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PREPARATION AND STUDY OF MAGNETIC PROPERTIES OF Gd^{3+} SUBSTITUTED MAGNESIUM-ZINC FERRITES

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ABSTRACT: Mixed ferrites belonging to the type Magnesium - Zinc ferrite substituted with rare earth ion Gd^{3+} are investigated. X-ray diffraction study of these compositions revealed the formation of single phase spinels. Study of initial permeability with temperature revealed that the long range ferromagnetic ordering in the compounds having x = 0.4. The sample with x ≤ 0.4,and y = 0.05 and 0.10 show peaking behavior near Curie temperature. Temperature dependence normalized AC susceptibility and Curie temperature of spinel ferrites study reveals that $MgFe_2O_4$ exhibits multi domain (MD) structure with high Curie temperature while on substitution of Zn^{2+}, MD to SD transitions occurs.

***Keywords*:** Permeability, susceptibility, Mg-Zn ferrites, Gadolium substitution.

INTRODUCTION

Structure-sensitive properties are those that are drastically affected by impurities. These elements tend to locate at interstitial sites in the crystalline lattice and consequently the lattice can be severely strained. As a result small concentrations of these elements can have large effects on some of the magnetic properties of the materials. Permeability, coercivity, hysteresis losses, remanence, and magnetic stability are all considered to be structure sensitive. The structure sensitive properties are controlled through processing of the material including mechanical and thermal treatments. Of all the ferrites, magnesium based mixed ferrites are the most commercially important class of material. There has been renewed interest in the last few years with increasing possibilities of applications in the field of technology on magnetic properties (Iyer, 2009),. Magnetic properties like permeability and susceptibility depends on grain size, grain boundary and domain structure (Globus et al., 1973). Domain wall motion and spin rotation contributes to permeability. Zn^{2+} and Cd^{2+} in spinel ferrite are considered as interesting substitutions in the literature (Chun Han et al,1995, Kolekar et al, 1994 and Bhosale 1995). Mixed ferrites as Mg-Mn (Dale Owens, 1956), Mg-Zn (Sugimoto Mitsuo, 1999) have numerous applications in electronic and microwave fields. The magnetic properties of mixed ferrites Mg-Zn (Ladgaonkar et al., 2000), Mg-Cd. (Karche, 1997), Mg-Mn (Dale Owens, 1956) and Mg-Cu-Zn (Bhosale et al., 1998) have been already studied. Trivalent substitution of Nd^{3+} ions causes a decrease in the magnetic moment of Mg-Zn ferrites (Ladgaonkar et al., 2000).

Soft ferrites consist of Multimomain (MD), single domain (SD) and superparamagnetic (SP) particle, which mainly depends on substitution (Kumar et al., 2011). These domain states can be distingushed by the technique of low field AC susceptibility (Radhakrishanamurty et al., 1978). The MD particles have domain walls (Murthy et al., 1978) and magnetic changes takes place due to domain wall (DW) motion. As particle size decreases, formation of domain walls becomes energitcally unfavorable, then it is said to be single domain (SD) particle. In these mañgnetic changes do not takes place through DW motion but require the rotation of spins resulting in larger coersivity. As the particle size further decreases, spins are affected by thermal fluctuations and the system becomes SP particle. SP particle nature reduces magnetic character of the material. Cd^{2+} substitution is interesting substitutioin in the spinels (Kolekar et al., 1994). Addition of Cr^{3+} in $NiFe_2O_4$ the domain structure changes from MD to SD (Ghatage et al., 1996). Al^{3+} substitued mixed Cu-Cd ferrites exhibit mixture of SD and MD particles (Suryawanshi et al., 1999). Magnetic properties of Cr-substituted Co-ferrite nano particles synthesized by citrate-gel auto combustion method were studied by (Raghasudha et al., 2013).

In the present investigation the efforts are made to understand the effect of Gd^{3+} substitution on permeability and susceptibility of Mg-Zn ferrite.

Experimental

Generally a number of techniques were used to prepare the ferrite powder including Ceramic synthesis, co-precipitation method, tartrate precursor method, hydrothermal method, combustion, auto combustion and sol gel technique etc. The standard ceramic method is widely used and is applicable for industry for preparation ferrite samples in bulk form. Polycrystalline ferrites with general formula $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$ (x=0, 0.2, 0.4, 0.6, 0.8 and 1.0; y= 0.05 and 0.10) were prepared by standard ceramic method using AR grade oxides of Fe_2O_3, MgO, ZnO and Gd_2O_3.The weighted oxides were mixed together and wet milled using acetone base. Dried powder of samples was heated at 700 ^{0}C for 10 hours and sintered at 1050 ^{0}C for 24 hours, cooled and grinded into powder. The pellets of samples were formed by applying 10^6 Kg cm^{-2} using hydraulic press. The pellets were again sintered at 1050 ^{0}C for 24 hours for better compaction. The powdered samples were characterized by XRD on Philips computerized X-ray diffractometer (PW 3710) using Cu-Kα radiation.

