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Magnetic ordering in Ni–Cd ferrite

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ABSTRACT

A series of $Ni_{1-x}Cd_xFe_2O_4$ ($0.0 \leq x \leq 0.8$) were prepared by conventional double sintering ceramic method and sintered at 1200 °C for 6 h. X-ray diffraction results confirmed the single-phase spinel structures of all the samples. The Curie temperature decreases linearly with increasing Cd content, which is explained due to the weakening of the A–B exchange interaction. The sample with $x=0.7$ shows re-entrant type of spin glass phase transitions. The magnetic moment and saturation magnetization at 20 K are found to increase with Cd content up to $x=0.5$ and then tends to decrease for $x>0.5$. The increase in magnetic moment with cadmium is attributed to Neel's two sublattice (A- and B-sublattice) collinear models according to which the magnetic moment is the vector sum of the lattice magnetic moment. The decrease in magnetization for $x>0.5$ obeys the Yafet–Kittel (Y–K) model. The increase in Y–K angles for $x>0.3$ indicates the increased tendency for triangular spin arrangements on B-sites. This suggests the existence of a canted spin structure in the ferrite system with higher content of Cd.

1. Introduction

Ferrites have gained importance because they possess the combined properties of magnetic materials and insulators. It has been shown by a number of workers that a variety of substitutional ferrites can be prepared with a fairly large range of saturation magnetization and Neel temperature values. These materials are also of considerable theoretical interest, as they are ideal for the study of the various types of spin arrangement and exchange interactions. The magnetic properties of ferrite materials are crucial in selecting material for various applications, such as the transformer cores, low-frequency inductors and deflecting systems in TV circuits [1–3].

Mixed Cd-ferrite is technically important due to their high resistivity, high permeability and comparatively low magnetic losses. Cd is non-magnetic divalent ions that occupy essentially tetrahedral sites when substituted in ferrites. The magnetic properties of spinel ferrites are governed by the type of magnetic ions residing on the A and B sites and the relative strengths of the inter-(J_{AB}) and intra-sublattice (J_{AA}, J_{BB}) exchange interactions. Many researchers have studied Li–Cd [4], Cu–Zn [5], Li–Mg [6], Cu–Cd [7], Ni–Zn [8], Co–Cd [9], Mg–Zn [10] ferrite systems. To our knowledge no systematic study has been performed on the detail magnetic properties of $Ni_{1-x}Cd_xFe_2O_4$ ferrite with wide range of composition. Therefore we decided to investigate on the magnetic properties of Ni–Cd ferrite and the results of this study are presented in this paper.

2. Experimental

Ferrite samples with chemical formula $Ni_{1-x}Cd_xFe_2O_4$ ($x=0.0$, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8) were prepared through the solid state reaction using conventional ceramic technique. The raw materials for the preparation of Ni–Cd ferrite were oxides of iron, nickel oxide and cadmium oxide. The constituent in required stoichiometric proportions were weighed first and then were thoroughly mixed using ceramic mortar and pestle. The resultant powders were then ball milled for 6–8 h to produce fine powders of mixed constituents. The slurry was dried and was pressed into disk shaped sample. The disk shaped sample was pre-sintered at 900 °C for 5 h and 40 min at a heating rate of 4 °C/min to form ferrite through chemical reaction and then cooled down to room temperature at the same rate as that of heating. The pre-sintered samples were again crushed and wet milled for another 6 h in distilled water to reduce it to small crystallites of uniform size. The mixture was dried and a small amount of saturated solution of polyvinyl alcohol is added as a binder. The resulting powders

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were pressed under a pressure of 2 t/cm^{-2} in a stainless steel die to make pellets and toroids respectively. The pressed pellets and toroids were then finally sintered at 1200 °C at an interval of 50 °C for 8 h in air and cooled in the furnace.

The spinel phase formation of the ferrite was confirmed by X-ray diffraction patterns obtained using Phillips (PW3040) X'Pert Pro X-ray diffractometer. The initial permeability as a function of temperature was measured using WAYNE KERR INDUCTION ANALYZER (model no. 3255B) at fixed frequency of 100 kHz from which Curie temperature, T_C has been determined. The magnetization measurements of the samples were done by a vibrating sample magnetometer (Model VSM-02, Hirstlab, UK) at room temperature as a function of field. At 20 K magnetization as a function of field was measured by a SQUID magnetometer (MPMS XL 5). The magnetic moment in Bohr magneton has been calculated using the saturation magnetization value. Field dependence of magnetization in zero field cooled (M_{ZFC}) and field cooled (M_{FC}) condition were measured using a Superconducting Quantum Interference Device (SQUID) MPMS XL magnetometer of the Dept of Solid State Physics, Uppsala University, Sweden.

3. Results and discussion:

Structural characterization and identification of phases is a prior for the study of ferrite properties. Optimum magnetic and transport properties of the ferrites necessitate having single phase cubic structure. Fig. 1 shows the XRD pattern of all the samples of $Ni_{1-x}Cd_xFe_2O_4$. The fundamental reflections from the planes of (220), (331), (222), (400), (422), (511), (440), (620), (533) and (622) characterizing the cubic spinel structures are observed. This indicates that the synthesized ferrite compositions are of single-phase cubic spinel since no ambiguous reflections other than the spinel structures are evidenced. This also demonstrates the homogeneity of the prepared samples.

