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Thermoelectric Power Studies of Ba-Co-Zr Hexagonal Ferrites

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ABSTRACT
Thermoelectric Power studies of Ba-Co-Zr ferrites having chemical formula $BaCo_xZr_xFe_{12-2x}O_{19}$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) were investigated from room temperature to 200 ^{0}C by differential method. The Seebeck Coefficient is negative for all the compositions showing that these ferrites behave as n-type semiconductors. Seebeck coefficient (S) in all the samples is found to increase with increasing temperature. The values of the charge carrier concentration have been computed from the observed values of Seebeck coefficient. On the basis of these results an explanation for the conduction mechanism in Ba-Co-Zr mixed ferrites is suggested.
Keywords: Seebeck coefficient, Charge Carrier Concentration, Ba-Co-Zr ferrites.

I. INTRODUCTION

Ba-Co-Zr hexagonal ferrites have been considered as promising materials for applications in high-density magnetic recording and microwave devices [1, 2]. Hall effect and thermoelectric power studies are widely used in the interpretation of the conduction mechanism in semiconductors. The interpretation of Hall effect results is more straight forward and it gives precise results. However, in the case of low-mobility semiconductors such as ferrites, it is sometimes difficult to measure the Hall effect. In such cases, the thermoelectric measurement is the only alternative. Moreover, the measurement of thermo e.m.f. is simple and its sign gives vital information about the type of conduction in semiconductors, i.e., whether they are n-type or p-type As per the authors' knowledge no information is available on thermoelectric power studies of mixed Ba-Co-Zr ferrites in the literature. The present investigation deals with the study of thermoelectric power of Co-Zr substituted Ba hexaferrites as a function of composition and temperature. The results of such study are presented in this paper.

II. EXPERIMENTAL DETAILS
2.1 SAMPLE PREPARATION

Ba-Co-Zr hexagonal ferrites having the compositional formula $BaCo_xZr_xFe_{12-2x}O_{19}$ (where x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) were prepared by conventional double sintering technique. The samples were pre sintered at 1000°C for 10 hrs and final sintering was carried out at 1250°C for 15 hrs, the sintering atmosphere being air.

2.2 COMPOSITION DEPENDENCE OF LATTICE PARAMETER

X-ray diffraction patterns for all the Ba-Co-Zr hexagonal ferrites under investigation have been obtained using Co K$_\square$radiation with λ=1.7902 A^0. Fig.1 shows the X-ray diffraction pattern for Ba-Co-Zr hexagonal ferrites. The diffraction patterns confirm the presence of M-Type hexaferrite phase structure.

The lattice parameters of the mixed ferrites were calculated from d-spacing and are given in Table 1. It can be seen from the table that the values of lattice parameters a & c continuously increase with the increase of Co and Zr concentrations. This can be expected in view of the fact that the ionic radii of 0.82 A^o for the Co^{2+} ions and 0.84 A^o for the Zr^{4+} ions are larger than that of 0.64 A^o for the Fe^{3+} ions.

Fig. 2 shows the SEM micrographs of Ba-Co-Zr hexa ferrites. These micrographs are obtained by using LEO-440I model instrument. It can be seen from the micrographs that the morphology of particles is similar and they are largely agglomerated. The average grain size of the samples calculated from line intercept method from SEM micrographs is in the range of 4 to 10 μm.

2.3 MEASUREMENTS

The thermoelectric power measurements were carried out by differential method [3,4] from room temperature to 473K. The temperature gradient across the sample was measured using two pairs of copper constant thermocouples. The sample was mounted on top of two well-separated copper blocks with silver paint. The temperature difference between the two ends of the sample was kept at 10K throughout the measured temperature range. To eliminate the effects of the reference leads, the absolute thermoelectric power of Cu was subtracted from the measured thermoelectric voltage. DMV-

OO1 was used for the voltage measurements. The thermoelectric power or Seebeck coefficient S was calculated using the relation, $S = \Delta E/\Delta T$, where $\square$ E is the thermo e.m.f. produced across the sample due to the temperature difference $\square$ T.

III. RESULTS AND DISCUSSION
3.1 COMPOSITION DEPENDENCE OF THE SEEBECK COEFFICIENT

The values of the Seebeck coefficient (S) at 310 K computed from the measured values of the thermo-e.m.f. are given in Table 2. It can be seen from the table that the sign of the Seebeck coefficient is negative for all the Ba-Co-Zr ferrites. On the basis of its negative sign, the Co-Zr substituted barium ferrites have been classified as n-type semiconductors. It can also be seen from the table that the absolute value of the Seebeck coefficient decreases with the increase of Co and Zr content in barium ferrites.

