Research Article

IJCRR
Section: General Science
Sci. Journal Impact Factor 4.016
ICV: 71.54

Electrical properties of Co-Zn ferrite Doped with Silicon

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ABSTRACT

Ferrites of the composition Co0.7+x Zn0.3 Six Fe2-2x O4 (x = 0.0, 0.1, 0.15, 0.2, 0.25, 3, 4) are prepared by double sintering ceramic method. X-ray diffraction patterns of all the samples show cubic spinal phase. By comparing the calculated and observed X-ray intensity ratios I220/I400 and I400/ I422 and using site preference of various ions, the cation distribution of the present system is determined. At lower concentration of Si up to x = 0.15, the dc resistively (ρdc) increases and for x > 0.15, ρdc is found to decrease. With increase in temperature, dc resistively decreases except for higher concentration of Si (x = 0.3 and 0.4) which show increase in resistivity at lower temperature. Activation energy values show one to one correspondence with resistivity values. Curie temperature is found to decrease with increase in Si concentration.

Key Words: Lattice Parameter, D.C. Resistivity, Activation Energy

INTRODUCTION

Studies of spinel ferrites are highly relevant to modern technologies. So that the synthesis of ferrites have become an important part of modern ceramic research. Cobalt-Zinc ferrite is one of the soft ferrites used in electronic devices such as transformer core, electric motors and generators. It is observed that incorporation of diamagnetic, tetravalent and pentavalent ions in the ferrites sufficiently improves their electrical properties. To increase the resistivity and develop the desired microstructure, tetravalent ions like Ti, Sn and Si are substituted in mixed ferrites. It is observed that the resistivity increases with increase in Si^{4+} content in Ni-Cd ferrite [1]. The addition of tetravalent ions like Ti and Si in Co-Cd mixed ferrite have been studied to understand the role of these ions on the physical properties of ferrites [2]. In our previous study we have reported electrical and dielectric properties of Si substituted Co ferrite [3]. As an extension of this work, present paper reports the effect of Si^{4+} substitution on electrical properties of Co-Zn mixed ferrite.

MATERIAL AND METHODS

The ferrites with chemical formula $Co_{0.7+x}Zn_{0.3}Si_x\,Fe_{2-2x}O_4$ (Where $x = 0.0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4$) were prepared by standard double sintering ceramic technique. AR grade oxides Fe_2O_3, CoO, ZnO and SiO_2 were thoroughly mixed in required proportions. The samples were then presintered at 700^0c for 12 hr in air medium and slowly cooled to room temperature. The presintered powder was milled and sieved to a fine powder. Pellets were prepared by subjecting a small quantity of powder to a pressure of about 12×10^6 kg/m^2 by keeping it in a die of diameter 1cm and these pellets were finally sintered at 1050^0C for about 24 hr in air medium. Cooling to room temperature was carried out at the rate of 80^0c per hr.

The structural and physical properties of the prepared samples were determined by X-ray powder diffractometry technique. The X-ray diffraction patterns were recorded on a diffractometer PW710 using Cu-K radiation of wavelength 1.54060 A^0 in the range of $2\theta = 10^0$ to 90^0 and scanning rate of 1^0 per minute.

The temperature and composition dependence of dc resistively (ρ_{dc}) of all samples was studied by two probe method. For good electrical ohmic contact the pellets were polished and silver pasted on both the surfaces.

RESULTS

The room temperature X-ray diffraction patterns of all the samples of the present series confirm the single phase formation of spinel. Typical X-ray diffraction patterns of the samples with Si concentration $x = 0.2$ and $x = 0.4$ are shown

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ISSN: 2231-2196 (Print) **ISSN:** 0975-5241 (Online) **DOI:** http://dx.doi.org/10.7324/IJCRR.2017.9968
Received: 04.04.2017 **Revised:** 19.04.2017 **Accepted:** 30.04.2017

in Figure.1. The values of lattice constant ‘a’ listed in Table 1 show slow decrease with increasing Si content from x = 0.1 to 0.4.

