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Magnetic and structural properties of Magnesium Zinc Ferrites synthesized at different temperature

S. S. Khot[1*], N. S. Shinde[1], B. P. Ladgaonkar[2], B. B. Kale[3] and S. C. Watawe[4]

[1]*D.B.J. College, Chiplun, Maharashtra, INDIA*
[2]*Shankarrao Mohite Mahavidhayalaya, Akluj, Solapur, Maharashtra, INDIA*
[3]*Center for Materials for Electronic Technology, Pashan, Pune*
[4]*Lokmanya Tilak Institute of Postgraduate Teaching and Research, Gogate, Jogalekar College, Ratnagiri, Maharashtra, INDIA*

ABSTRACT

The magnetic properties of $Mg_{1-x}Zn_xFe_2O_4$ (where x = 0.3,0.4,0.5,0.6) ferrites have been studied. Magnesium Zinc Ferrites was synthesized by oxalate co-precipitation method at different synthesis temperature and characterized by X-ray diffraction and far IR absorption techniques, scanning Electron microscopy .The lattice parameter were computed . The X-ray diffraction studies reveal the formations of single phase cubic spinel structure.IR absorption bands are observed around 600 cm^{-1} and 400 cm^{-1} on the tetrahedral and octahedral sites respectively. Magnetization parameters such as saturation magnetization, and magnetic moment were calculated and the results are discussed with the help of the existing theories. Saturation magnetization was found to be in the range 2 emu/gm to 8.28 emu/gm when the samples were synthesized below 100ºC. The variation of A.C. susceptibility with temperature shows the existence of super paramagnetic nature. The Curie temperature was determined from the measurement of the susceptibility verses temperature. The SEM micrographs shows the uniform distribution of the particles, the average size was estimated to be 0. 350 µm.

Key words: Polycrystalline ferrites; Oxalate precursor; X-ray diffraction; Saturation magnetization; susceptibility; Curie temperature; SEM micrographs.

INTRODUCTION

Ferrites have wide range of applications depending upon their properties. The basic properties of ferrites such as structural, magnetic and electrical etc. have been the subject of tremendous interest to Physicist, Chemists and Ceramists. The academic interest in the study of ferrites is due

to the fact that they are the most important electronic and magnetic ceramics. The potential applications of ferrites in electronics, microwave and computer technologies have focused the attention of many research workers on these materials. Recently, there has been a growing interest in low temperature sintered ferrite for the application in producing multilayer type chip inductors because of its better properties at high frequencies than MnZn ferrites [1]. Among the magnetic ceramics, magnetic oxides are the most important and relevant materials, from the point of view of their applications. The low frequency applications of soft ferrites include magnetic heads, inductors and transformer cores, electromagnets, filter cores and many other applications.

Among the technically important of ferrites are the mixed Zinc ferrites with a high initial permeability .The magnetic properties of a spinel ferrite are strongly dependent on the distribution of different cations among tetrahedral (A) and octahedral (B) sites in the crystal lattice. The cation distribution of in magnesium ferrite has been studied by various authors and was found to be strongly temperature dependent [2-5]. Magnesium ferrite requires high sintering temperatures of the order of 1350 ^{0}C to achieve the desired control on the Mg^{2+} ions distribution on octahedral and tetrahedral sites [6].Most of Mg^{2+} ions are located on B sites and small fraction migrate to A-sites [7].In order to study the influence of of Zn^{2+} ion on the structure and magnetic properties of magnesium ferrite , the composition of mixed ferrite $Mg_{1-x}Zn_xFe_2O_4$ (where x = 0.3,0.4,0.5,0.6)have been prepared and reported in the present work.

It is well known that the Zn^{2+} ions show a preference for the tetrahedral sites of the spinel lattice [7,8]. So that, the distribution of cations are over octahedral and tetrahedral sites determines to a great extent the physical properties.

MATERIALS AND METHODS

The Mg Zn ferrites having general formula $Mg_{1-x}Zn_xFe_2O_4$ (where x= 0.3,0.4,0.5,0.6) were prepared by co-precipitation method at different reaction temperatures – room temperature (38^{0}C), below room temperature (10^{0}C) and above room temperature (70^{0}C). The AR grade Magnesium sulphate, zinc sulphate, and ferrous sulphate were weighed carefully on single pan microbalance (make – Conteque and L.C. – 0.001 gm) to have proper stoichiometric proportion required in the final product. The synthesis was carried out at room temperature (38^{0}C), in which 200ml distilled water was taken and magnesium (mg), zinc (Zn), and ferrous (Fe) were added in stoichiometry proportion to the water at that temperature. A clear solution was obtained. Ammonium oxalate was taken in burette and was added drop by drop until the precipitation was completed.

The precipitate was filtered through whatman filter paper No. 41. The filtrate was washed with distilled water to remove unreacted chemicals. The residue was checked for the absence of sulphates using BaCl test. The solution was maintained at same temperature. Similar reaction was carried out using ice bath below room temperature at 10^{0}C and above room temperature at 70^{0}C where the magnetic stirrer was maintained at 70^{0}C to carry out the reaction. The precipitate was dried using electric lamp. The solid state reaction was carried out in muffle furnace maintained at 600^{0}C for 6 hours, and the powders so obtained were finely ground using agate mortar to obtain fine powders. The pellets of diameter 1 cm and thickness 0.5 cm were formed

with the hydraulic press at the pressure of 9 kg/cm^2 for five minutes, for the study of saturation magnetization. The palletized samples were finally heated in a furnace at 700^{0}C for 7 hours, for hardening.

