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ORIGINAL ARTICLE

INFLUENCE OF MN^{4+} TETRAVALENT SUBSTITUTION ON MAGNETIC VARIATION OF NICKEL FERRITE

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Abstract:

Ferrite sample have general formula $Ni_{1+x}Mn_xFe_{2-2x}O_4$ have been prepared by standard ceramic route and investigated by means of a. c. susceptibility measurement and Mossbauer spectroscopy. The Mossbauer data and a. c. susceptibility measurements suggest the Mn^{4+} ions occupy octahedral (A) and tetrahedral (B) sites. The Mossbauer spectra at temperature 300K have been fitted with two sextets in the ferrimagnetic state. The temperature dependence a. c. susceptibility behaviour is attributed to magnetic anisotropic Ni^{2+} ions in the system.

KEYWORDS:

Ferrite, a. c. susceptibility, Mossbauer spectroscopy.

INTRODUCTION

The Ni-Fe_2O_4 was prepared by using chemical formula $Ni_{1+x}Mn_xFe_{2-2x}O_4$ (where x=0.0 to 0.6 in the step of 0.1) and have been studied by a. c. susceptibility technique and Mossbauer spectroscopy. The present paper reports the preparation of selected system and investigation of effect of tetravalent Mn^{4+} substitution on the magnetic properties of Ni-Fe_2O_4 through low field a. c. susceptibility and Mossbauer spectroscopy measurement.

The formations of samples were analyzed by using X-ray diffraction. The X-ray data confirmed that the present series forms single phase and lattice constant obtained from Vegard's law [1].

1.EXPERIMENTAL:

By taking variation in Mn^{4+} ions concentration in the step of 0.1 from x= 0.0 to 0.6 in composition, $Ni_{1+x}Mn_xFe_{2-2x}O_4$ The samples were prepared by double sintering standard ceramic technique using stoichiometeric calculations carried out by following equation.

$$(1+x)NiO+xMnO_2+ (1-x)Fe_2O_3 Ni_{1+x}MnxFe_{2-2x}O_4$$

The values of x are taken in the range 0.0 to 0.6 in the step of 0.1.

The calculated amount of row material [NiO, MnO, and $FeO]_2$ were thoroughly mixed and ground for good homogeneity. The samples were pre-sintered at 950C for 12 hours and then cool to room temperature, again ground the sample and from a homogeneous mixture. This mixture again sintered at 1100C for 18 hours. After cooling the sample were sieved though five micron shiver.

The pellets were formed under pressure 6 ton/cm3 using hydraulic press. These pellets were then

sintered at 1100^{0}C for 18 hours. X-ray pattern were taken to confirm the FCC structure of the sample.

Low field a. c. susceptibility measurement of powder sample were made on a double coil set up operating d. c. voltage in the range 0-30 volt and d. c. current 0-5 amperes. [2] The measurement of a. c. susceptibility was carried out in the temperature rage of 300-500K.

Mossbauer measurements were undertaken with constant acceleration transducer and 512-multichannel analyzer operating in time mode using 57Co source. The absorber were made using sample in the powder from of thickness between 1.5 to 2 mg/cm in circular shape.

2.RESULTS AND DISCUSSION:

The variation of lattice constant 'a'with Mn content x is shown in Fig.1. From Fig.1, it can be observed that the lattice constant of $Ni_{1+x}Mn_xFe_{2-2x}O_4$increases from x= 0.0 to x= 0.1, beyond x= 0.1 the lattice constant decreases with Mn content x. The decrease in lattice constant with Mn 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 $^{3+}$ (0.67Å) ions are replaced by combination of Ni $^{2+}$ (0.72Å) ions and Mn $^{4+}$ (0.52Å), thereby decreasing the lattice constant of the system should decrease. A similar result of variation of lattice constant in tetrahedral substitution is observed [3].The decrease in lattice constant is almost linear and obeys Vegard's law.

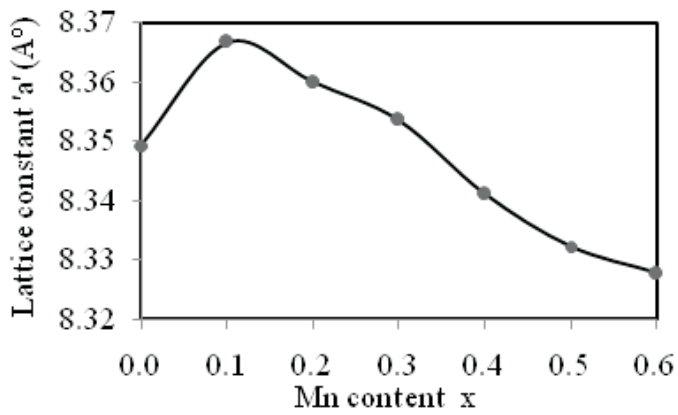

Fig.1. The variation of lattice constant 'a'with Mn content x.