3.2 TEMPERATURE DEPENDENCE OF THE THERMOELECTRIC POWER

The variation of the thermoelectric power S with temperature is shown in Figs.3. It can be seen from the figures that the value of thermoelectric power for all Ba-Co-Zr ferrites increases with increasing temperature. The negative value of the thermoelectric power found over the entire temperature range studied shows that the majority of the charge carriers are electrons. This is due to electron hoping between Fe^{3+} to Fe^{2+}.

A similar variation of the thermoelectric power with temperature was observed by several investigators in various ferrite systems [5-7].

3.3 COMPOSITION DEPENDENCE OF CARRIER CONCENTRATION (n)

Table 2 gives the experimental values of the Seebeck coefficient at room temperature. The carrier concentration for these ferrites was also calculated using the formula given by Morin and Gebella [8], $n = N \exp(-Se/K)$, where S is Seebeck coefficient, e is charge of electron, K is Boltzman constant and N is density of states or concentration of electronic levels involved in the conduction process. For low mobility semiconductors, such as ferrites, the density of states can be taken as $10^{22}/cm^3$ [8]. The computed values of the carrier concentration are presented in Table 2. It may be seen from the table that the values of the carrier concentration decrease with the increase of Co

and Zr content. This observation is in agreement with the result reported by Rezlescu *et al*. [9] who found that the resistivity of Li-Zn ferrites increased with the increase of the zinc content. Among all the ferrites the specimen with composition $BaCo_{1.0}Zr_{1.0}Fe_{10}O_{19}$ has a minimum value for the Seebeck coefficient and minimum value of carrier concentration.

IV. CONCLUSIONS

Ba-Co-Zr ferrites were prepared through soild-state route. X-ray diffractograms reveal the single phase formation and the structure is found to be hexagonal. The crystalline size of the present ferrites were found to be in the range of 4-10 $\square$ m. The Seebeck coefficient decreases with increasing the Co-Zr content in the Ba ferrite. Among all the compositions, $BaCo_{1.0}Zr_{1.0}Fe_{10}O_{19}$ showed minimum Seebeck coefficient as well as charge carrier concentration. Based on the results, the present ferrites were found to be n-type, where majority charge carriers are electrons.

V. ACKNOWLEDGEMENTS

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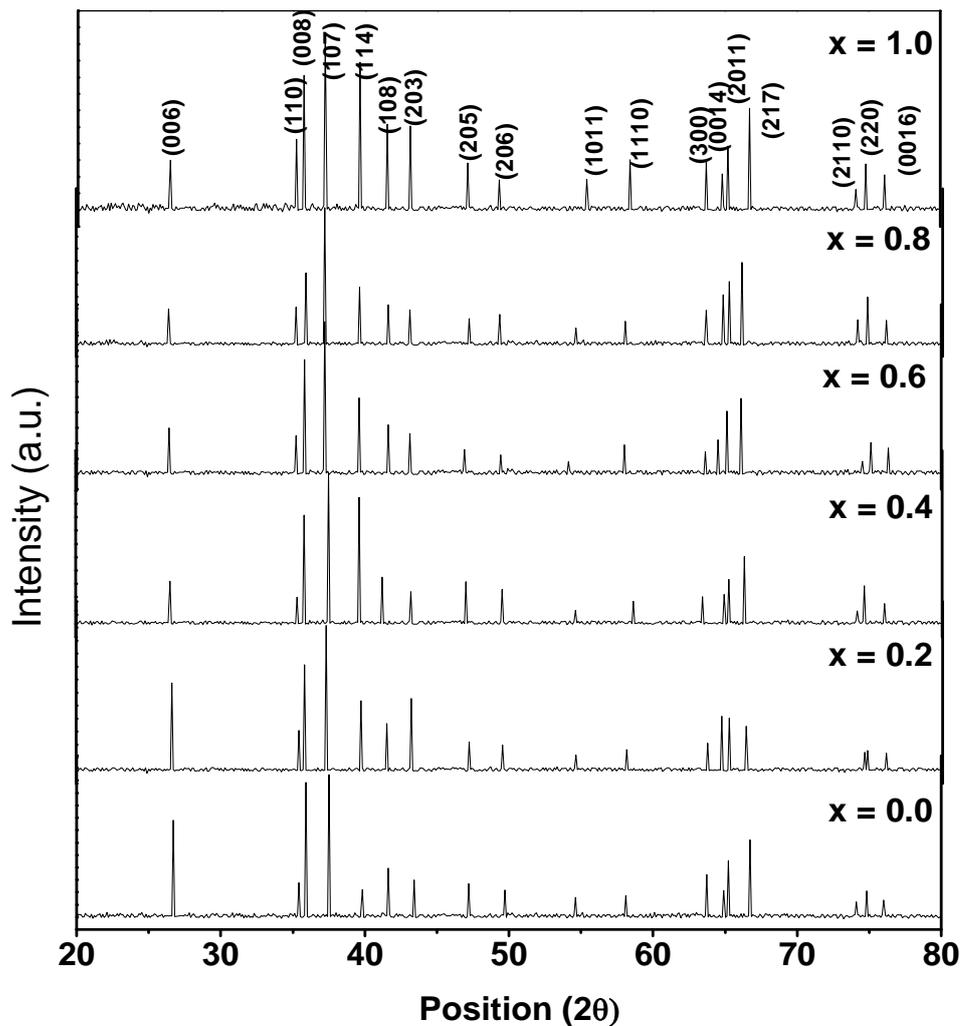
Fig. 1 : X-ray diffraction patterns of BaCo$_x$Zr$_x$Fe$_{12-2x}$O$_{19}$ hexaferrite samples