RESULTS AND DISCUSSION

3.1 X-ray analysis

The X-Ray diffraction patterns obtained for the samples $Mg_xZn_{1-x}Fe_2O_4$ using Cu Kα radiation (λ = 1.5418 AU) are shown in Fig 1-4. The (h,k,l) values which diffracts in X-ray spinels are (220), (311), (400), (422), (333) and (400) . All the planes are the allowed planes, which indicate the formation of single-phase cubic spinel structure [9].The lattice parameter were calculated using the standard relation [10] for the cubic system and presented against composition and temperature of synthesis shown in fig. 5 and 6.

The lattice parameter obtained using the XRD data is found to be in the range 8.42A° to 8.45 A°. The variation may be attributed to the ionic size difference between Mg^{2+}(0.06 nm) and Zn^{2+} ion (0.074 nm) [9] where Zn^{2+} ion replaces Mg^{2+} ion on B site. For high concentration of Zinc (X=0.6), the lattice parameter is found to decrease, which may be attributed to shifting on some Fe^{3+} ions from A site to B site for higher composition [10]. The Temperature of synthesis does not seem to show variation in lattice parameter indicating that the range of temperature chosen for synthesis does not appreciably affect the lattice parameter.

Mazen et.al[9] have synthesized the sample at temperature above 1000ºC and obtained lattice parameter 8.41Aº, Ladgaonkar et.al[10] have synthesized the sample at temperature 1000ºC and sintered for 24 hrs by using standard ceramic method and obtained lattice parameter 8.35Aº, where as in the present case the samples have been synthesized below 100ºC but the lattice parameter obtained is slightly higher. From Fig.6 it can be seen that the samples synthesized at room temperature shows largest values for lattice parameter. Hence it can be concluded that room temperature synthesis gives greater values of lattice parameter, which may affect the force constant.

3.2 Magnetization

The saturation magnetization and the magnetic moment at room temperature are obtained by using hysteresis loop. Generally it can be seen that the magnetization M increases with increasing H. The basic composition shows the lowest magnetization value and the sample of X=0.4 shows the highest magnetization value. In the present case the variation of saturation magnetization with Zn content is shown in Fig 7. From the Graph it is observed that the saturation magnetization (Ms) are found to be increasing with increasing Zn content for X=0.3 and X=0.4 obeying Neel's two sub lattice model for magnetization and decreasing for X>0.4 suggesting the existence of noncolinear spin interaction. The similar behavior of canted spin was reported by Yafet and Kittle [11].

For higher concentration of Zn, saturation magnetization decreases. This behavior may be attributed to the migration of zn^{2+} ions to B site. Also the number of Fe^{3+} ions which will decrease on the A site and increase on B site by the same amount. These replacements will weaken the net magnetization of the whole lattice [9]. The magnetization of each composition

depends on the distribution of Fe^{3+} ions among the two sites A and B, where Mg^{2+} and Zn^{2+} ions are nonmagnetic.

The sample has the value of saturation magnetization in the range 2 emu/gm to 8.28 emu /gm. Joshi et.al [12] have reported the saturation magnetization in the range 58emu/gm to 21 emu/gm for Mg-Zn Ferrites..Vital et .al[13] reported the value of saturation magnetization 88.3 emu/gm for Mg-Zn Ferrites. Pradeep et.al [14] reported the value of saturation magnetization 41.41 emu/gm for Mg-Zn.

The saturation magnetization is related with grain size. The composition X=0.4 shows smaller grain size and greater saturation magnetization.

Variation of saturation magnetization with temperature of synthesis is shown in Fig 8. It can be seen that saturation magnetization for the samples which are synthesized below room temperature (10^0C) is lower as compared to the samples which are synthesized at room temperature (38^0C) and above room temperature (70^0C). The composition X=0.3 and X=0.4 shows increase in saturation magnetization with increase in temperature of synthesis for all temperature of synthesis. The composition X=0.5 and X=0.6 shows increase in saturation magnetization for 10ºC and 38ºC temperature of synthesis and then decrease in saturation magnetization when synthesized at 70º.

