Thermoelectric Power, Conductivity and Dielectric Behavior of (x)$Ba_{0.7}Pb_{0.3}TiO_3$ + (1-x)$Ni_{0.4}Co_{0.6}Fe_2O_4$ Composite

Sukadeo. L. Kadam

Post Graduate Department of Physics, New Arts, Commerce and Science College, Parner, (Savitribai Phule Pune University), Parner, Dist. Ahmednagar, Maharashtra-414302, India

Abstract

Particulate composites with composition (x)$Ba_{0.7}Pb_{0.3}TiO_3$ +(1-x)$Ni_{0.4}Co_{0.6}Fe_2O_4$ in which X varies as 1, 0.85, 0.70, 0.55 and 0 (in mol%) were prepared by the conventional double sintering ceramic technique. The presence of two phases viz. ferromagnetic ($Ni_{0.4}Co_{0.6}Fe_2O_4$) and ferroelectric ($Ba_{0.7}Pb_{0.3}TiO_3$) was confirmed by x-ray diffraction analysis. The thermoemf measurement was carried out with variation of temperature, The ac conductivity measurement investigated in the frequency range 100Hz to 1mHz conclude that the conduction in these composites is due to small polarons. The temperature dependence of dielectric behavior at frequencies 1kHz, 10kHz, 100kHz and 1mHz was studied. Dielectric constant is maximum for 1kHz frequency.

KEYWORDS: Composite, XRD, TEP, Conductivity.

1. Introduction: Recently, electronic composite materials have attracted many researchers and led to an improvement in the piezoelectric, piezomagnetic and mechanical properties, which are applicable for a particular application [1]. The electroceramic composite materials synthesized from two different phases account for the sum, combination and product properties [2]. The composite materials of the ferroelectric and the ferromagnetic phases have the ability to show the product and sum properties. In such composite materials, electromagnetic coupling occurs and accounts for an excellent magnetoelectric effect [3-5]. They are also significant for the fundamental research on novel coupling properties such as electro-optic, electro-magnetic and other couplings [6] and have potential applications in electromagnetic interference filters, capacitors, transducers and integral chip inductors. Hence, additional efforts are necessary to study the ferroelectric-ferromagnetic materials [7].

The selection of ferrite and ferroelectric materials depends on the various factors like high magnetostriction coefficient and piezoelectric coefficient poling strength [8]. In the present work, the measurements of thermoelectric power, ac conductivity and dielectric constant are done on the composites having the general formula (x)$Ba_{0.7}Pb_{0.3}TiO_3$ +(1-x)$Ni_{0.4}Co_{0.6}Fe_2O_4$ where x=0.85, 0.70 and 0.55 to understand the conduction mechanism in the composites.

2. Experimental:

2.1. Preparation of composites:

Piezomagnetic phase (ferrite) was prepared by solid state reaction by using NiO, CoO and Fe_2O_3 in molar proportion as starting materials. Similarly, the piezoelectric phase (ferroelectric) was prepared by using PbO and TiO_2 in molar proportions. Ferrite was presintered at 700^0C and the ferroelectric at 900^0C. After presintering, the constituent phases were ground to fine powder. The composites were prepared with compositions mentioned above. The composites were again ground for

3h to mix them thoroughly. The powder was then pressed into pellets and final sintering was carried out at 1100^0C for 12h.

2.2. Characterization:

The structural analysis of the composites was done by x-ray diffraction studies using Philips x-ray diffractometer (Model PW 1710) with CuKα radiation (λ=1.5418A^0). The average particle size was calculated using Scherrer's formula.

The thermoelectric power of the composites was measured by producing a temperature gradient of $\Delta T = 20^0$C across the samples with the help of small heater attached to one of the hard electrodes of the sample holder. Silver foils are used as electrodes for measurement. The temperature gradient ΔT was measured using differential thermocouple. Thermoemf (ΔV) across the sample was measured at different ambient temperatures of the samples using digital microvoltmeter. Thermoelectric power was calculated using the relation $\alpha=\Delta V/\Delta T$.

