International e-Conference on Recent Trends in Nano-Materials and Its Applications
In Association with International Journal of Scientific Research in Science and Technology
Volume 9 | Issue 11 | Print ISSN: 2395-6011 | Online ISSN: 2395-602X (www.ijsrst.com)

DC Resistivity of La3+ Substituted Mg-Zn Ferrite Nanoparticles by Co-Precipitation Method

R.A. Bugad[1]*, B.G. Pawar[1], B.B. Navale[2], P.G. Pawar[3]

[1]Department of Science, Sangola Mahavidyalaya, Sangola, Dist. Solapur 413307, Maharashtra, India
[2]Department of Science, Vidnyan Mahavidyalaya, Sangola, Dist. Solapur 413307, Maharashtra, India
[3]Department of Science, Shivaji Polytechnic College, Sangola, Dist. Solapur 413307, Maharashtra, India

ABSTRACT

Lanthanum (La) substituted magnesium zinc ferrite nanoparticles with general formula Mg0.6Zn0.4La2yFe2-2yO4 (where y = 0.00, 0.05, 0.10, 0.15, 0.20 and 0.25) have been synthesized by co-precipitation method. The XRD analysis was carried out to confirm the single –phase cubic structure of La3+ substituted Mg-Zn ferrite. The nature of DC resistivity of ferrite was studied with substitution of La3+ content. The effects of La3+ substitution in Mg-Zn ferrite on structural and electric properties were studied.

Keywords: Lanthanum, Mg-Zn ferrites, Co-precipitation, DC Resistivity,

I. INTRODUCTION

Ferrites are usually non-conductive ferrimagnetic ceramic material. Most of the ferrites have a spinel structure [1]. The general formula of a spinel can be written as AB_2O_4. Nano-particles of mixed spinel ferrites have been the subject of current interest because of their interesting electric, optical and magnetic properties, which are considerably different from that of their bulk ferrites [2]. The ferrites are also widely used in high frequency cores, antennas, high frequency transformers, deflecting coil, motor generator and microwave devices such as modulators, phase shifter and circulators etc.[3]. The coercive force is related with saturation magnetization, anisotropy, internal stresses and porosity. The ferrites having low coercive force (HC) is known as Soft ferrites [4]. Generally, soft ferrite shows high electrical resistivity, superior magnetic and structural properties and hence they have low eddy current losses at high frequency [5]. Demand for electronic and computer components with high density and light weight performance is greatly increasing, which step up the demand for soft ferrites with high performance and thus contributes to the development of soft magnetic ferrites on the direction of higher frequency and lower power consumption [6].

II. EXPERIMENTAL

2.1. Synthesis of La^{3+} substituted Mg-Zn ferrite

The $Mg_{0.6}Zn_{0.4}La_{2y}Fe_{2-2y}O_4$ (where y= 0.00, 0.05, 0.10, 0.15, 0.20 & 0.25) have been prepared by the oxalate co-precipitation method as per reported in earlier literature [7]. The high purity AR grade starting materials $MgSO_4.7H_2O$, $ZnSO_4.7H_2O$, $LaSO_4.7H_2O$ and $Fe_2SO_4.7H_2O$ were used for preparation of samples. These chemicals were weighted in desired stoichiometric proportion and dissolved in distilled water. The pH of the solution was maintained at 4.8 by drop wise addition of concentrated H_2SO_4. The resulting solution was heated at 80 ^{0}C for 1 h in order to complete the ionization of metal sulfates. The precipitating regent was prepared in distilled water by adding required proportion of AR grade ammonium oxalate. Ammonium oxalate was taken in burette and was added drop by drop until the precipitation was formed. The co-precipitate product was dried and calcined at 450 ^{0}C for 5h in air. The calcined powders were milled in an agate mortar with AR grade acetone as a base. The powders were pre-sintered at 700 ^{0}C for 5h.The pre-sintered powders were pressed under hydraulic pressure of 5 tones /cm^3 to form pellet using polyvinyl alcohol as binder. Then pellets were finally sintered at 900 ^{0}C for 12h.

2.2. Characterization Techniques

XRD patterns of lanthanum substituted magnesium zinc ferrites sintered at 900^{0}C for 12h were recorded by Philips X-Ray Diffractometer model PW 1710 using Cu Ka radiation (λ = 1.5405 A^0). Two probe method was used for measurement of the dc electrical resistivity of ferrite in the temperature range 25^{0}C to 575^{0}C. The resistivity was obtained by using formula $\rho = \frac{\pi r^2}{t} \times \frac{V}{I} = \frac{\pi r^2 R}{t}$, Where, t is thickness and r is radius of the pellet in cm.