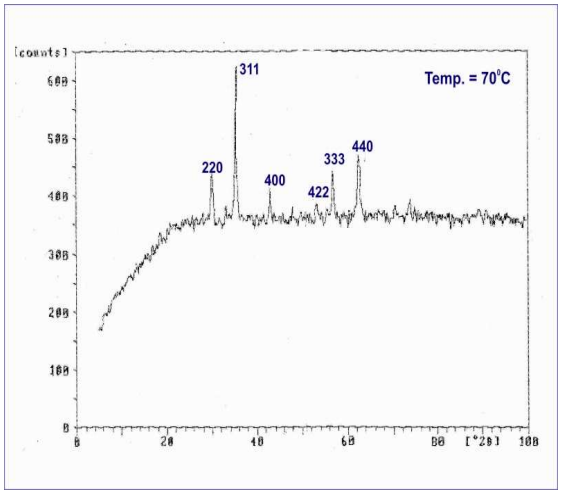
Joshi et.al have synthesized the sample at temperature above 1100ºC and obtained saturation magnetization in the range 58emu/gm to 21 emu/gm, Ladgaonkar et.al have synthesized the sample at temperature 1000ºC for 24 hrs by using standard ceramic method and obtained saturation magnetization 45.46 emu/gm to 12.80 emu/gm , Vital et.al have synthesized the sample at temperature above 900ºC by flame spray analysis and obtained saturation magnetization 88.3 emu/gm , Pradeep et al have observed value of saturation magnetization 41.41 emu/gm when the synthesize temperature is 135ºC ,where as in the present case the samples have been synthesized below 100ºC and the saturation magnetization obtained is 2 emu/gm to 8.28 emu /gm, which is smaller as compared to the other reported value due to lower temperature of synthesis.

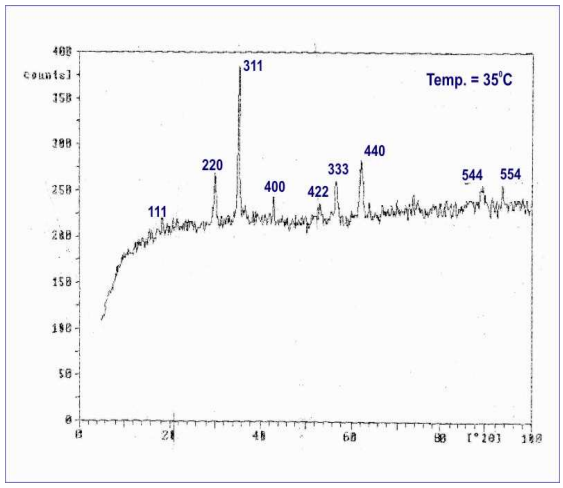
3.3 ac susceptibility

The Temperature dependence of normalized a.c susceptibility χ for the sample $Mg_{1-x}Zn_XFe_2O_4$ is shown in Fig .9-11. On inspection of this Figure, it is seen that the normalized susceptibility drops to zero in the temperature range 27ºC to 75ºC.

The plots do not show any peak with increase in temperature hence it may be concluded that the present sample exhibits superparamagnetic nature. Mazen et al [9] have observed the single domain for $Mg_{1-x}Zn_XFe_2O_4$ when samples were synthesized by ceramic technique. Joshi et.al [12] have observed ferrimagnetic behaviour for $Mg_{1-x}Zn_XFe_2O_4$ when samples were synthesized by ceramic technique. Ladgaonkar et.al [10] have observed multidomain behaviour for Mg-Zn Ferrites when samples were synthesized by standard ceramic technique. D.N.Bhosale et.al [15] have reported multidomain behaviour for Mg-Zn Ferrites when samples were synthesized by co precipitation method.

X = 0.3





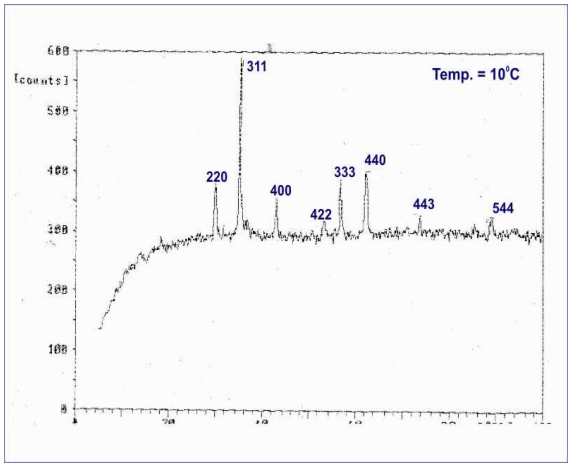
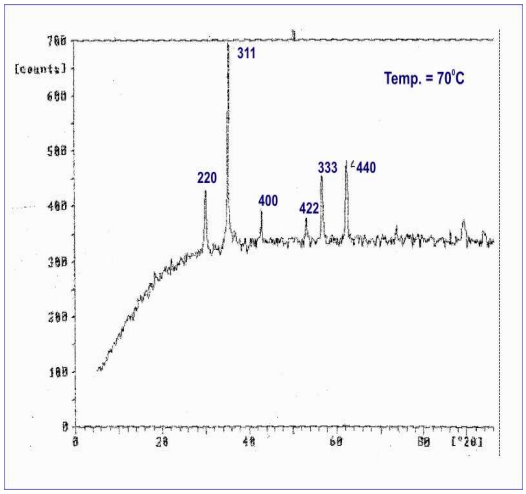
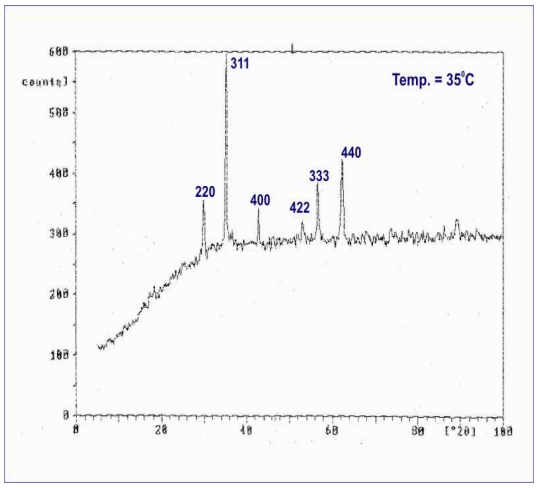


