

Influence of Mn⁴⁺ tetravalent substitution on the magnetic properties of nickel ferrite

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Abstract. In this work we have investigated structural and magnetic properties of Ni_{1+x}Mn_xFe_{2-2x}O₄ (0.0 ≤ x ≤ 0.6) spinel ferrite. The prepared samples were characterized by X-ray diffraction (XRD) technique to confirm the formation of single phase cubic spinel structure. XRD data used to investigate structural parameters. Lattice constant is decreases with increase in manganese ions substitution. The magnetic properties investigated by using pulse field hysteresis loop tracer technique. Magnetic parameters show the strong influence of Mn⁴⁺ tetravalent substitution. The magnetic properties of the spinel system (Ni_{1+x}Mn_xFe_{2-2x}O₄) are explained on the basis of Neel's two sub lattice model of ferrimagnetism. The magnetic properties are also supported by Mossbauer data.

Keywords: Spinel ferrite, Structural properties, Magnetization, Mossbauer spectroscopy.

INTRODUCTION

Ferrites play an important role because of their extensive technological applications and due to their fundamental, electrical and magnetic twin behavior. [1]. Ferrite crystallizes in spinel structure in which both divalent and trivalent is distributed among tetrahedral (A) and octahedral [B] sites. Nickel ferrites have high electrical resistivity, low eddy current, high Curie temperature and dielectric losses over a wide range of frequencies. The basic electrical and magnetic properties can be tailor made by careful control of composition and microstructure, by method of preparation [2, 3] Nickel ferrite belongs to a class of compound having the general formula M²⁺Fe₂²⁺O₄ and crystallizes with the inverse spinel structure. The substitution of tetravalent ions like silicon, germanium etc in nickel has modified the properties of nickel ferrite [4, 5].

In this work, we report influence on magnetic properties of tetravalent manganese (Mn⁴⁺) substituted nickel ferrite.

EXPERIMENTAL

The samples were prepared by using a standard ceramic technique [6]. The A.R. grade forms (SD Fine) of oxides are mixed in stoichiometric ratio according to the formula Ni_{1+x}Mn_xFe_{2-2x}O₄; where 0.0 ≤ x ≤ 0.6. X-ray diffraction (XRD) patterns were taken at room temperature to confirm the single phase cubic spinel crystal structure of the prepared samples. The magnetic properties were studied using pulse field hysteresis

loop technique and also Mössbauer spectroscopy measurements were carried out at room temperature.

RESULTS AND DISCUSSION

Room temperature XRD patterns of typical samples (0.2, 0.4 and 0.6) of Ni_{1+x}Mn_xFe_{2-2x}O₄ ferrite system as shown in Fig. 1.

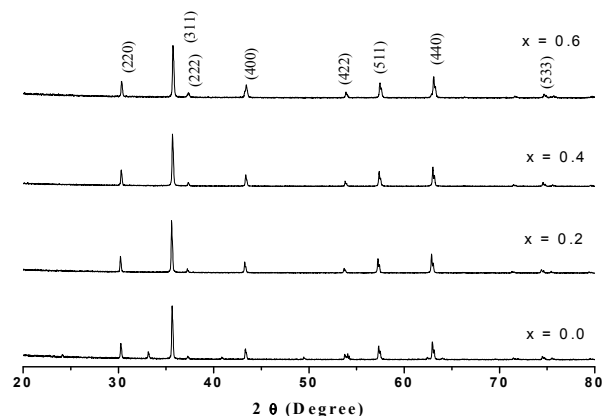


FIGURE 1: XRD pattern of Mn content x (= 0.0, 0.2, 0.4 and 0.6) of Ni_{1+x}Mn_xFe_{2-2x}O₄ ferrite system.

XRD data showed that the samples under investigation have single phase cubic spinel structure; no extra peak has been observed other than the cubic spinel. All the peaks are sharp, clear and intense. The XRD data was used to calculate the lattice constant listed in table 1.

The lattice constant of Ni_{1+x}Mn_xFe_{2-2x}O₄; increases beyond x = 0.2 the lattice constant decreases with Mn⁴⁺ content x. The decrease in lattice constant with Mn⁴⁺

content x is attributed to the difference in ionic radii of constituent ions i.e. Ni^{2+} , Fe^{3+} and Mn^{4+} . In the present series $Ni_{1-x}Mn_xFe_{2-2x}O_4$; two Fe^{3+} (0.67\AA) ions are replaced by combination of Ni^{2+} (0.72\AA) ions and Mn^{4+} (0.52\AA), thereby decreasing the lattice constant of the system should decrease. The decrease in lattice constant is almost linear and obeys Vegard's law [8]. From table 1, it is observed that X-ray density decreases because the fact that decreases in mass over takes the decrease in volume of the unit cell [9]. The particle size of each sample was determined by considering most intense (311) peak of the XRD pattern by using Scherrer formula given in the table 1.