RESULTS AND DISCUSSION

Characterization

The X-ray diffraction patterns of the synthesized ferrite is shown in Fig. 1 The existence of the (220), (311), (400), (422), (511) and (440) major lattice planes in the XRD patterns confirms the formation of spinel cubic structure with the Fd3m space group, which is consistent with the powder diffraction file of JCPDS. Also the presence of the (620), (533), (622), (444), (642) and (731) minor lattice planes in the XRD patterns agrees well with the powder diffraction of spinel cubic JCPDS file. From these observations all samples are considered to be formation of single-phase spinel structure. The crystalline size was calculated using Debye Scherrer's formula by using the full width at half maximum (FWHM) intensity of (311) plane of the pattern (Prithviraj Swamy et al., 2011). The lattice constant calculated increases with Zn^{2+} concentration and decreases slightly with Gd^{3+}. Such variation in the lattice constant was reported in the literature. The crystallite size and lattice constants are presented in the table 1.

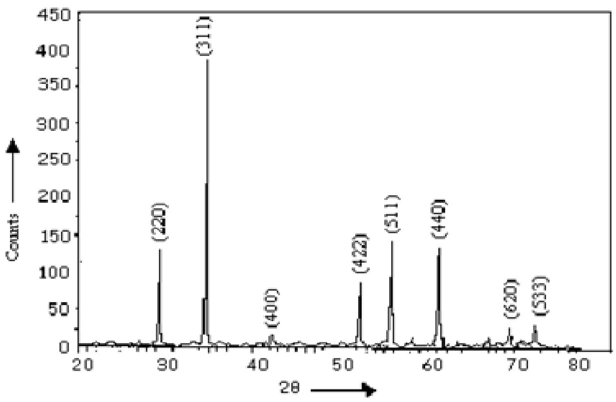

Fig1. Typical X-ray diffractogram of $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$ ferrites with x= 0.2 and y = 0.05.

Table 1. Experimental data of the ferrites $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$.

Conc.Cd^{2+} (x)	Conc.Cr^{3+} (y)	Lattice Constant ^{0}A	Crstallite Size nm	T_C from AC susceptibility measurement ^{0}K	T_C from Loria-Sinha Method ^{0}K
0.00		8.32	65	669	670
0.20		8.40	64	550	555
0.40	0.05	8.46	60	445	435
0.60		8.57	56	420	425
0.80		8.59	55	----	----
1.00		8.62	54	----	----
0.00		8.33	63	613	600
0.20		8.40	62	520	520
0.40	0.10	8.44	59	430	425
0.60		8.51	54	395	390
0.80		8.55	52	----	----
1.00		8.60	50	----	----

Initial permeability

The initial permeability (μ_i) was measured in the temperature range from 300 K to 750 K at a constant frequency of 1 KHz. The plots of μ_i versus temperature (T) are presented in Fig 2-3. The temperature dependent initial permeability (μ_i –T) curves shows increase in μ_i with the increase in temperature for the compositions x =

0, 0.2, 0.4 for y = 0.5 and 0.1. The initial permeability increases slowly up to Curie temperature, near the Curie temperature (T_c) the peaking behavior was observed. Such variation of μ_i with temperature can be attributed to the domain spin rotation as well as wall displacement. The peak in μ_i–T curves, can be attributed to zero crossing of magneto crystalline anisotropy constant K_1 (Standley, 1972). In the range of K_1 zero crossing bulging of domain wall is maximum and they become thick, the pores are reduced and μ_i increases to extremely high values (Ruess Ferrites, 1970). For the compositions with x = 0.6, y = 0.05 and 0.1, μ_i decreases exponentially with temperature, suggesting their transition temperatures are below room temperature. The compositions with x = 0.8 and 1.0 for y = 0.05 and 0.1, have no permeability showing paramagnetic nature at and above room temperature.

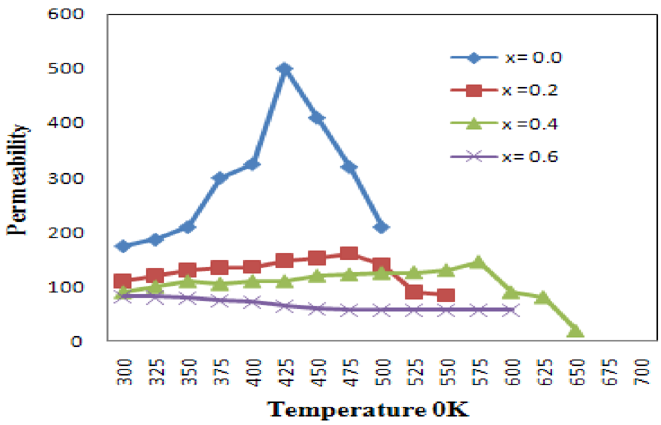

Fig 2. Plot of Permeability with Temperature (^{0}K) of $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$ ferrites with y = 0.05.

