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Dielectric Behavior, Resistivity and Thermoelectric Power of Multiferroic Composite

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ABSTRACT

Composite materials were prepared by ceramic method. Initially Ferrite and ferroelectric phases were prepared separately by solid state reaction. By thoroughly mixing required mole percent of sintered phases composites were prepared. Composites were pressed into pellets. The XRD patterns of all the samples were taken by using CuKœ radiation (λ=1.5418 A^0) on Philips PW 1710 diffractometer. XRD patterns shows well defined peaks. The occurrence of the peaks with specific indices characteristic of spinel and perovskite structure confirms the formation of cubic spinel structure in ferrite and tetragonal perovskite structure in ferroelectrics. The well defined XRD peaks show crystalline nature of the samples.

Keywords: ME output, Dielectric properties, XRD, Composite.

INTRODUCTION

The existence of the magnetoelectric effect in some materials was given by Pierre Curie [1]. The magnetoelectric effect is a coupled, two –field effect in which the application of an electric field induces magnetization and a magnetic field induces electric polarization [2]. Such magnetoelectric composites are prepared by sintering together powders of piezoelectric and piezomagnetic phases. These particulate and in situ grown composites have been developed to overcome the problem of single phase magnetoelectric materials, which

have complicated crystal structures and show low outputs at low temperatures [3, 4]. In 1978, Boomgard *et. al* [3] outlined the requirements for a good ME effect in composites, which can be summarized as: **i-** the two phases should be in equilibrium; **ii-** no chemical reaction should occur between the constituent phases; **iii-** the resistivity of magneto-strictive phase should be as high as possible to avoid the leakage of current during electric poling; **iv-** a proper poling strategy should be adopted to get a large ME effect in a composite. These ME materials are used as transducers, actuators, magnetic sensors for dc and ac magnetic field measurements [5,6]. In the present work cobalt ferrite is used as it is highly magnetostrictive and resistive [7-9]. Also BaTiO3 ferroelectric has high permittivity.

PREPARATION

Piezomagnetic phase (ferrite) was prepared by solid state reaction by using CoO & Fe_2O_3 in molar proportions as starting materials. Piezoelectric phase (ferroelectric) was prepared by using BaO, PbO & TiO_2 oxides in molar proportions. The constituent phases were presintered at 900^0C. for 12 hrs. separately. After presintering, the constituent phases were ground to fine powder. ME composites were prepared by thoroughly mixing 85, 70 & 55 mole % of ferroelectric material with 15, 30 & 45 mole % of ferrite material respectively. The mixture was presintered again at 1100^0C for 24 hrs .in programmable furnace. The composites were again ground for 3hrs. to mix them thoroughly. The powder was then pressed into pellets and final sintering was carried at 1100^0C for 24hrs.

CHARACTERIZATION

The samples were characterized by X-ray Diffractometer (Philips Model PW 1710). The XRD patterns show the presence of constituent phases. The patterns do not indicate any chemical reaction between the components during sintering. The dc resistivity measurements were performed by means of a two probe method. A low value of constant voltage was applied across the sample and current through the sample was measured as a function of temperature

in the range from room temperature to 600^0C. Flat surfaces of the pellets were silver pasted for good ohmic contacts. The seebeck coefficient was measured at different temperature by maintaining the thermal gradient of 25^0C across the sample surface. The frequency dependent measurements of parameters such as capacitance and dissipation factor (tan δ) were carried out using LCR meter bridge (Model HP 4248 A) in the frequency range 20 Hz to 1MHz at room temperature. To realize the magnetoelectric voltage coefficient, the electric poling was carried out by heating the samples at about $30\text{-}40^0C$ above the ferroelectric Curie temperature in an external electric field of 2-2.5 kV/cm. The ME output $(dE/dH)_H$ was determined by measuring the electric field generated across the sample using the Keithley electrometer (Model 2000) when dc magnetic field up to 5.0 K Oe was applied to it. The variation of ME output voltage as a function of dc magnetic field was used to measure the magnetoelectric coefficient.

RESULTS AND DISCUSSION

The XRD pattern of one of the representative composite is shown in **Fig.1**. It reveals that the composites consist of ferrite and ferroelectric as predominant phases with no unidentified peaks observed. The intensity of ferrite peaks increases with its content in the composites. The indexing of patterns confirms that ferrite has cubic spinel structure and ferroelectric has tetragonal perovskite structure [10, 11]. The lattice parameters of both the phases in these composites do not vary much with the composition.