The behavior of law field a. c. susceptibility as a function of temperature for Mn content x = 0.1 to 0.6 as shown in Fig.2. It has been observed that T/RT against temperature curves no dips have been observed which exhibit normal ferrimagnetic behavior. The dips show the co-existence of two ferrimagnetic oxidation state.

A sharp maximum curve was observed near a curie temperature (Tc) which shows that the substance changes from one oxidation ferromagnetic state to other oxidation paramagnetic state. The nature of curve can be explained on the basis of magnetic order in the system. Zubov and Skrebneva [4] have studied the temperature depends of initial susceptibility. A tailing effect is observed when the sample shows a transition from ferromagnetic to paramagnetic state or spin clusters.

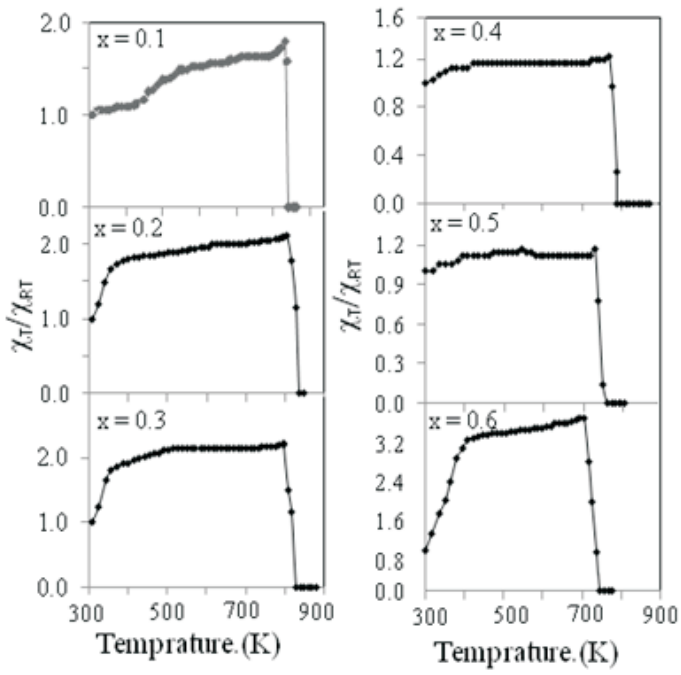

Fig.2. Curves of T/RT against temperature of Ni1+xMnxFe2-2xO4 for x=0.1 to 0.6.

The peak corresponds to change in spin ordering (paramagnetic to ferrimagnetic) from the plots of T/RT, it is seen that the ferrimagnetic peak gradually diminishes on Mn4+ dilution indicating shift towards lower temperature and decreasing Neel temperature (TN). The Neel temperature determined from the plots listed in table 1.

Table1 .
Neel temperature (T_N) for $Ni_{1+x}Mn_xFe_{2-2x}O_4$

x	0.0	0.1	0.2	0.3	0.4	0.5	0.6
T_N (K)	853	843	836	826	788	757	745

It is seen from table 1, TN decrease with increase in x. This happens due to replacement of Fe^{3+} (5μB) in the system $Ni_{1+x}Mn_xFe_{2-2x}O_4$ results in the decrease in TN. The decreasing in peak value is attributed to decrease in coercive force which is due to number and strength of A-B magnetic linkage [5].

The room temperature Mossbauer spectra exhibit Zeeman effect due to Fe^{3+} ions at both tetrahedral (A) site and octahedral [B] site for Mn content x = 0.1 to 0.6 as shown in Fig. 3.

