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THERMOELECTRIC PROPERTIES OF (CuTi) SUBSTITUTED Ni-Zn FERRITES

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Abstract: A series of $Ni_{0.8}\ Zn_{0.2}\ Cu_x\ Ti_x\ Fe_{2-2x}\ O_4$ ferrites with x = 0.0, 0.2, 0.4, 0.6, 0.8 were prepared by ceramic sintering method. The presence of an f.c.c phase has been confirmed by x-ray diffraction. The variation of lattice constant vs. CuTi concentration shows a large positive deviation from the Vegard's law. Values of room temperature resistivity show a minimum at x = 0, that may be due to the hopping of electron between the ions $Fe^{2+} \Leftrightarrow Fe^{3+}$ and $Cu^{2+} \Leftrightarrow Cu^{1+}$. Above x = 0 the resistivity increases at higher (CuTi) concentration due to Ti^{4+} ions that act as scattering centers at B-sites. Seebeck coefficient (α) was measured and it shows that the samples are degenerate type semiconductors.

Keywords: Ceramic method, lattice constant, resistivity, thermoelectric power.

INTRODUCTION

Ferrites are ferrimagnetic materials with useful electrical properties. They have a large number of technological applications. They are used in antenna rods, inductors, magnetic cores, filters components etc. Ferrites have very low conductivity, which make them useful for microwave applications [Henaish *et al.* 1991]. The electrical resistivity of ferrites at room temperature usually depends on their chemical composition [Rana and Islam 1996] and the method of preparation. Islam *et al.* [2001] has reported electrical properties of Mg-Zn ferrites. Temperature dependent electrical resistivity was measured after final heat treatment in the temperature range 20-200°C that reveals the semi conducting nature of these ferrites. Electrical resistivity and thermoelectric power of $Cd_{0.5}Ni_{5+x}Mn_xFe_{2-2x}O_4$ have been studied by Patil *et al*. [1996]. They showed that at lower Mn concentration ($x \leq 0.2$) the increase in dc resistivity with concentration (x) could be attributed to the hindering of $Fe^{3+} \Leftrightarrow Fe^{2+}$ transition. The decrease in resistivity with higher concentration was attributed to the formation of Ni^{3+} and to the $Mn^{3+} \Leftrightarrow Mn^{4+}$ transition. The observed high value of activation energy for the system under consideration was related to Mn^{3+} Jahn-Taller trapping. The compositional variation of thermo-emf shows n-type conduction for samples with $x \leq 0.2$ and p-type conduction for samples with $x > 0.2$. The n-p type transition was explained by the formation of p-type carriers such as Ni^{3+} and Fe^{2+} cations vacancies.

Bhise *et al.* [1996] studied the dc resistivity and thermoelectric power for Mn-substituted ferrites with general formula $Zn_{0.3}Ni_{0.7+x}Mn_xFe_{2-2x}O_4$. The increase in dc resistivity at lower concentration of Mn was explained on the hindering of Verway mechanism $Fe^{2+} \Leftrightarrow Fe^{3+}$ due to stable bonds of

Mn^{3+} + Fe^{2+} pair. The decrease in resistivity at higher Mn concentration (x > 0.5) could be due to the formation of Mn^{3+} cluster and $Ni^{2+} \Leftrightarrow Ni^{3+}$ [Van Uitert 1955]. The activation energy values show one to one correspondence with resistivity values. The compositional variation of thermoelectric power shows n-type behavior for the samples with x <0.2 whereas p-type behavior for samples with $x \geq 0.2$. The p-n transition is attributed to the formation of Ni^{3+}, Fe^{2+} +vacancies which act as p-type carriers.

Patil *et al*. [1996] observed that resistivity of $Cd_{0.5}Cd0._{5+x}Ni_xFe_2O_4$ ($0.5 \leq x \leq 0.4$) ferrites increases slowly upto x = 0.2 and monotonously for x = 0.3 and 0.4. Samples show regions of activation energies with changing slope at different temperatures.

In this paper thermoelectric properties of $Ni_{0.8}Zn_{0.2}(CuTi)_xFe_{2-2x}O_4$ were measured and discussed on the basis of some conduction mechanism in these materials.