III. RESULTS AND DISCUSSIONS

3.1. XRD studies

The XRD patterns of $Mg_{0.6}Zn_{0.4}La_{2y}Fe_{2-2y}O_4$ (where y = 0.00, 0.05, 0.10, 0.15, 0.20, 0.25) ferrite system sintered at temperature 900^{0}C for 12h are shown in Fig. 1. The XRD patterns reveals the well resolved, sharp and intense peaks corresponding to planes (220), (311), (222), (400), (422), (511), (440), (620) and (533). The peaks obtained in the diffractogram closely match the data in the JCPDS file card number (04-002-5442). The XRD peak pattern corresponds to all allowed planes, which hint outs single phase cubic structure with the traces of secondary phase. Moreover, the peak at 2θ= 32.10° corresponds to plane (121) which is attributed to secondary phase for $LaFeO_3$ indexed as per ICDD file No. 01-74-9045. With increase in La content, intensity of characteristics peak (311) for Fe_2O_4 gradually decreases, while intensity of peak (121) of $LaFeO_3$ increases. It implies that the substituted La^{3+} ion has a solubility limit in the spinel lattice.

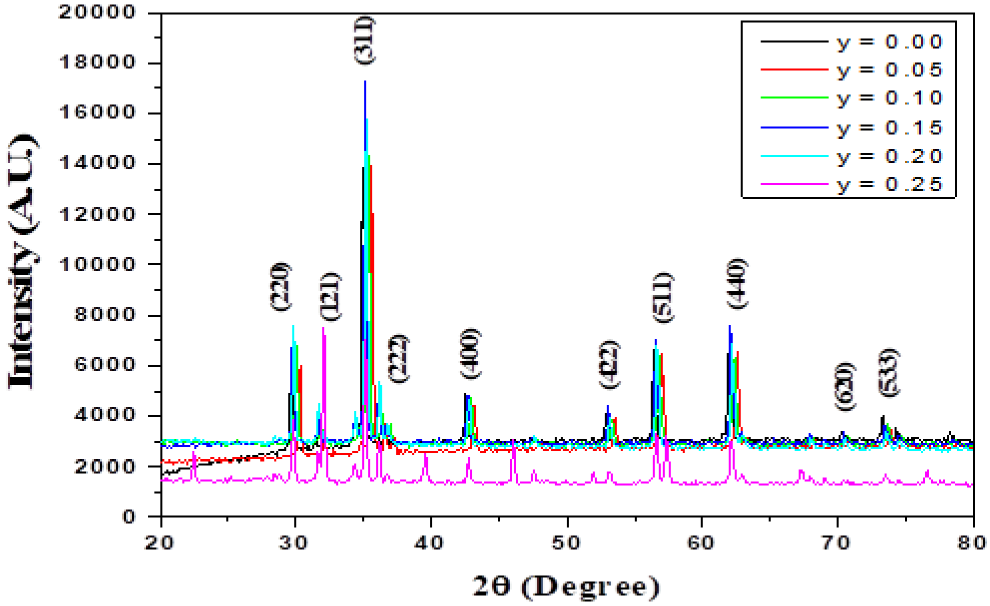

Fig.1 : XRD patterns of $Mg_{0.6}Zn_{0.4}La_{2y}Fe_{2-2y}O_4$ ferrite system

The degree of substitution of $Fe^{3+}$by La^{3+} ion is limited in the spinel lattice due to larger ionic radii of La^{3+} ions compared to Fe^{3+}ions. There is always some La^{3+} ions do not enter into spinel lattice. These La^{3+} ions react with Fe^{3+} ions and form second phase $LaFeO_3$ usually locating at the grain boundaries. Similar observation have been reported in lanthanum substituted nickel [8], Cadmium [9] and Ni-Zi ferrites [10, 11].