Fig. 1 - XRD of 15 % $CoFe_2O_4$ • 85 % $Ba_{0.8}Pb_{0.2}TiO_3$ Composite
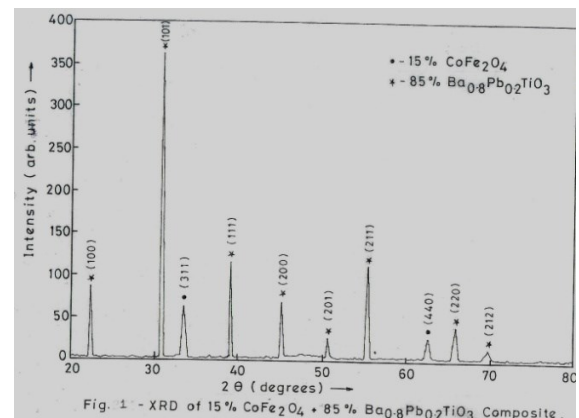
Fig. 1: The XRD pattern of one of the representative composite

The variation of dielectric constant with frequency is shown in **Fig.2**. The dielectric constant decreases with increase in frequency showing dispersion in a certain lower frequency range. It attains a constant value independent of frequency thereafter. All the samples reveal dispersion due to Maxwell-Wagner type interfacial polarization in agreement with Koop's Phenomenological theory [12].
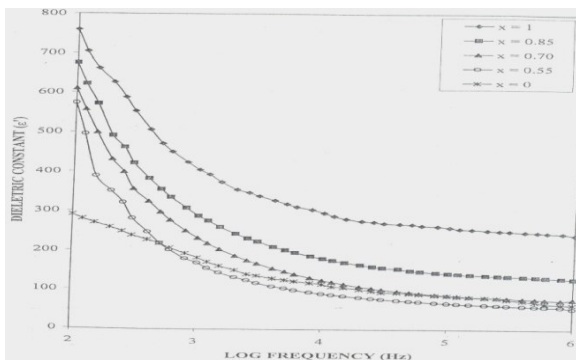

Fig.2: variation of dielectric constant with frequency for $xBa_{0.8}Pb_{0.2}TiO_3 + (1-x) CoFe_2O_4$ composites

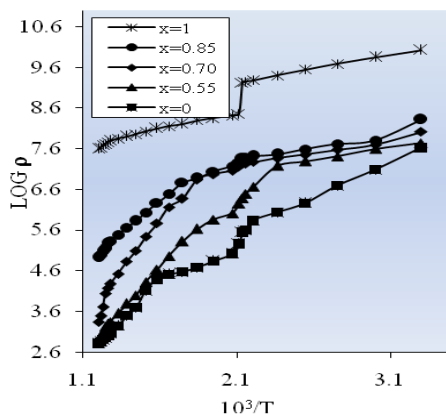**Fig.3**-variation of log resistivity (ρ) with temperature for $xBa_{0.8}Pb_{0.2}TiO_3 + (1-x)CoFe_2O_4$ composites

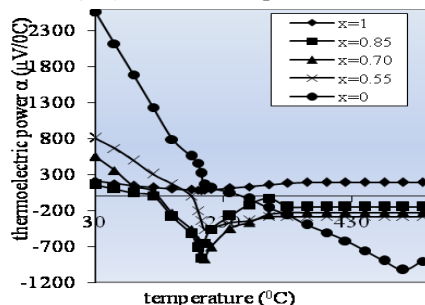**Fig.4**- variation of thermoelectric power with temperature for $xBa_{0.8}Pb_{0.2}TiO_3 + (1-x)CoFe_2O_4$ composites

The variation of resistivity with temperature is shown in **Fig.3**. There are two regions in the resistivity plots.

The first region at low temperature is attributed to the ordered state of the ferroelectric phase in the composite while the second region is for the paraelectric state of the composite. The first region indicating insulating behavior while second region indicating the conduction due to hopping mechanism. The change in activation energy is observed when transition from ferroelectric to paraelectric state takes place [13].

The variation of Seebeck coefficient (α) with temperature is shown in **Fig. 4.** At lower temperature all the samples show positive values of Seebeck coefficient, which indicates that the charge carriers are of p-type. A p-n transition is observed in all the composites and pure ferrite. It is well known that the evidence for polaron hopping conduction of p-n transition and the temperature independence of Seebeck coefficient [14].

Conflicts of interest: The authors stated that no conflicts of interest.

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