Figure 1-Variation of most intense (311) peak with temperature of chemical reaction for the composition x = 0.3

X= 0.4





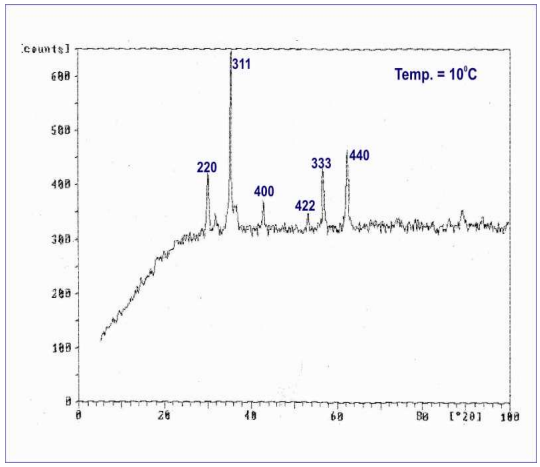
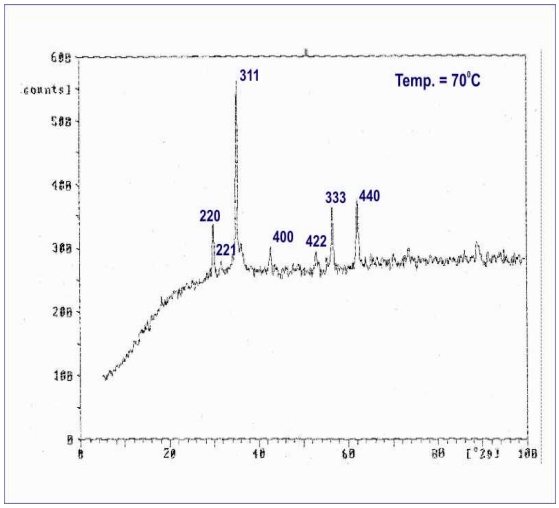
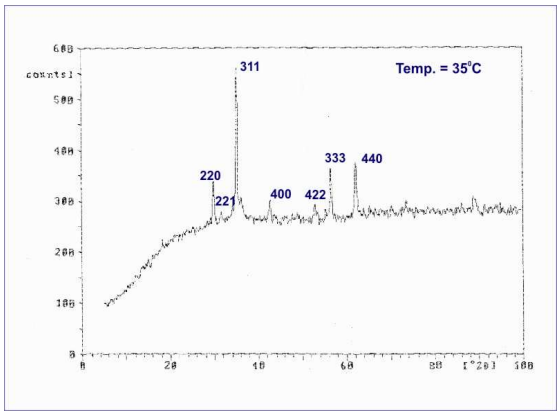


Figure 2 -Variation of most intense (311) peak with temperature of chemical reaction for the composition x = 0.4

X=0.5





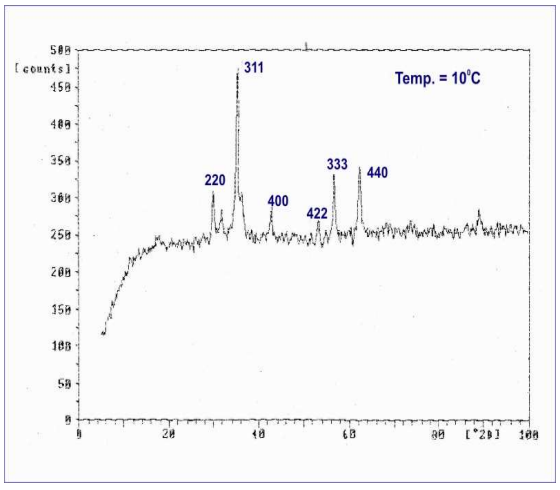
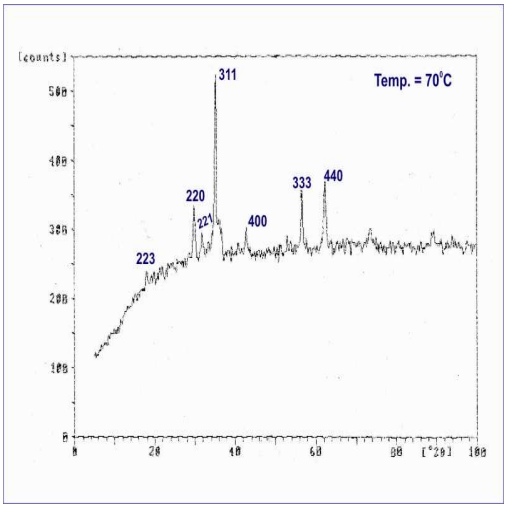
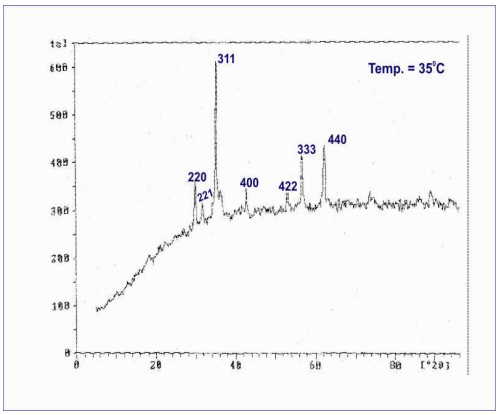