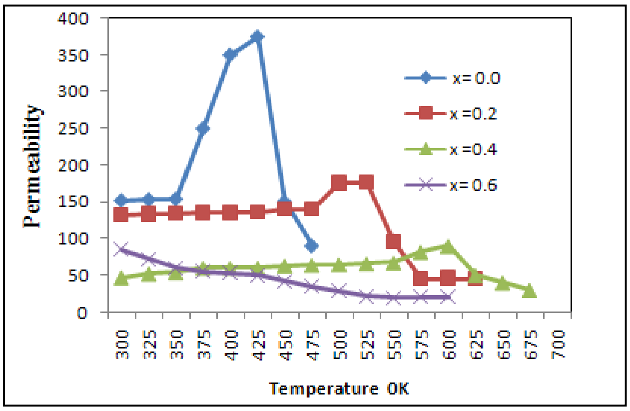

Fig 3. Plot of Permeability with Temperature (^{0}K) of $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$ ferrites with y = 0.10.

Normalized susceptiblity

The magnetic susceptibility is a dimensionless proportionality constant that indicates the degree of magnetization of a material in response to an applied magnetic field. The typical plot of normalized susceptiblity (χ/χ_{RT}) verses temperature is presented in the Fig.4. The susceptibility is found to be increases slowly and reaches peak value with temperature and then drops to zero (Sankpal et al., 1998). The increase in susceptibility with peak values suggests there is existance of multidomain(MD)particles in the material (Basu et al., 2008). The peak is found to suppresed with substitution of Gd^{3+} in $MgFe_2O_4$ and also Curie temparature (Tc) decreases with Gd^{3+} content. For the composition x=0.2; y = 0.05 and 0.1, susceptiblity is found to be independent on temperature upto Tc and after Tc it suddenly drops to zero. Such nature of curve indicates that the presence of SD particles in the materials (Basu et al., 2008). Also (Kumar et al., 2009) reported similar behaviour in Mg-Zn ferrite system. The compositions with x=0.4 and x=0.6 for y= 0.05 and 0.1 with shows exponential decrease in susceptibility indicating SD to SP transition. The composition with x=0.8 and x=1 shows paramagnetic behavior at and above room temperature. Curie tempratures (Tc) obtained from susceptibility plots are presented Table 1. On substitutiion of Zn^{2+} in $MgFe_2O_4$ Curie temperature found to decrease. Such variation was also reported (Feng Wang et al., 2013). This is because substituted Zn^{2+} ion invariably occupies tetrahedal (A) site, resulting into decrease in A-B interaction (Mariño-Castellanos et al., 2004). Substitution of Gd^{3+} ion, Curie temperature of each composition is found to decrease, this is because of decrese in strength of B-B interaction (Mariño-Castellanos et al., 2004).

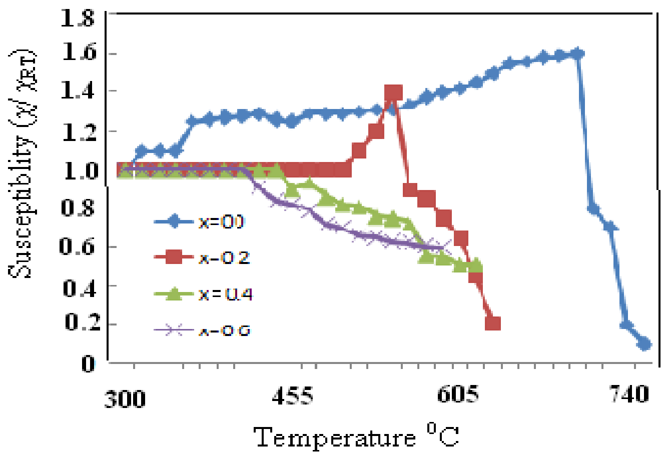

Fig 4 . The plots of normalized susceptibility (χ/χ_{RT}) verses Temperature of $Zn_xMg_{1-x}Fe_{2-y}Gd_yO_4$ ferrites with y = 0.05.

CONCLUSIONS

We have investigated experimentally how properties of spinel nickel ferrites are affected because of substitution of Zn and Gd. The crystallite size of synthesized ferrites was calculated using Scherrer"s formula in the range of 50 -68 nm. The variation of permeability with temperature suggests that there is motion of domain spin as well as domain wall with temerature. Susceptibility measurements reveals that $MgFe_2O_4$ exhibit MD particle and on substitution of Zn^{2+},domain structure changes from MD to SD and for higher concentration SD to SP. Curie temperature was found to decrease on substitution of Zn^{2+}, which is attributed to the dilution of A-B interaction. On substution of Gd^{3+}, peak obtained in the graph of normalized susceptibility of $MgFe_2O_4$ is suppresed may be attributed to the decrease in grain size and dilution of B-B interation.

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