3.2. DC Resistivity study

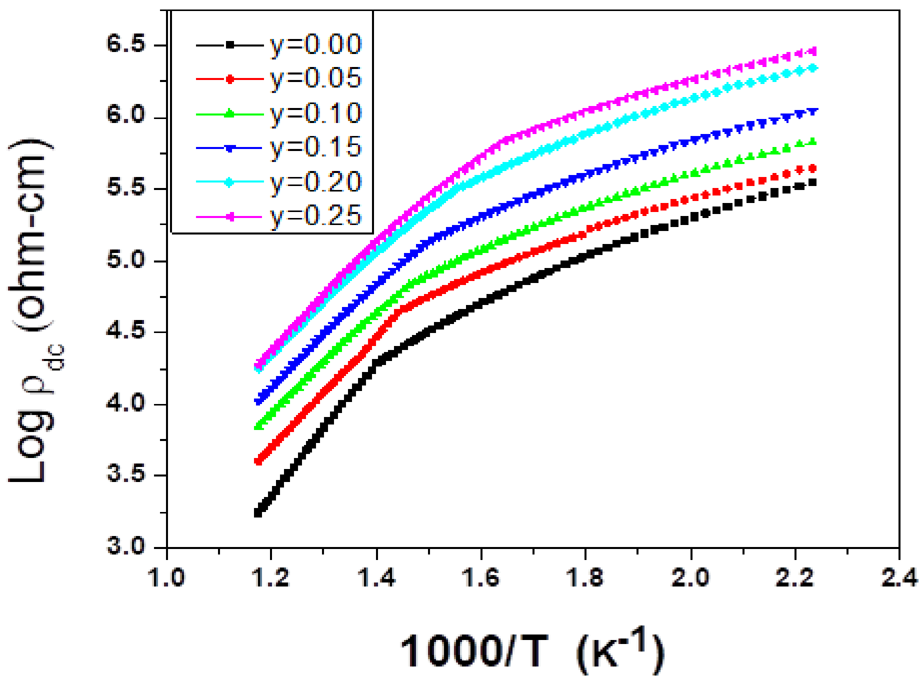

Fig.2: Variation of dc resistivity with inverse temperature for $Mg_{0.6}Zn_{0.4}La_{2y}Fe_{2-2y}O_4$ ferrite system

The variation of log of dc electrical resistivity ($\log\rho_{dc}$) as a function of inverse of temperature (1000/T) for various composition of $Mg_{0.6}Zn_{0.4}La_{2y}Fe_{2-2y}O_4$ ferrite is shown in Fig.2, The dc resistivity of the ferrites gets decreased with an increase in temperature indicates semiconducting behavior of the ferrites. All the samples show the break at Curie temperatures due to the change in conduction mechanism [12]. The slope change at Curie temperature corresponds to the samples transform from an ordered ferrimagnetic state to disordered paramagnetic state [13]. The value of Curie temperature obtained from the graph of $\log\rho_{dc}$ verses 1/T is calculated. The Curie temperature decreases with increase of La^{3+} content. It is due to the nonmagnetic nature of La^{3+} ions, which may break linkage between magnetic cations [14]. Rare earth La^{3+} ions have a strong preference to occupy on octahedral site and therefore replace Fe^{3+} ions at octahedral site (B) in spinel lattice by La^{3+}. Upon increase in the La^{3+} ions in B-site, A-B interaction weakens. Thus the decrease in Curie temperature is probably due to weakening of the A-B interaction. Similar observations have also been reported by Patil et al. [15]. The conduction phenomenon in polycrystalline ferrites was explained on the basis of Verwey and de Boer mechanism [16]. The conduction in ferrites is due to hopping of electrons between Fe^{2+} and Fe^{3+} ions on the octahedral (B) sites. In addition of lanthanum in Mg-Zn ferrite, it found that, Zn^{2+} ion have strong tendancy to occupy A-site and Mg^{2+} ion have strong preference to occupy B-site, While La^{3+} preferencelly occupy to octahedral B site, where it replaced Fe^{3+} ions in the B-site as per the modified cation distribution in magnetic study. The resistivity of the ferrite is controlled by Fe^{3+} concentration on B-site. The increase in resistivity with La^{3+} content is due to overall decrease in Fe^{3+} ions concentration on B-site. It causes decrease in hopping of electrons between Fe^{2+} and Fe^{3+} ions, results in decrease in conduction in ferrite with increase of resistivity [17]. Several researchers have been reported that resistivity of ferrites increases with the substitution of rare earth [18]. Gul and Ahmed also reported the effect of grain size, porosity and grain boundary area on resistivity of ferrite [19]. It is found that one of the factors for higher resistivity in ferrite is the decrease in grain size upon the addition of lanthanum. Smaller grain size produces larger number of insulating grain boundaries which produces inhomogeneous structure and greater energy is required for electron conduction which affects on AC and DC resistivity of ferrites [20].

IV. CONCLUSIONS

In conclusion, we report the preparation of lanthanum substituted magnesium zinc ferrites by co-precipitation method. A study on DC resistivity shows all sample have semiconducting behavior and break at Curie temperature. The conduction phenomenon in polycrystalline ferrites was explained on the basis of Verwey and de Boer mechanism. The increase in DC resistivity is due to low concentration of Fe^{2+} ions, which is responsible for decrease in electronic polarization. The increase of dc resistivity with La content was mainly attributed to decreases in drift mobility with lanthanum content. This study reveals that Lathanum substitution alters the structural and electric properties of Mg-Zn ferrites.

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