Figure 3 -Variation of most intense (311) peak with temperature of chemical reaction for the composition x = 0.5

X=0.6





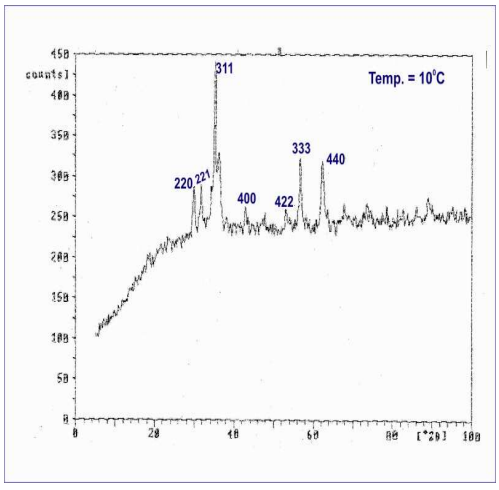


Figure 4 -Variation of most intense (311) peak with temperature of chemical reaction for the composition x = 0.6

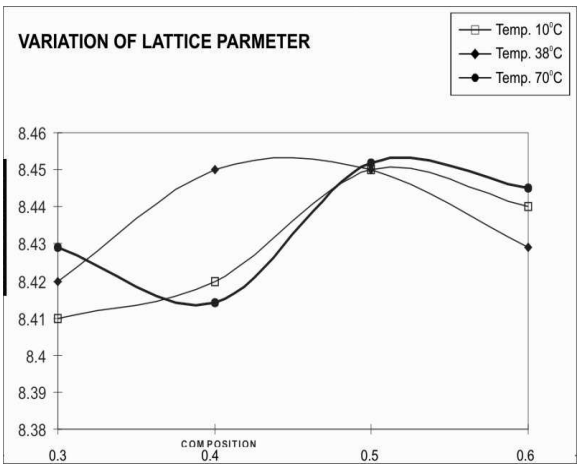


Figure 5- Variation of lattice parameter with composition

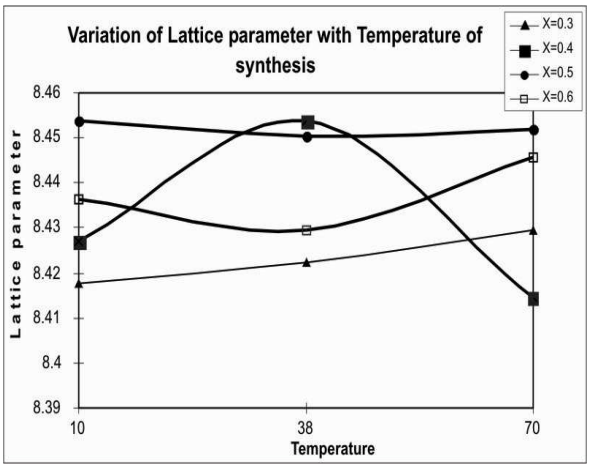


Figure 6- Variation of lattice parameter with temperature of synthesis

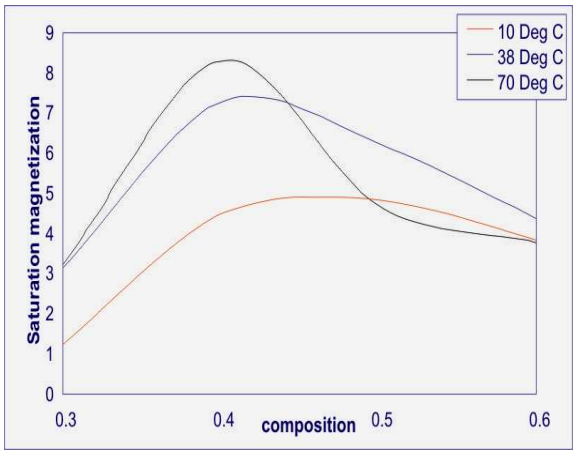


Figure 7- Variation of saturation magnetization with compositions x = 0.3,x = 0.4,x = 0.5 and x = 0.6 for the different temperature of chemical reaction