MATERIALS AND METHODS

The samples were prepared by standard ceramic sintering technique. Stoichiometric calculations were carried out using the equation:

$$0.8\ NiO + 0.2\ ZnO + x\ CuO + x\ TiO + (1-x)\ Fe_2O_3 \longrightarrow Ni_{0.8}Zn_{0.2}\ (CuTi)_xFe_{2-2x}O_4$$

where x = 0.0, 0.2, 0.4, 0.6 and 0.8 for five samples under investigation. The calculated amount of raw materials (NiO, ZnO, CuO, TiO_2, and Fe_2O_3) were thoroughly mixed and ground to obtain a good homogeneity. The pellets were formed under a load of 30KN using a hydraulic press. Heat treatment of these samples was carried out for about 25 hours at 800°C and finally all the samples were sintered at 1000°C for 58 hours. Then samples were quenched in air to obtain equilibrium for positions of the cations on A and B sites. Electrical resistivity was measured at different temperatures by two-probe method. Ohmic contacts were developed on both sides of the samples using silver paste. An electrometer model 610 C, a constant dc power supply and a sensitive digital multimeter (Keithley 197A) were used for resistivity measurements. Seebeck coefficient (α) was measured by differential method. In this method a small temperature difference ΔT (20°C) was established across the sample to induce a small thermoelectric voltage ΔV (T). The Seebeck coefficient was then determined from the relation [Islam *et al*. 2002]:

$$\alpha = \lim_{\Delta T \to 0} \Delta V\ (T)/\ \Delta T$$

RESULTS AND DISCUSSIONS

Fig. 1 shows the lattice constant vs. CuTi concentration. It can be seen that lattice constant shows a large positive deviation from Vegard's Law. This large deviation from Vegard's Law may be due to anomalous distribution of cations on A and B sites [Philips Research Report 1953].

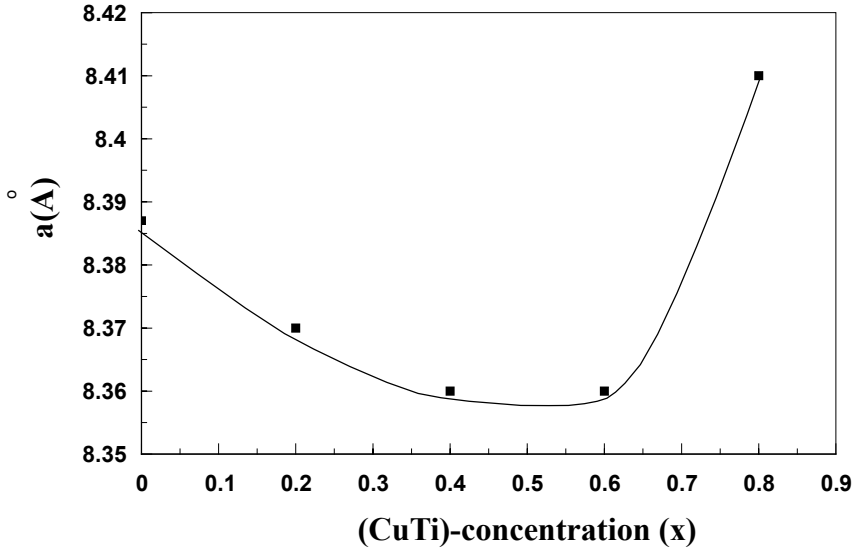

Fig.1: Lattice constant (a) vs. (CuTi) concentration for various $Ni_{0.8}Zn_{0.2}$ $(CuTi)_xFe_{2-2x}O_4$ ferrites.

