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ORIGINAL ARTICLE

EFFECT OF MN DOPING LEVEL ON MAGNETIC PROPERTIES OF Ni SUBSTITUTED COBALT FERRITE

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Abstract:

It has been observed that for cobalt ferrite the coefficient of magnetostriction is maximum but the observed value of magnetoelectric coefficient is low as compared to the value calculated on the basis of known piezo-electric coefficient(d) and magnetostrictive strain(λ). It was predicted that a large value of magneto crystalline anisotropy of cobalt ferrite reduces the effective magneto electric coupling. To improve the magneto electric coupling still maintaining the values of resistivity (p), Saturation magnetization (Ms) and initial permeability (μ) high, substitution of Nickel is found to be useful. Further substitution of Mn at A or B-site is reported to improve , and reduce anisotropy energy k1, coeresive field (Hc) and curie temperature (Tc) of the material. Therefore it was interesting to determine Magnetic properties of submicron level Ni and Mn substituted Cobalt Ferrite.

KEYWORDS:

Magnetostriction, Remnant and Anisotropy energy, Mn substituted cobalt ferrite.

I.INTRODUCTION:

Increasing power densities and decreasing dimensions of the devices are the hallmarks of the modern computer chips. In recent years, researchers in the field of computer turn their attention to study the magnetic properties of highly resistive cobalt ferrite because of their wide applications in computer memory. For typical desktop computer magnetic hard disc drives, has a capacity of more than 40 Gbyte/disc, it was 1Gbyte/disc in 1995. For magnetic data storage, the key parameter is the electron spin which can be thought of magnetic moment. A key advantage of magnetic materials memory device is that they are non-volatile since they use ferromagnetic materials that by nature have remanence [1]. These materials therefore offer realistic prospects for the development of contactless sensors and in their other applications. Also, there has been increasing interest in materials with specific nano-morphologies with expectations of getting enhanced electric and magnetic properties.

Mainly the present paper reports the effect of manganese substitution for iron on structural, saturation magnetization test (Ms), coercivity (Hc), and remnant magnetization(Mr), initial permeability (μ) and coefficient of magnetostriction (λ) of sub-micron particulate of $Co_{0.9}Ni_{0.1}Fe_{2-x}Mn_xO_4$ (CNFMO) for x=0, 0.1,0.2,0.3, and 0.4 compositions. To arrive at a composition possessing the optimal values of , Ms &

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and low Hc, CNFMO for x=0, 0.1,0.2,0.3, and 0.4 compositions are synthesized via hydroxide co-precipitation route and are subjected to the measurement of their electric and magnetic properties. The reason for selecting x values from x= 0 to 0.4 is that Mn ion being larger in size as compared to iron ion, hence cannot be substituted in crystal structure perfectly beyond x=0.5[2]. The structural characterizations of the sample were done by X-ray diffraction analysis while, magnetic properties were measured using a Hysteresis loop tracer and custom designed tensometric magnetostriction instrumentation setup.

II.EXPERIMENTAL PROCEDURE

We have been interested in hydroxide co-precipitation method for the synthesis of series of manganese doped cobalt ferrite with compositions of $Co_{0.9}Ni_{0.1}Fe_{2-x}Mn_xO_4$, where x ranges from 0 to 0.4 because the method ensure ease of preparation, chemical homogeneity at precipitates, purity and uniform grain growth. For good magnetic properties attention is made on sintering temperature, chemical composition and the preparation condition on which the magnetic properties mostly depends.

The AR-grade $Fe(NO_3)_3.9H_2O$, $Co(NO_3)_2.6H_2O$, $Ni(NO_3)_2.6H_2O$, $MnCl_2$ $4H_2O$, are used as precursors while, a mixture of NH_4OH and KOH is used as precipitating agent. The details of the co-precipitation route are similar as reported earlier [3, 4]. The precipitate formed is washed thoroughly and calcinated at 1100ºC for 12 hrs to achieve complete ferrite phase formation. The CNFMO powder is pelletized in the form of disc of 1.2cm diameter using pressure of nearly $2T/cm^2$. Further, the pellets are sintered at 1200ºC for 10 hrs to achieve a dense ferrite composition. In this case the compositions are synthesized by adopting co-precipitation route similar to the process reported by bhame et al., only difference is in the final sintering temperature and duration of sintering [5]. In the present case, the final sintering is carried out at1200ºC for 24 hrs. The samples were then investigated for their structural characterizations and magnetic properties.

