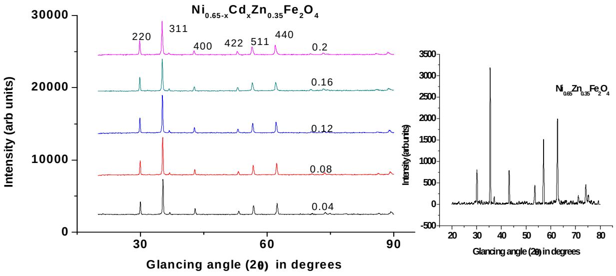
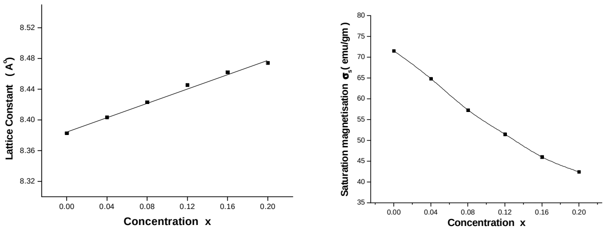


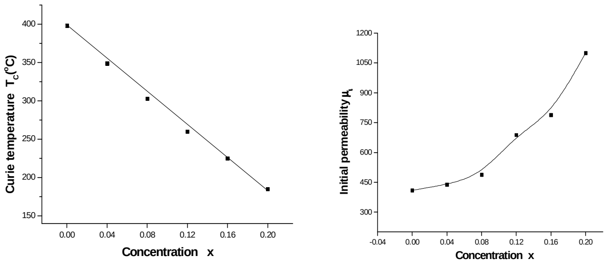
Figure 1: *X-ray diffraction patterns of* $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$

Figure 3 shows a systematic linear decrease in room temperature specific saturation magnetization with increasing cadmium concentration, reaching a minimum value of 42 emu/g. The observed decrease in saturation magnetization can be explained on the basis of site occupancy of cadmium ions which in turn modify the exchange interactions between both tetrahedral and octahedral sites of the ferrite lattice. Occupancy of cadmium towards tetrahedral sites results in migration of iron ions from A-site to B-site thereby improving B-sublattice magnetic moment.



Figures 2&3: *Lattice constant and Saturation magnetization of* $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$

Diamagnetic contribution of cadmium ions thus decreases A-site magnetic moment and consequently the difference between the magnetic moments of A and B sublattices, $M= M_B-M_A$, increases with increasing cadmium content. Contrary to this, the observed magnetization decreases which can be attributed to reduction of A-B exchange interaction due to diamagnetic cadmium and development of B-B interaction due to migration of iron ions. This modification in exchange interactions could tend to decrease the saturation magnetization by the addition of cadmium. Therefore a lesser amount of thermal energy is needed to offset the effect of exchange interactions and the same is reflected through Curie temperature measurements (fig.4).



Figures 4&5: *Curie temperature and initial permeability of* $Ni_{0.65-x}Cd_xZn_{0.35}Fe_2O_4$

An appreciable increase in the room temperature initial permeability and a gradual decrease in magnetic loss have been noticed throughout the cadmium concentration studied (fig 5). The value of initial permeability for the basic Ni-Zn composition found to be 430 which is in close agreement with that of the reported [10, 11]. The increase in initial permeability is attributed to increase in grain size with increasing cadmium content through the relation $\mu_i \propto D^{1/3}$. In addition to this, initial permeability of ferrites intimately depend on anisotropy constant according to the relation $\mu_i \propto M_s^2 / K_1^{1/2}$. Substitution of cadmium replacing nickel would reduce the small negative anisotropy contribution of Ni^{2+} ions thereby causing an increase in the initial permeability. Increase in grain size would cause increase in density and contain more number of domain walls exhibiting no hindrance to the domain wall motion.

Conclusion:

Spinel structure of ferrites has been confirmed in Ni-Cd-Zn ferrites. They exhibited a linear decrease in saturation magnetization and Curie temperature which is attributed to the diamagnetic nature of cadmium. A gradual increase in initial permeability was noticed which might be due to the increased grain size and lowered anisotropy constant of the ferrite throughout the series.

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