TABLE 1: Lattice constant, particle size and X-ray density of $Ni_{1-x}Mn_xFe_{2-2x}O_4$ ferrite

| Mn content x | Lattice constant (\AA) | Particle size (\AA) | X-ray density (gm/cm^3) |
|----------------|-----------------------------------|--------------------------------|------------------------------------|
| 0.0 | 8.337 | 499 | 6.781 |
| 0.2 | 8.355 | 555 | 6.198 |
| 0.4 | 8.336 | 501 | 5.696 |
| 0.6 | 8.323 | 416 | 5.179 |

Magnetic properties:

The magnetic hysteresis curves of $Ni_{1-x}Mn_xFe_{2-2x}O_4$ for typical sample $x = 0.0, 0.2, 0.4$ and 0.6 are shown in Fig. 2.

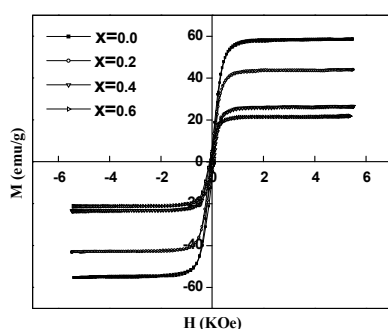


FIGURE 2: Variation of Magnetization (M_s) with magnetic field strength (H) for $Ni_{1-x}Mn_xFe_{2-2x}O_4$ ferrite.

From figure, the samples show ferrimagnetic behavior which decreases with increase in Mn^{4+} content x . These hysteresis curves are used to determine magnetic parameter listed in table 2.

TABLE 2: Magnetization (M_s), remanance magnetization (M_r), magneton number (n_B), and coercivity (H_c) of $Ni_{1-x}Mn_xFe_{2-2x}O_4$ ferrite

| Mn content x | Magnetization parameter | | | |
|----------------|-------------------------|----------------|-------|------------|
| | M_r (emu/gm) | M_s (emu/gm) | n_B | H_c (Oe) |
| 0.0 | 6.08 | 47.19 | 2.0 | 68.65 |
| 0.2 | 0.30 | 65.89 | 1.8 | 15.11 |
| 0.4 | 4.78 | 57.86 | 1.6 | 29.33 |
| 0.6 | 13.20 | 36.52 | 1.4 | 116.60 |

From table 2, saturation magnetization (M_s) increases with Mn^{4+} content x . The observed behavior of saturation magnetization is similar to that of other well

known spinel ferrite. The observed behaviour of magneton number (n_B) is tried to explain on the basis of Neel's two sub-lattice collinear spin model. According to Neel's model magneton number ' n_B ' is calculated by formula, $n_B = |M_B - M_A|$

Mössbauer studies:

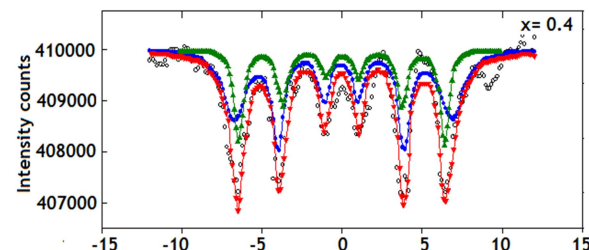


FIGURE 3: Mössbauer spectra of Mn content $x = 0.4$ of $Ni_{1-x}Mn_xFe_{2-x}O_4$ ferrite.

The Mössbauer spectrum of typical sample $x = 0.4$ of $Ni_{1-x}Mn_xFe_{2-2x}O_4$ ferrite obtained at room temperature is shown in Fig. 3. The Mössbauer spectrum is fitted with two magnetic sextets. The two magnetic sextets have been assigned to the magnetic order ions and the tetrahedral (A) and octahedral [B] sites.

TABLE 3: Mössbauer parameter of $Ni_{1-x}Mn_xFe_{2-2x}O_4$ ferrite

| Mn Content x | Site | Isomer shift (mm/s) | Quadrupole splitting (mm/s) | Hyperfine field (T) |
|----------------|------|---------------------|-----------------------------|---------------------|
| $x = 0.4$ | A | -0.053 ± 0.03 | 0.005 ± 0.04 | 481 ± 0.2 |
| | B | -0.205 ± 0.03 | -0.049 ± 0.03 | 504 ± 0.2 |

Table 3, gives the parameter such as isomer shift quadrupole splitting, hyperfine field and relative intensity at room temperature derived from Mössbauer spectrum of typical sample $x = 0.4$. The quadrupole splitting in the Zeeman sextet is very much less for tetrahedral (A) and octahedral [B] sites which is a general observation for a spinel ferrite system

Conclusions:

The single phase cubic spinel ferrite formation of the $Ni_{1-x}Mn_xFe_{2-2x}O_4$ was confirmed through XRD patterns. The lattice constant from XRD data initially increases up to $x = 0.2$ and decrease to $x = 0.6$. The saturation magnetization (M_s), and magneton number (n_B) decrease with increase in Mn content x in Ni-ferrite. The Mössbauer parameters are in good agreement with the reported spinel ferrite.

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