According to site preference clergies given by Miller [5], Fe^{3+} ions get statistically distributed over B-site and A-site, Co^{2+} has a strong preference on B-site and Zn^{2+} has strong preference for A-site. Also it is reported that Si^{4+} ions have strong preference to A-site [6]. X-ray intensities were calculated using the formula suggested by Buerger [7].

$$I_{hkl} = |F_{hkl}|^2 \; P \, L_p$$

where notations have their usual meanings. By comparing the calculated and observed X-ray intensity ratios I_{220}/I_{400} and I_{400}/I_{422} and using the above stated site preference of various ions, the cation distribution of the present system can be written as

For x = 0.0 $(Zn_{0.3}\,Fe_{0.7})\,[Co_{0.7}\,Fe_{1.2}]$
For x = 0.1 to 0.4 (Co0.05 Zn0.3 Si0.05+y Fe0.6-y) [Co0.75+y Si0.05Fe1.2-y]

Where $x = 0.1 + y$, y = 0.0, 0.05, 0.1, 0.15, 0.2, 0.3

The values of dc resistivity ρ_{dc} obtained at 400 K are listed in Table 1. It is evident that dc resistively increases with increase in Si concentration up to x = 0.15 and then decreases with increase in x.

The activation energies (Eg) in ferrimagnetic and paramagnetic regions obtained from the plots of Log ρ_{dc} and temperature are listed in Table1. The activation energy values in the paramagnetic regions are higher than that in ferrimagnetic region. This is in agreement with the theory developed by Irkhin and Turvo [12].

The values of Curie temperature (Tc) determined from the resistivity plots are listed in Table 1. It is evident that the Curie temperature decreases with increasing Si concentration

DISCUSSION

Decrease in lattice constant ‘a’ with increasing Si content from x = 0.1 to 0.4 show that present samples obey Vegard’s law [4]. Usually in a solid solution of spinels within the miscibility range, a linear change in the lattice constants in observed [4]. The linear slow decrease in lattice constant in the present system can be explained on the basis of following facts.

(i) The larger Fe^{3+} ions (0.64 A^0) are replaced by smaller Si^{4+} ions (0.42 A^0) on A site so that ionic radius at A site decreases.
(ii) The smaller Fe^{3+} ions (0.64 A^0) are replaced by larger Co^{2+} (0.72 A^0) on B site so that ionic radius of B site increases.
(iii) The difference in ionic radii of Fe^{3+} and Si^{4+} (0.22 A^0) on A Site is more than the difference in the ionic radii of Fe^{3+} and Co^{2+} (0.12 A^0) on B site. Thus decrease in ionic radius of A site is greater than the increase in ionic radius of B site.

Due to complex structural composition of these ferrites, the conduction mechanism can be attributed to a combination of several processes. Fe^{2+} ions are formed due to partial reduction of small fraction of Fe^{3+} to Fe^{2+} ions due to volatilization of Zn at elevated firing temperature during sintering process [9]. In the present system, $2Fe^{3+}$ ions are replaced by Si^{4+} ions and Co^{2+} ions. Also CoO has an affinity for oxidation resulting in the formation of Co^{3+} ions. So increasing resistivity of the system with increase of Si content up x = 0.15 may be due to electron – hole compensation.

The decrease in ρ_{dc} with increase of Si content from x = 0.2 to 0.4 may be attributed to the presence of larger number of cobalt ions on B-site, which favor the hoping mechanism between Co^{2+} and Co^{3+} ions. Thus P-type, i.e. hole hoping between Co^{2+} and Co^{3+} in predominant conduction mechanism in the ferrites containing higher concentration of cobalt. These observations are consistent with the earlier reported results [10].

Variation of Logρ_{dc} with temperature (Fig.2) show decrease in resistivity with increasing temperature. This indicates semiconductor behavior having negative temperature coefficient of resistance. For higher concentrations of Si with x = 0.3 and 0.4, Log ρ_{dc} is found to increase with temperature up to about 250 K. (Figure 2.) This anomalous variation of resistively at lower temperature for Si concentration x = 0.3 and 0.4 may be due to ionic drift current. The variation of ρ_{dc} with temperature can be attributed to current due to electrons in conduction band, current due to electro hoping and ionic drift current [11]. The present results indicate that contribution from ionic drift current is more predominant in the anomalous region of ρ_{dc} for samples with Si concentration x = 0.3 and 0.4. The ionic contribution becomes negligible at higher temperatures and ρ_{dc} becomes entirely electronic.