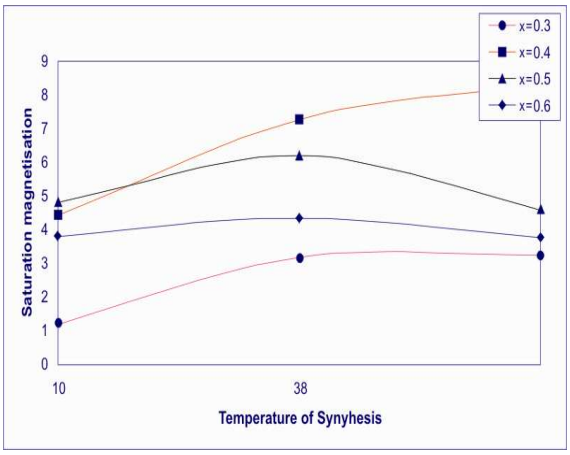


Figure 8- Variation of saturation magnetization with temperature of chemical reaction for the compositions x = 0.3,x = 0.4,x = 0.5 and x = 0.6

3.4 Curie Temperature

The Temperature dependence of normalized susceptibility χ_m for the sample $Mg_{1-x}Zn_XFe_2O_4$ is shown in Fig 12-13. Near Curie temperature the normalized Susceptibility drops to Zero. The variation of Curie temperature Tc of the sample $Mg_{1-x}Zn_XFe_2O_4$ with Zn content (X) and with the temp of synthesis is shown in Fig 12-13. It is found that the Curie temperature goes on decreasing with increasing Zn concentration. In Mg-Zn Ferrites , Mg^{2+} and Zn^{2+} are non-magnetic ions [9]. The re-placement of Mg^{2+} by Zn^{2+} ions lead to cation distribution as explained in IR study. The presence of Mg^{2+} and Zn^{2+} ions either on A site or on B-site will cause a decrease in A-B magnetic interaction thereby lowering the curie temperature .

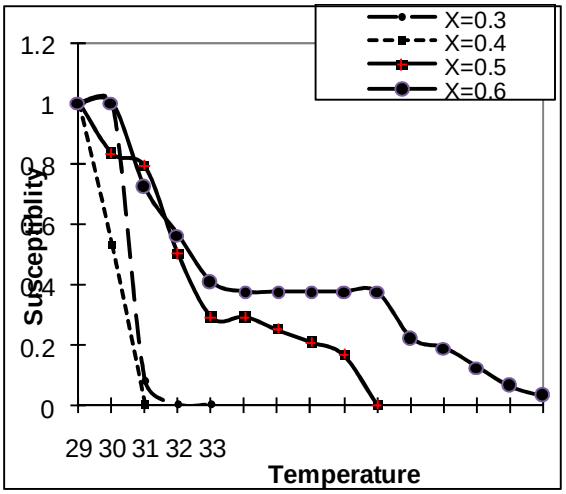


Fig-9 Variation of normalized Susceptibility of composition at 10 Deg C temperatures of synthesis

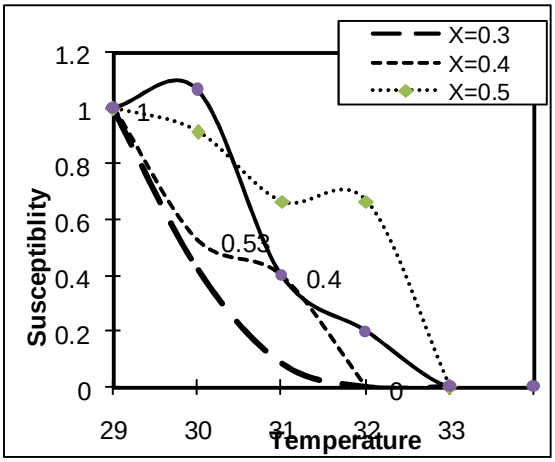


Fig-10 Variation of normalized Susceptibility of composition at 35 Deg C temperatures of synthesis

Table No 1 - Variation of magnetic properties with composition and chemical reaction temperature

Temperature of Reaction (^{0}C)	Composition Parameter X	Ms (emu/gm)	Curie Temperature (^{0}C)	Grain Size (µm)
10	0.3	1.20	46	-
	0.4	4.44	33	2.50
	0.5	4.81	31	1.52
	0.6	3.80	30	-
35	0.3	3.15	33	-
	0.4	7.26	32	0.30
	0.5	6.21	31	0.35
	0.6	4.34	35	-
70	0.3	3.21	55	-
	0.4	8.28	41	0.75
	0.5	4.63	45	0.80
	0.6	3.75	51	-

The curie temperature obtained in the present case is found to be in the range 27ºC to 75ºC. Joshi et.al [12] reported the curie temperature in the range 227ºC to 325ºC for $Mg_{1-x}Zn_XFe_2O_4$. Mazen et al [9] have observed the curie temperature in the range 127ºC to 225ºC for $Mg_{1-x}Zn_XFe_2O_4$. Vital et.al [13] reported the curie temperature 34ºC for $Mg_{1-x}Zn_XFe_2O_4$. D.N.Bhosale et.al [15] have reported the curie temperature in the range 5ºC to 27ºC for $Mg_{1-x}Zn_XFe_2O_4$.

The sample has been synthesized below room temperature and above room temperature also. As temperature of synthesis increases, the value of Curie temperature also increases. The composition X=0.3,0.5,0.6 shows lower value of Curie temperature when synthesized below room temperature and shows greater value when synthesized above room temperature. The samples for composition X= 0.4 shows the highest value of Curie temperature when it is synthesized at room temperature and above room temperature.