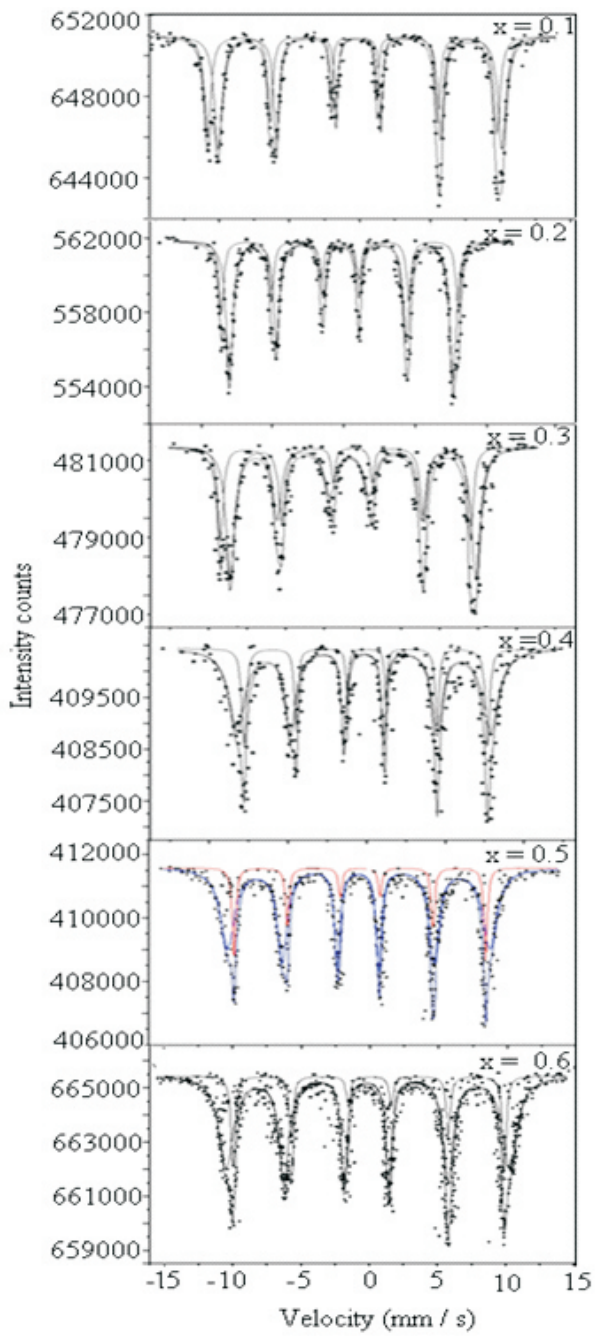

Fig.3.Mossbauer spectrum of Ni1+xMnxFe2-2xO4 for x=0.1 to 0.6.

All measured spectra have been fitted with two magnetic sextets. The magnetic sextets have been assigned to the magnetically ordered Fe^{3+} ions at the tetrahedral (A) and octahedral [B] sites.

The hyperfine interaction has been made to analyze this spectrum. The hyperfine interaction parameters deduced from the spectrum are given in table 2.

The variation of hyperfine field for A-site and B-site as a function of Mn concentration (x) depicted in Fig. 3 and the values are listed in table 2.The fitting parameter have value in table 2 good agreement those published in earlier [6]

It is evident from table 2 suggest that the values of hyperfine field for B-site is greater than A-site indicating that both side very little change with Mn4+ substitution (x) causes S-electron charge distribution

on Fe^{3+} ions in negligibly influenced by Mn^{4+} substitution. The value of isomer shift (Is) less for B-site comparatively from A-site indicating that the shifting of Fe^{3+} ions is less from site due to Mn^{4+} which is general observation in spinel ferrite [7].Similar behavior is also obtain in case of quadrupole shift

Table 2.
Values of Isomer shift (mm/s), Quadrupole Shift (mm/s), Hyperfine field (T) for the $Ni_{1+x}Mn_xFe_{2-2x}O_4$ system at room temperature.

Mn content x	Site	Isomer shift (mm/s)	Quadrupole Shift (mm/s)	Hyperfine field (T)
0.1	A	-0.044	0.085	503
	B	-0.397	-0.101	510
0.2	A	-0.168	0.006	472
	B	-0.254	0.030	491
0.3	A	-0.027	0.026	471
	B	-0.378	-0.256	494
0.4	A	-0.053	0.005	481
	B	-0.205	-0.049	504
0.5	A	-0.063	-0.023	470
	B	-0.154	0.028	490
0.6	A	-0.069	-0.068	470
	B	-0.187	0.093	540

The Mossbauer spectra indicative of paramagnetic Fe^{2+} state and ferrimagnetic Fe^{3+} state. This suggests that Fe^{2+} oxidation state has lower TN than Fe^{3+} oxidation state. These Mossbauer result are in excellent agreement with susceptibility data.

CONCLUSIONS:

From the low field a. c. susceptibility and Mossbauer data both oxidation state are in ferrimagnetic state.

Low field a. c. susceptibility data suggest that TN decreased linearly with Mn4+ substitution (x) for all composition. This variation is well support for collinear magnetic ordering which is further supported by the Mossbauer data

The Mossbauer data exhibit Zeeman sextets pattern indicate that $Ni_{1+x}Mn_xFe_{2-2x}O_4$(where x=0.0 to 0.6 in the step of 0.1) is spinal ferrite.

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