For complete characterization of these compositions, the dc resistivity (ρ_{dc}) is measured using potential divider arrangement. The physical density (d_{Bulk}) is measured using the liquid displacement method while saturation magnetization (M_s) is measured using Hysteresis loop tracer from Ms. Arun Electronics, Mumbai (India). The permeability (μ) is measured using a LVDT arrangement.

III.RESULTS AND DISCUSSION

Here figure 1 shows XRD spectra of the calcined CNFMO powder for x=0.2 and 0.4. The spectra for other compositions of the series are similar as shown in figure 1. It is observed that the XRD spectra are in accordance with JCPDS data of the parent composition (JCPDS card Nos.77-0428, 80-0072). Within the detectable limit, no peak corresponding to any impurity phases is recorded in the XRD spectra. Thus from the XRD spectra it is revealed that the ferrite compositions were synthesized in the desire spinal cubic crystal structure without any detectable impurity phase. Using the XRD spectra, the lattice parameter 'a' is calculated and is given in table 1. It is observed that the lattice parameter increases slowly with increasing Mn contain and may be attributed to the slightly larger ionic radius of Mn as compare to Fe ions [6]. Using the Scherrer formula the particle size is calculated and is also given in the table 1. Here the particle size of sintered co-precipitate is above 50 nm. This is expected because of long time cacination at 1100ºC for of 12 hrs.

The ferrite compositions are further subjected to the determination of ρ_{dc}, X-ray density ($d_{X\text{-}ray}$), physical density (d_{Bulk}) and porosity (p). The observed variations of ρ_{dc} and 'p' are also given in table 1. It is observed that ρdc is very high at x=0 and decreases slowly with increasing Mn content. The presence of Fe^{2+} and Fe^{3+} or $Co^{2+}$and Co^{3+}ions on equivalent sites is known to cause polaronic conduction in case of ferrites. At very low concentration the Mn and Ni reduce the percentage of fractional Fe^{2+} and Co^{2+} ions formed during the process of synthesis. This causes an increase in resistivity for substitution of Mn/Ni below 0.04 atom percentage [2]. For further increase in Mn/Ni concentration ρdc decreases slowly with increasing x. The present observations are in confirmation with the earlier reports [7]. The magnitude of porosity is as shown in table 1. It is observed that the samples are dense and porocity is than 10% comparable with the porosities reported earlier for similar sintering conditions [8]. Here the aim of the present studies is to prepare a composition with similar sintering conditions as reported earlier; nevertheless the final sintering is kept limited to 1200 ºC for 24hrs. This may reduce the value of Ms and λ by less than 5% as the porosity is slightly high.

Figure 2 shows the variation of λ_{η} with applied magnetic field (H). It is observed that, the λ_{η} increases and appears to saturate for H > 4 kOe. This could be because of the demagnetization factor of disc shape sample and higher value of Hc for $CoFe_2O_4$. The highest value of λ_{η} for H=4.5kOe is termed as sat.

Along with the observed magnetic property like λ , Ms, Hc, and μ of the CNMFO series, these parameters for $CoFe_2O_4$ are reported in same table 2 . These values are also in confirmation with the earlier reports on similar systems [5, 6]. From table 2 it is observe that the saturation magnetization (Ms) increases with increasing Mn content as expected for Mn substituted cobalt ferrite [9]. The present observations show that the Mn fractionally occupies B-sites also, and Ms increases in nonlinear fashion with x instead of following the linear behavior. The variation of Ms with x is shown in figure 3(a). This suggests that the majority of Mn ions are present on the A-sites. Further, Hc for all the compositions are closer to 100 Oe and this observation too is in confirmation with the earlier report [5]. This behavior of substitution of Mn is interesting as compared to the earlier reports on Mn substitution in $CoFe_2O_4$ [6, 10]. It is observed that for substitution of Mn in $CoFe_2O_4$, the Ms increases up to x=0.2 only and decreases for further increasing Mn concentration. The present report on the other hand show that Ms increases monotonically for Mn even up to x=0.4. Thus the substitution of Ni at x=0.1 has caused majority of Mn to occupy its preferential A-sites than occupying the B-sites.

The table 2 shows the variation of μ as a function of x. The μ is expected to be proportional to Ms^2 and the present observations are in concurrence with this prediction. The figure 3(b) shows that μ varies linearly with Ms^2. The earlier reports on the Mn substituted cobalt ferrite predict that the presence of Mn ions on A-sites causes an increase in and magneto mechanical coupling of CNFMO [6]. The table 2 shows variation of sat as a function of x. With small decrease, the increases with x for x=0.2 and 0.3 and then reduces slightly for x=0.4. Though is observed to follow Ms, that is occupancy of Mn on A-sites, up to x=0.3 for higher values of x, the does not follow this relation. This could be because of the fractional occupancy of Mn on B-sites as x increases.