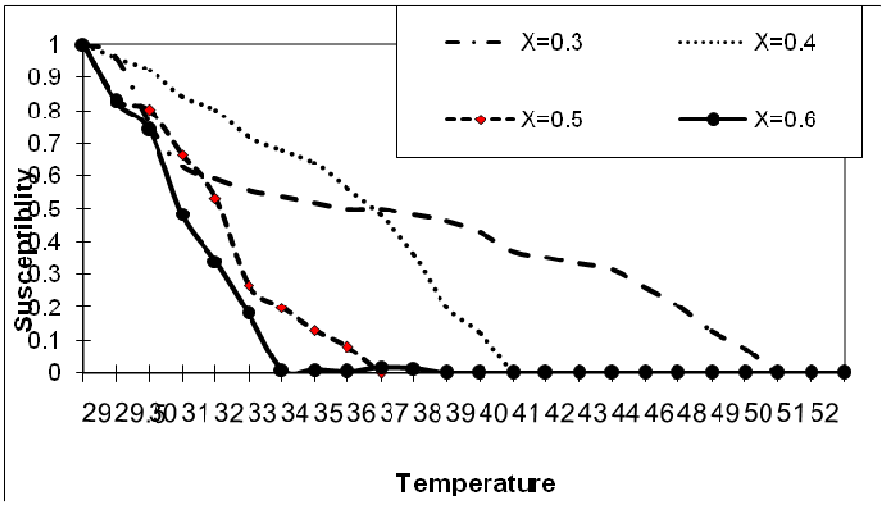


Fig-11 Variation of normalized Susceptibility of composition at 70 Deg C temperatures of synthesis for $Mg_{1-x}Zn_XFe_2O_4$

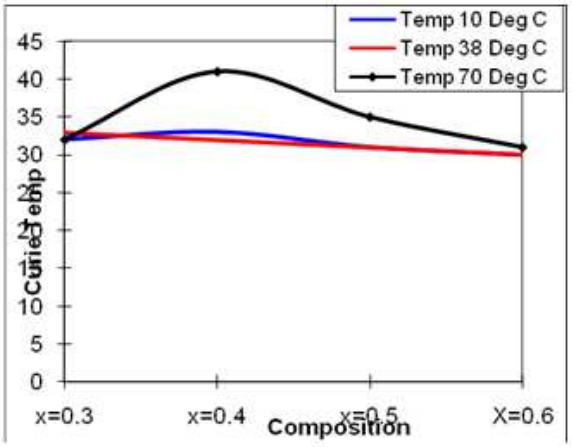


Fig.12 Variation of Curie temperature with composition for $Mg_{1-x}Zn_XFe_2O_4$

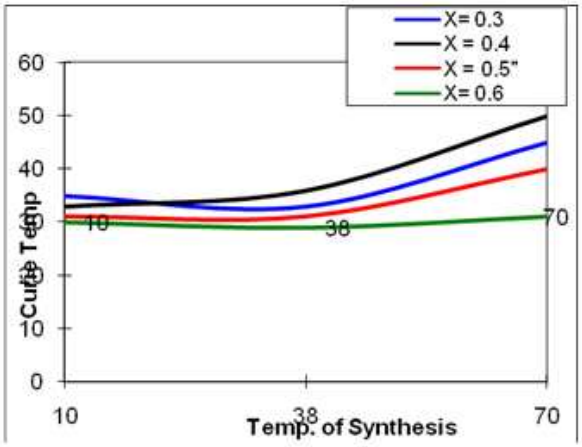


Fig.13 Variation of Curie temperature with Temperature of synthesis for $Mg_{1-x}Zn_XFe_2O_4$

The values of Curie temperature of the present sample are in good agreement with those reported by Vital et.al and D.N.Bhosale et.al for Mg-Zn Ferrites.

3.5 Scanning Electron Microscope

The SEM micrographs of the finally sintered samples of different composition are shown in fig 14., indicate the distribution of grains .The variation of the grain size of the samples with composition and temperature of synthesis are given in Fig. 14 and table 1. The grain size is greater for X=0.3 and it decreases for X=0.4 and then it increases with further increases in zinc content. The variation in grain size can be attributed to the grain growth mechanism involving diffusion coefficients, firing temperature and the concentration of dissimilar ions. [**16,17**]. The grain growth mechanism is compromised between driving force for grain boundary movement

and retarding force of pores and inclusion during the sintering process. The strength of the driving force depends on diffusivity of constituent ions [**16**].