The Ac parameters such as capacitance(c) and dissipation factor (tanδ) of the samples were measured in frequency range 100Hz to 1mHz using LCR meter Bridge (model HP 4284A). The variation of dielectric constant with temperature for four different frequencies (1 kHz, 10 kHz, 100 kHz and 1 MHz) was studied. The dielectric constant was calculated using the relation [9]. $\varepsilon= cd/\varepsilon_0 A$ ……… (1)

Where c = capacitance, d = thickness of pellet, A= area of the surface of the pellet and ε_0 permittivity of free space ($8.805*12^{-12}$ F/m).

The ac conductivity data was derive from dielectric constant and measured loss tangent using the relation [10]

6ac = $\varepsilon\varepsilon_o\omega \tan \delta$ ………….. (2)

Where ω is the angular frequency and tanδ is loss factor.

3. Result and Discussion:

The XRD pattern of one of the representative composite i.e. 70% ferroelectric phase (X=0.70) + 30% ferrite phase, is shown in fig.1. From the figure, it is revealed that both the ferroelectric as well as ferrite phases are present in the composite. No additional phases were noted. The occurrence of peaks with specific indices characteristic of spinel and perovskite structure confirms the formation of cubic spinel in ferrite phase and tetragonal perovskite structure in the ferroelectric phase [11-12].

The compositional variation of the seebeck coefficient (α) as a function of temperature is shown in fig.2. All the samples show p-type charge carriers at lower temperature. In all the composites p-n transition is observed. A negative value of α confirms n-type charge carriers and a positive value confirms p-type charge carriers. The most probable mechanism for n-type conduction is electron hopping between Fe^{3+} to Fe^{2+} ions [13,14] such as , $Fe^{2+} \leftrightarrow Fe^{3+} + e^-$

The number of such ion pairs depends upon the sintering conditions and amount of reduction of Fe^{3+} to Fe^{2+} at elevated temperatures. The resistivity of ferrite is controlled by the Fe^{2+} concentration on the B-site. The occurrence of p-n transition confirms polaron hopping conduction. To know wheather small or large polarons are responsible for the conduction, ac conductivity was studied.

The variation of log (6ac-6dc) with log ω^2 is shown in fig. 3. The plots are almost linear indicating that the conductivity increases with increasing frequency. Adler and Feinleib [15], have shown that for conduction by small polarons, the conductivity increases with frequency. It has been shown that for ionic solids, the concept of small polaron is valid [16].

The variation of dielectric constant ε with temperature of one of the representative composite i.e. 70% ferroelectric phase (x=0.7) +30% ferrite phase, is shown in fig.4. Two dielectric maximum are obtained. One at the ferroelectric Curie temperature (190^0C) for all test frequencies and another at ferromagnetic Curie temperature (520^0C) at 1 and 10 kHz test frequencies. The ferroelectric and ferrite curie temperatures for a given composite are found to be independent of test frequency. Variation in the ferrite Curie temperature from 520 to 525^0C is observed as the ferrite content changes from 30 to 45 mol%, but the ferroelectric Curie temperature remains unchanged irrespective of variations in the molar ratio of components in the composites. The absence of a dielectric maximum for the 100 kHz and 1 MHz test frequencies can be explained on the basis that the electron exchange mechanism cannot follow the applied electric field above a certain frequency [9, 17]. The electric field induced magnetic phase transition depends on the strength of interaction between electric and magnetic ordering, which in turn depends on the molar ratio of the phases.