Curie temperature T_c corresponds to the temperature at which a magnetically ordered material becomes magnetically disordered, i.e., becomes paramagnet. Curie temperature also signifies the strength of the exchange interaction between the magnetic atoms. T_c of all the $Ni_{1-x}Cd_xFe_2O_4$ ferrite samples have been determined from the temperature dependence of initial permeability except for the sample with $x=0.8$, which is paramagnetic at room temperature. Fig. 2 shows the temperature dependence of permeability for the samples $x=0.0$–0.6. It was observed that the initial permeability, μ' increases with increasing sintering

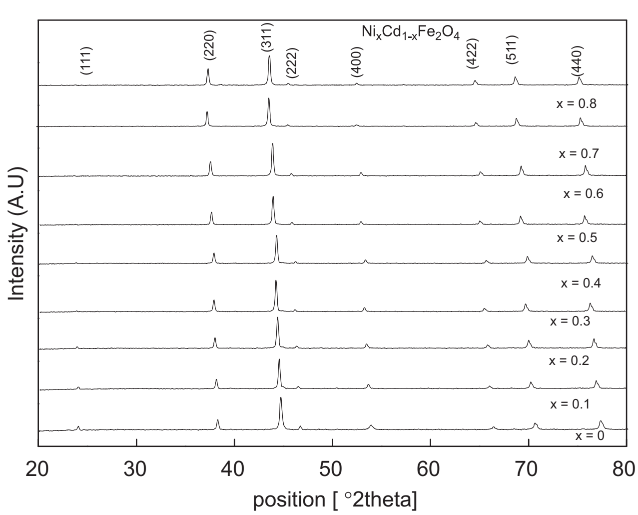

Fig. 1. XRD patterns of $Ni_{1-x}Cd_xFe_2O_4$ ferrites.

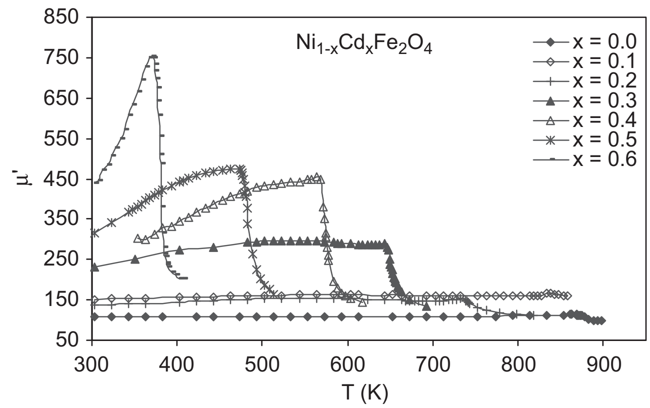

Fig. 2. Permeability vs temperature for different compositions of $Ni_{1-x}Cd_xFe_2O_4$.

temperature attaining a maximum value just below T_C. The maximum value of μ' just below the Curie temperature (T_C), is a manifestation of Hopkinson peak attributed to the minimization of anisotropy energy with temperature. Beyond this peak value μ', initial permeability sharply falls to very low value indicating the ferro-paramagnetic phase transition. T_C has been taken as the temperature at which a sharp fall of permeability is observed i.e. where $d\mu/dT$ attains its maximum value. The sharpness of the permeability drop at the Curie point can be used as a measure of the degree of compositional homogeneity according to Globus et al. [11]. The present ferrites show good homogeneity as shown in Fig. 2, where an abrupt drop in permeability occurs within the temperature range less than 5 °C near Curie point and it have also been confirmed by X-ray diffraction by our previous XRD experiment. According to Globus model μ' is given by

$$\mu' = \frac{M_s^2 D}{\sqrt{K_1}} \tag{1}$$

where M_s is the saturation magnetization, D is the average grain size and K_1 is the magneto-crystalline anisotropy constant. The anisotropy constant (K_1) and saturation magnetization (M_s) usually decrease with increase in temperature. But decrease in anisotropy constant (K_1) with temperature is much faster than the decrease in saturation magnetization (M_s). When the anisotropy constant (K_1) reaches zero, μ' attains its maximum value and then drops off to the minimum value near the Curie point. Thus, the effect of K_1 appears to be more significant in controlling the μ' of the ferrite materials [12,13].

When the non-magnetic substitution greater than 0.6, intra-sublattice interaction J_{BB} become comparable and/or more predominant than inter-sublattice interaction, J_{AB}. In such a situation disorder and frustration takes place in the spin subsystem leading to the manifestation of various magnetic structures: ferrimagnetic order, local spin canting (LSC), antiferrimagnetic order, re-entrant spin glass and spin glass etc [14,15]. Scholl et.al. reported a value of site percolation threshold of 0.429 for A site and 0.39 for B-site [16]. In our case re-entrant spin glass behavior occurs at $x=0.7$. A re-entrant spin glass or semi spin glass is a highly frustrated and disordered magnetic system in which there is a competition between spin glass order and long range ferromagnetic order. In other words a system where there exists a majority of ferromagnetic couplings between the individual spins but a sufficiently large number of antiferromagnetic couplings to create substantial frustration leading to a competition between spin glass order and long range ferromagnetic order when the temperature is lowered. In such a material it exhibits a transition from a paramagnetic (PM) to a ferromagnetic (FM) state and on further lowering the temperature spin glass like behavior generally