The activation energy (Eg) values are found to correspond to the dc resistivity values of the samples. The plots of Log ρ_{dc} against reciprocal of temperature show abrupt change in the slope at the transition temperature. The change in the slope of the curves may be due to change in conduction mechanism [13] or it may be due to phase transition..

The decrease in Curie temperature Tc is because of net reduction of Fe^{3+} ions which reduces the number of active magnetic linkages

CONCLUSION

Decrease in Lattice constant is due to replacement of larger ions by smaller ions. Decrease in X-ray density shows that decrease in mass overtakes decrease in volume of unit cell. Particle size is found to be consistent with the ceramically prepared ferrites. Increase in dc resistivity for lower concentration of Si is due to electron hole compensation while decrease in ρ_{dc} at higher concentration of Si is due hole hopping between Co^{2+} and Co^{3+} ions. The anomalous variation of resistivity for higher concentration of Si is due to ionic drift current.

ACKNOWLEDGEMENT

Author acknowledges the immense help received from the scholars whose articles are cited and included in references of this manuscript. The author is also grateful to authors/editors/publishers of all those articles, journals and books from where the literature for this article has been reviewed and discussed.

Conflict of Interest: Author declare no conflict of interest.

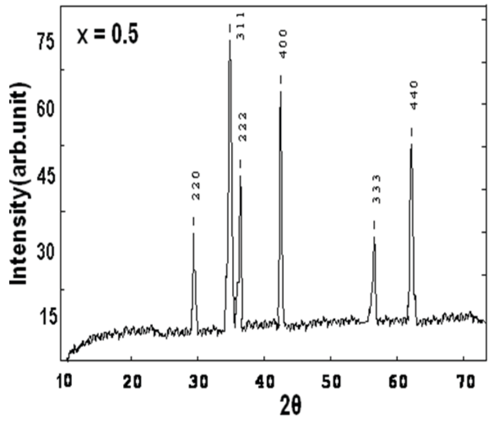

Figure 1: Typical X-ray diffractograms of the $Co_{0.7+x}Zn_{0.3}Si_xFe_{2-2x}O_4$ for x=0.1.

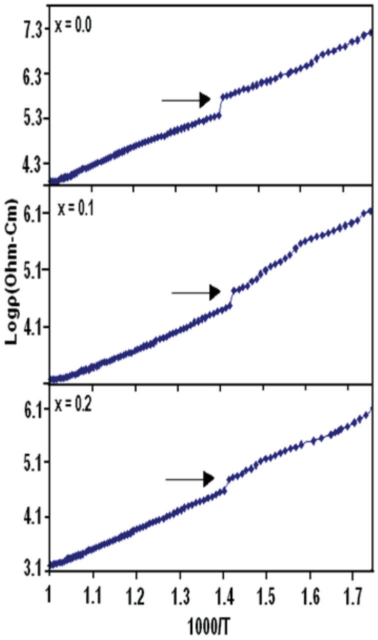

Figure 2: Plots of log ρ_{dc} vs 10^4/T for Si concentration in $Co_{0.7+x}Zn_{0.3}Si_xFe_{2-2x}O_4$.

Table 1: D.C. resistivity, activation energy and Curie temperature.

Composition. x	DC resistivity at 400 K ρ_{dc} (Ω-cm)	Activation energy ΔE (ev)		Curie temperature Tc (K)
		Para	Ferri	
0.0	1.31×10^6	0.59	0.50	630
0.10	3.53×10^6	0.63	0.63	580
0.15	3.92×10^6	0.79	0.79	576
0.20	2.84×10^6	0.71	0.71	570
0.25	1.61×10^6	0.65	0.65	563
0.30	0.34×10^6	0.55	0.55	550
0.40	0.07×10^6	0.54	0.54	525

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