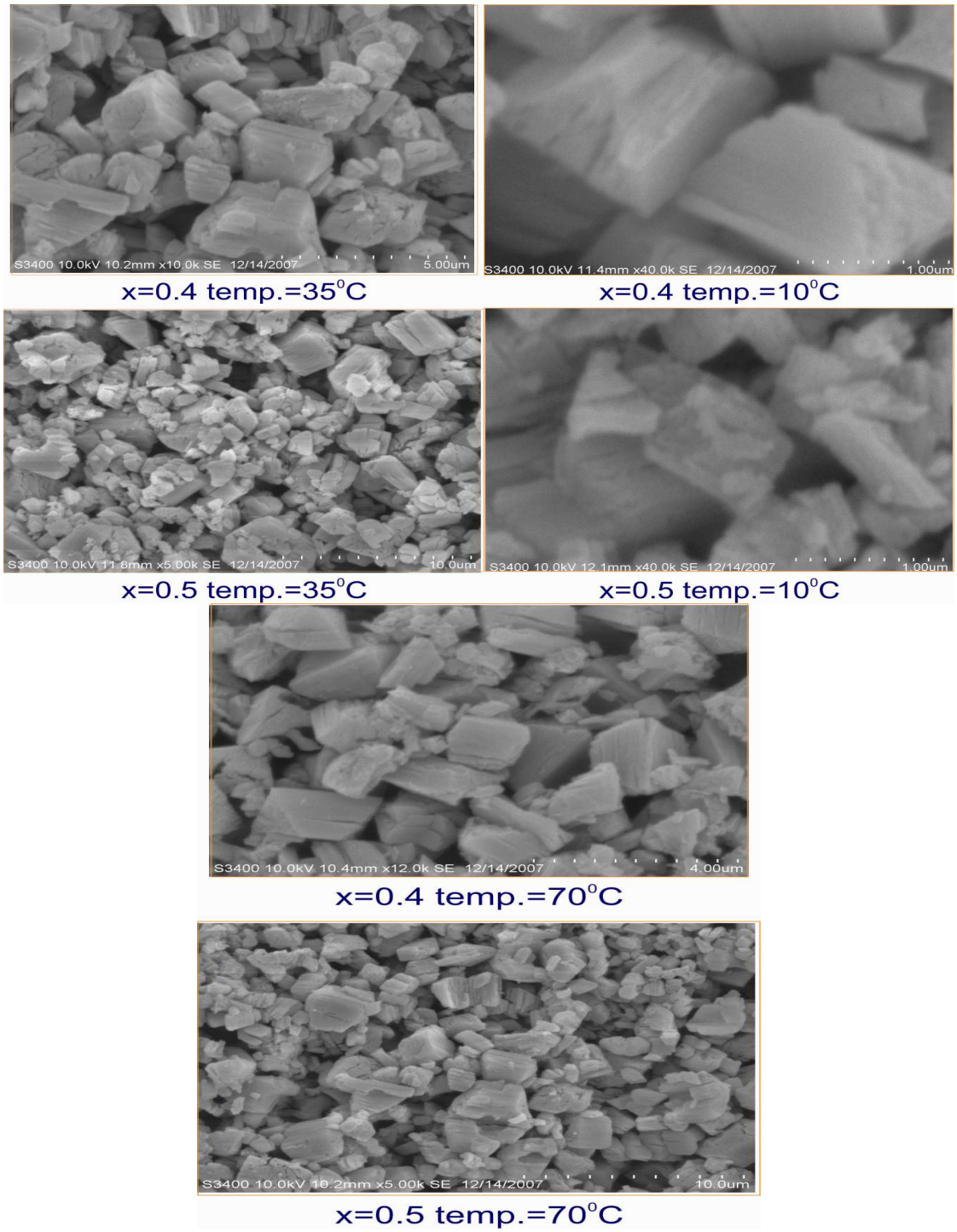


Figure 14 – SEM micrographs for X = 0.4 and X=0.5 at reaction temperatures 10 °C , 35 °C and 70 °C

The average grain size of the samples calculated using the line intercept method from the SEM micrograph, and given in the table 1 which is in the range 0.300 µm to0.335 µm. Ladgaonkar et.al.[10] also have observed the grain size in the range 1.44 µm –0.92µm for Mg-Zn Ferrites. Vital et .al [13] reported the value of grain size in the range 5.5nm to 13.5 nm for Mg-Zn Ferrites. Pradeep et.al [14] reported the value of grain size 41.9 µm for Mg-Zn Ferrites. D.N.Bhosale [15] reported the value of grain size 5.85µm for Mg-Zn Ferrites. S.C.Watawe et al [16] reported the similar feature for Lithium Ferrites.

The grain size is related with the saturation magnetization. The lower value of (Ms) is consistent with particle size. The saturation magnetization has greater value when grain size has lower value .The size effects could explain the reduced magnetic saturation. The reduction in grain size can be further attributed to both elemental composition and site occupancies of the metal cations in the oxygen lattice [16].

The samples have been synthesized below and above room temperature also, to find the effect on grain size. The grain size of the composition is also depends upon temperature of synthesis and the method employed for the preparation of sample .For lower temperature of synthesis, the grain size is more. The grain size is smaller for the composition X=0.4 and X=0.5, when synthesized at room temperature. The grain size increases with increase in temperature of synthesis therefore grain size increases up to 0.750 µm at 70ºc.

In the present case the samples have been synthesized below 100ºC and the grain size obtained is 0.300 µm to 0.335 µm, which is smaller as compared to the other reported value [13-15].

Hence it can be concluded that room temperature synthesis gives smaller grain size, which may affect the saturation magnetization.

CONCLUSION

It can be concluded that the Mg-Zn ferrite with greater values of lattice parameter, smaller value of saturation magnetization, super paramagnetic behavior, better results for curie temperature. can be synthesized at room temperature (38^0C) and above room temperature (70^0C) by employing oxalate co-precipitation method.

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