IV.CONCLUSIONS

The present aim of the paper to investigate physical, magnetic and magnetostrictive properties of Ni substituted $CoFe_{2-x}Mn_xO_4$ ferrite had shown a substantial increase in the magnetostriction coefficient (λ). Fine particles of $Co_{0.9}Ni_{0.1}Mn_xFe_{2-x}O_4$ ($0 \leq x \leq 0.4$) compositions were successfully synthesized by the hydroxide co-precipitation route. Measurements on $Co_{0.9}Ni_{0.1}Fe_{2-x}Mn_xO_4$ show that CNFMO formed in the desire crystalline phase and exhibit the magnetic properties as reported.Also from the studied compositions, the composition x = 0.4 is the most suitable constituent phase for the magnetoelectric (ME) composite due to its higher magnetostriction and magnetization value. Presence of Mn on A-sites appears responsible for increase in Ms and λ with increasing x and we hope that it will show a better magnetoelectric response and may be used as a field sensor in future.

V.ACKNOWLEDGEMENT

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FIGURE

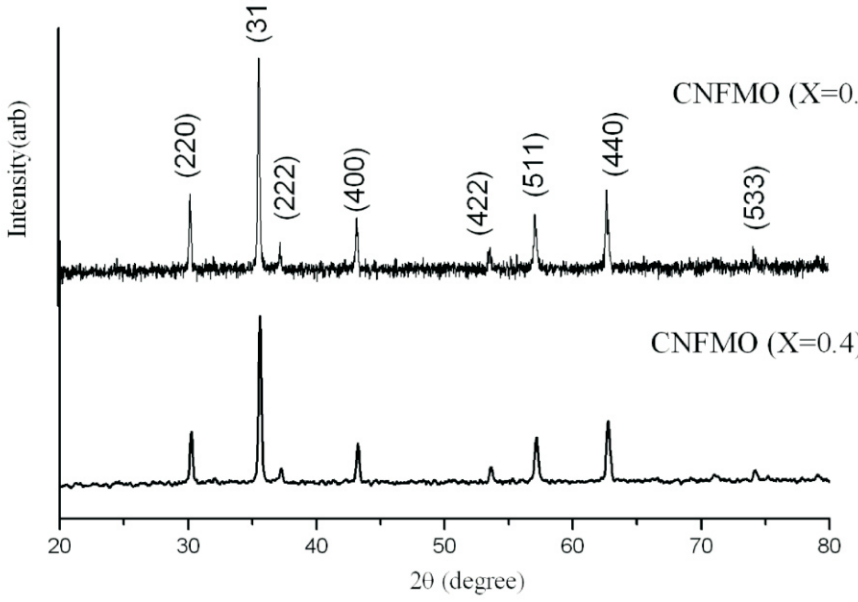


Figure 1 XRD spectrum of calcined CNFMO ferrite powder for x=0.2 and 0.4.