Table 1 : Lattice parameters (a & c) of BaCo$_x$Zr$_x$Fe$_{12-2x}$O$_{19}$ hexaferrites

Sl. No.	Ferrite composition	a(A^0)	c (A^0)
1	BaFe$_{12}$O$_{19}$	5.890	23.208
2	BaCo$_{0.2}$Zr$_{0.2}$Fe$_{11.6}$O$_{19}$	5.891	23.213
3	BaCo$_{0.4}$Zr$_{0.4}$Fe$_{11.2}$O$_{19}$	5.893	23.224
4	BaCo$_{0.6}$Zr$_{0.6}$Fe$_{10.8}$O$_{19}$	5.897	23.243
5	BaCo$_{0.8}$Zr$_{0.8}$Fe$_{10.4}$O$_{19}$	5.903	23.267
6	BaCo$_{1.0}$Zr$_{1.0}$Fe$_{10}$O$_{19}$	5.905	23.271
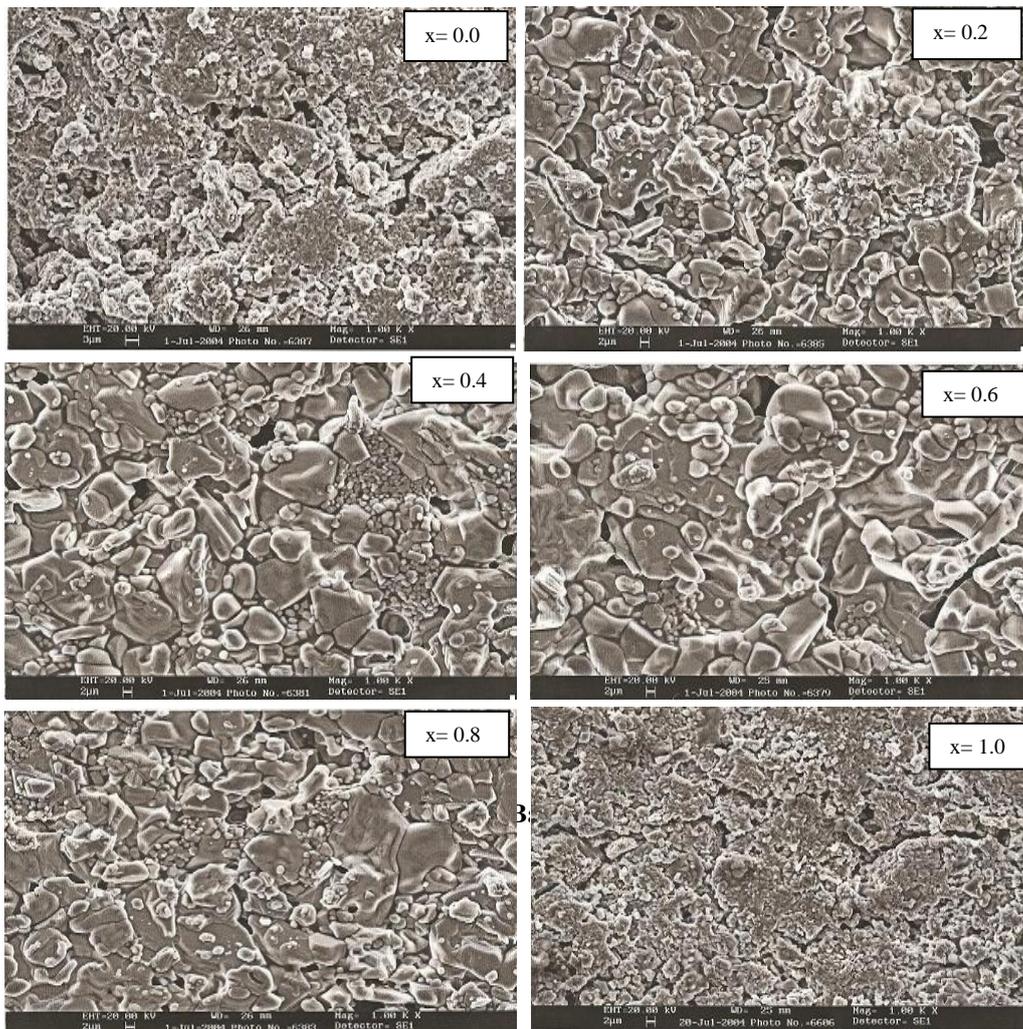
Table 2 : Electrical transport data for BaCo$_x$Zr$_x$Fe$_{12-2x}$O$_{19}$ hexaferrites at 310K