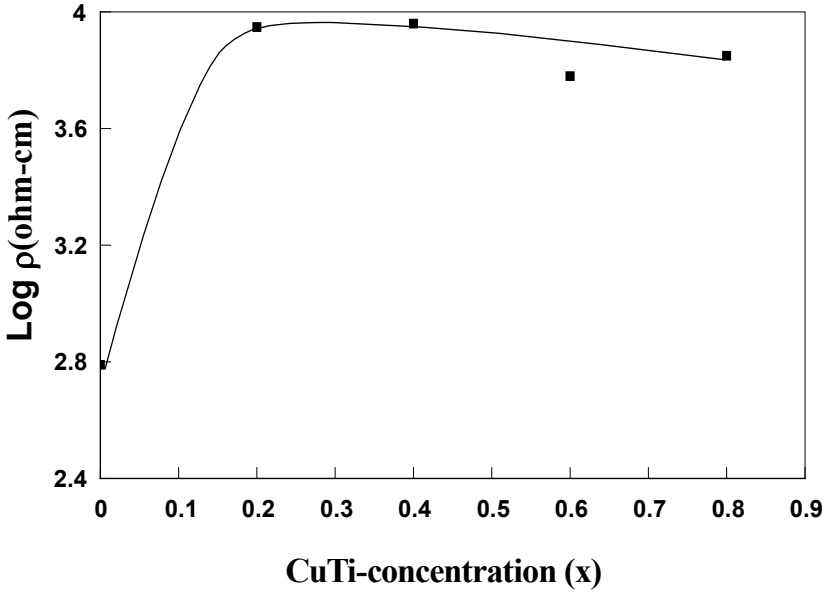

Fig. 2: Room temperature electrical resistivity (ρ) vs. (CuTi) concentration for various $Ni_{0.8}Zn_{0.2}$ $(CuTi)_x$ Fe_{2-2x} O_4 ferrites.

The variation of room temperature resistivity vs. CuTi concentration is plotted in Fig. 2. At x = 0, the value of resistivity is minimum which may be due to the hopping of electron between $Fe^{2+} \Leftrightarrow Fe^{3+}$ and $Cu^{2+} \Leftrightarrow Cu^{1+}$

states. The increase in resistivity at higher (CuTi) concentration (x > 0.0) may be due to the presence of Ti^{+4} that does not contribute to conduction but acts as a scattering center at B-sites [Bhise *et al*. 1995, Mazen *et al*. 1995]. This hinders the Verway mechanism. At x > 0.0 electrical resistivity increases which may be attributed to change of charge species from n to p type [Na *et al*. 1992, 1993, Abbas *et al.* 1995].

The thermopower (α) vs. (CuTi) content has been plotted in Fig. 3. It was observed that the thermopower is initially negative and then it becomes positive in all samples. This shows that both types of charge carriers are taking part in the conduction process. The α increases and becomes positive which is indicative of the presence of p-type carriers. It is observed that Verway mechanism is hindered in the present system with the increase of (CuTi) substitution. The variation of α with temperature shows that values of thermo-emf of all the samples depend on temperature, which in turn indicates that these samples belong to the class of degenerate type of semiconductors [Islam *et al*. 2002].

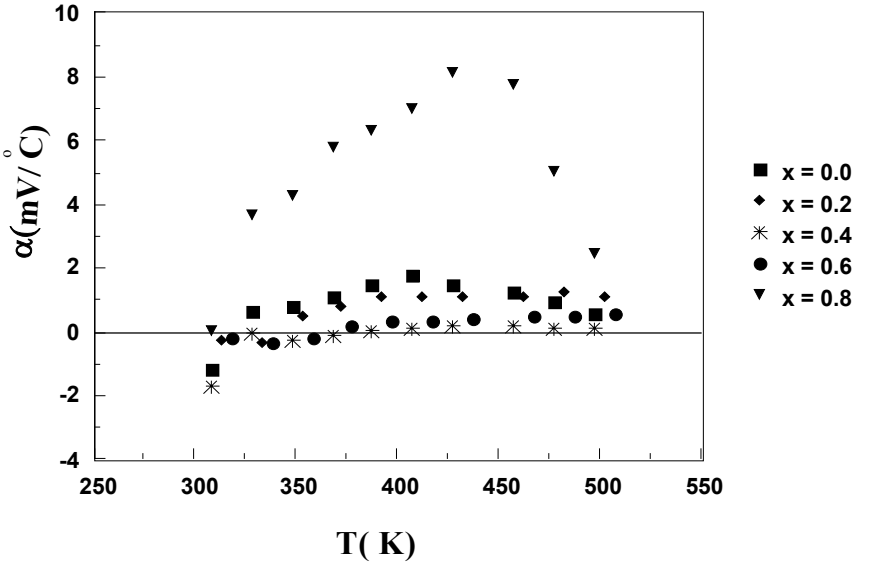

Fig. 3: Thermopower (α) vs. (CuTi) concentration for various $Ni_{0.8}Zn_{0.2}$ $(CuTi)_xFe_{2-2x}O_4$ ferrites.

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