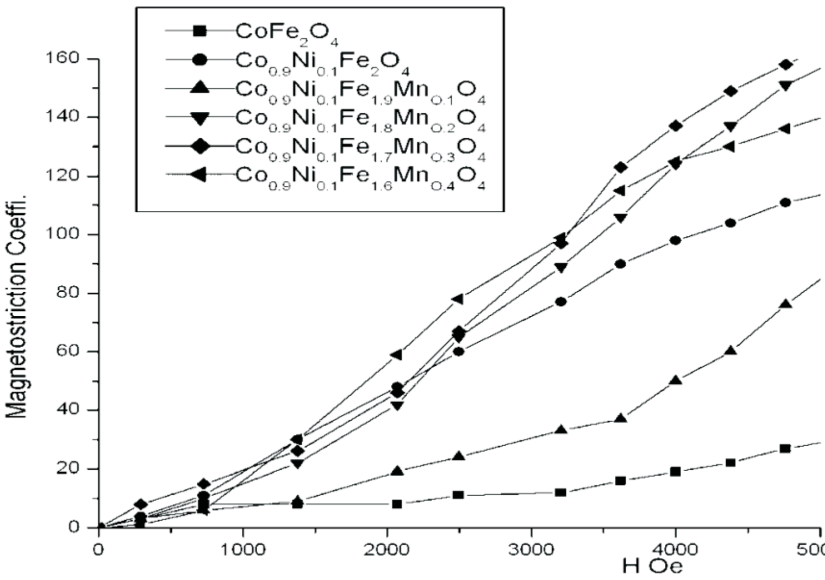
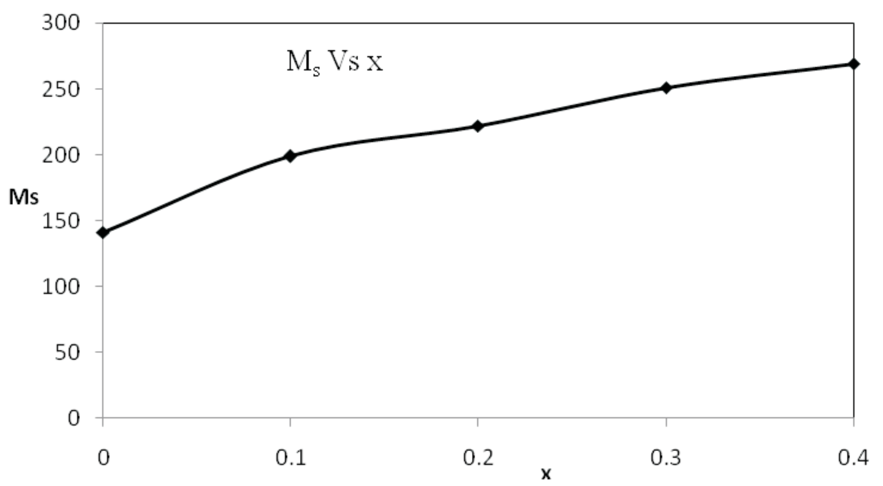
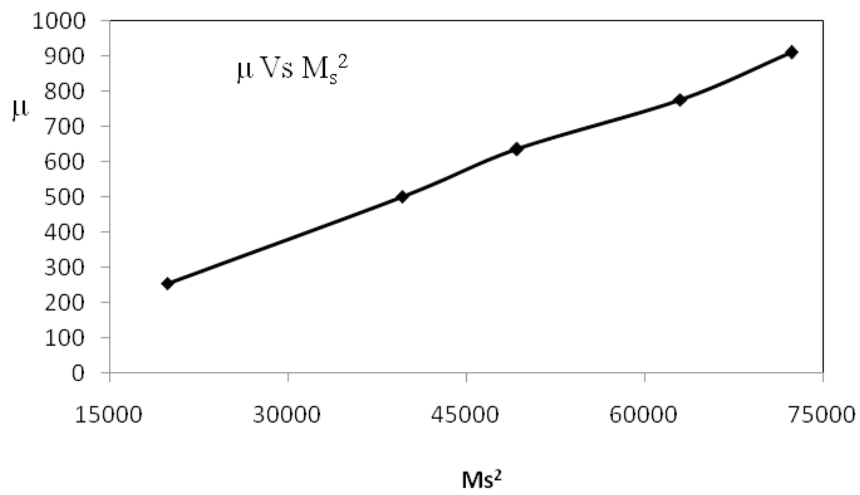


Figure 2. Variation of $\lambda_{\parallel}$ with applied magnetic field H for Cobalt ferrite and CNFMO ferrite.



(a)



(b)

Figure 3. (a) Variation of Magnetization (M_s) Vs Composition(x);
(b) Permeability (μ) Vs M_s^2.

Tables

Table 1. a: Lattice parameter, D:Crystallite size, ρdc:dc resistivity, dBulk:bulk density, dX-ray:X-ray density and p:Porosity.

CNFMO	'a' Å	Crystallite size 'D' (nm)	'ρ_{dc}' x10^6Ω-met	d_{Bulk} gm/cm^3	d_{X-ray} gm/cm^3	p (%)
x=0.0	8.18	134	67.1	5.38	5.69	5.37
x=0.1	8.23	119	12.92	5.29	5.58	5.24
x=0.2	8.25	114	10.76	5.03	5.54	9.17
x=0.3	8.28	121	2.236	5.01	5.46	8.10
x=0.4	8.31	129	0.145	5.00	5.54	9.75

Table 2. M_s: Saturation magnetization, H_c: Coeresive field, μ: Permeability, λ_{sat}: Saturation magnetostriction.

CNFMO	M_s emu/gm	H_c Oe	μ	λ_{sat} x10^{-6}
$CoFe_2O_4$	265	60	558	92
x=0	141	100	255	115
x=0.1	199	112	502	90
x=0.2	222	112	636	160
x=0.3	252	93	777	167
x=0.4	269	81	911	142

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