Sl. No.	Composition	Seebeck coefficient (S) μv/K	Charge carrier concentration (n) /cm^3
1	BaFe$_{12}$O$_{19}$	-137	1.13×10^{22}
2	BaCo$_{0.2}$Zr$_{0.2}$Fe$_{11.6}$O$_{19}$	-115	1.10×10^{22}
3	BaCo$_{0.4}$Zr$_{0.4}$Fe$_{11.2}$O$_{19}$	-105	1.09×10^{22}
4	BaCo$_{0.6}$Zr$_{0.6}$Fe$_{10.8}$O$_{19}$	-100	1.08×10^{22}
5	BaCo$_{0.8}$Zr$_{0.8}$Fe$_{10.4}$O$_{19}$	-85	1.06×10^{22}
6	BaCo$_{1.0}$Zr$_{1.0}$Fe$_{10}$O$_{19}$	-60	1.05×10^{22}
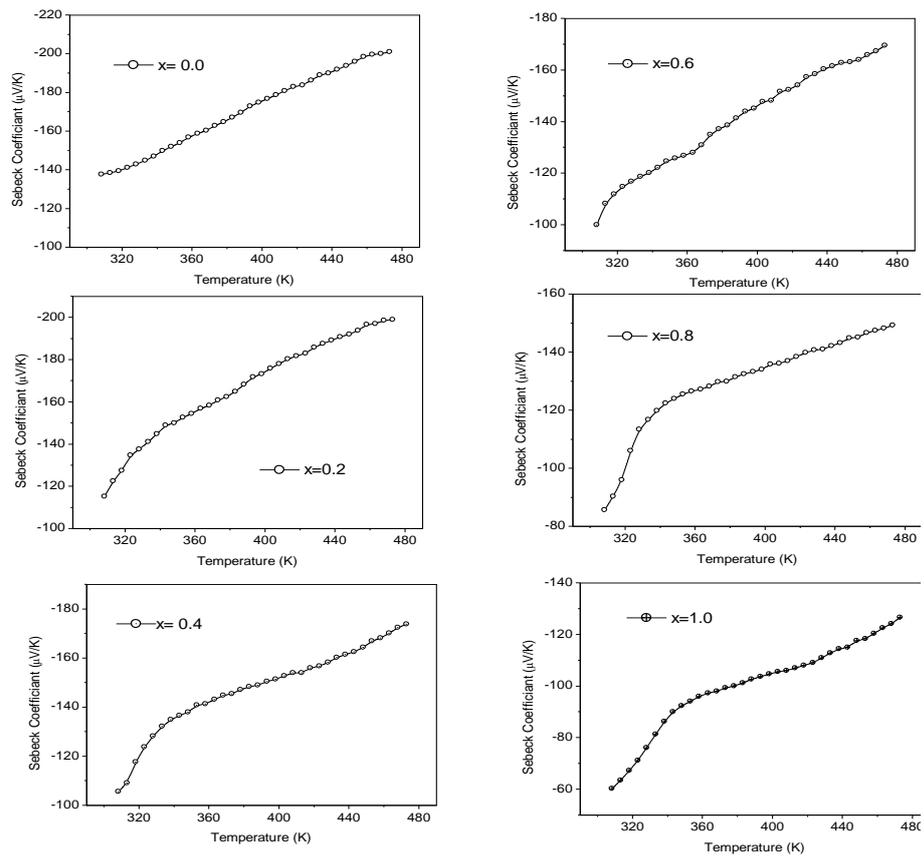
Figs. 3 : Plots of Seebeck coefficient (S) vs temperature for BaCo$_x$Zr$_x$Fe$_{12-2x}$O$_{19}$ hexaferrites