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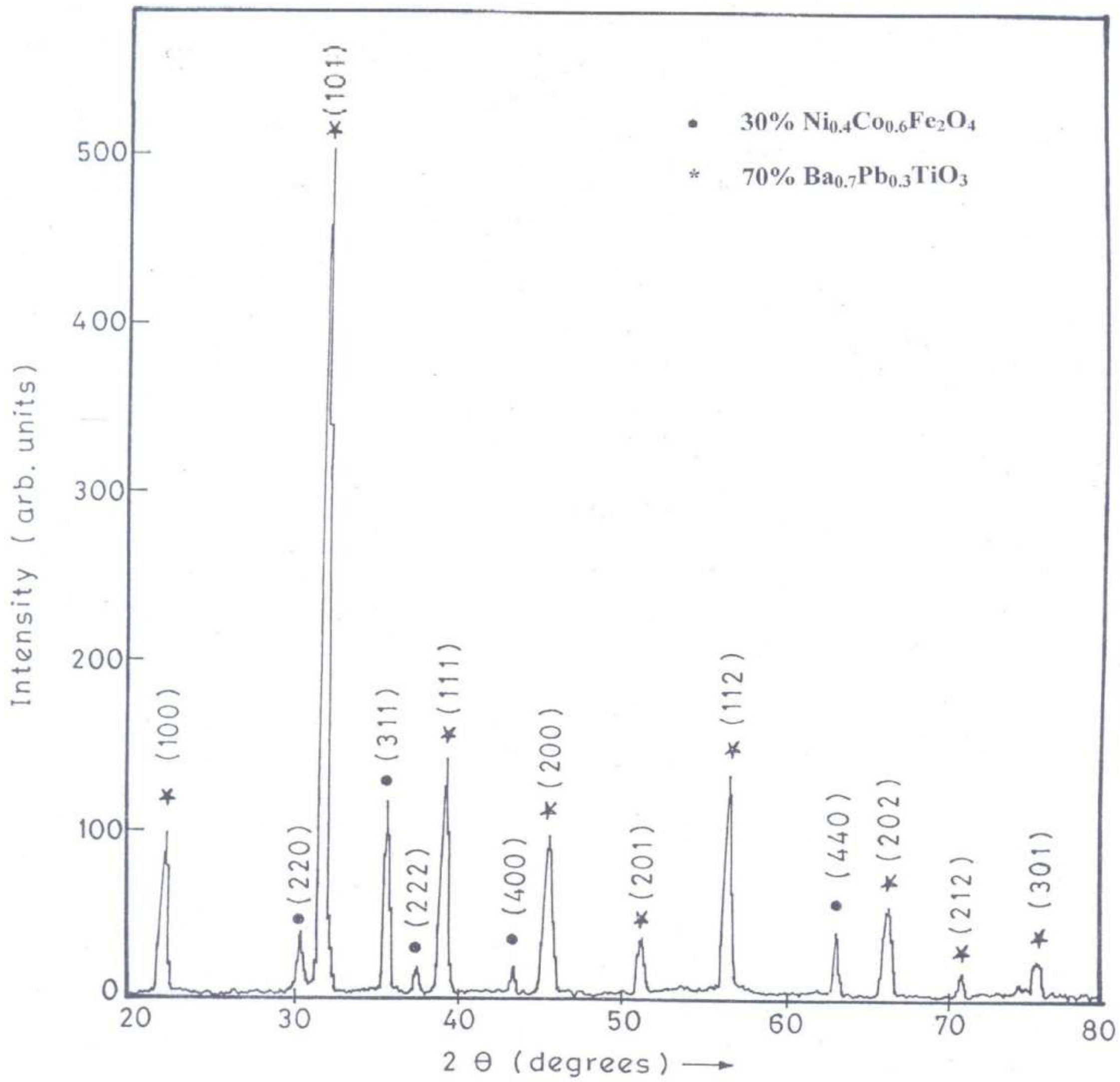

Fig. 1 XRD OF 30% $Ni_{0.4}Co_{0.6}Fe_2O_4$ + 70% $Ba_{0.7}Pb_{0.3}TiO_3$ COMPOSITE

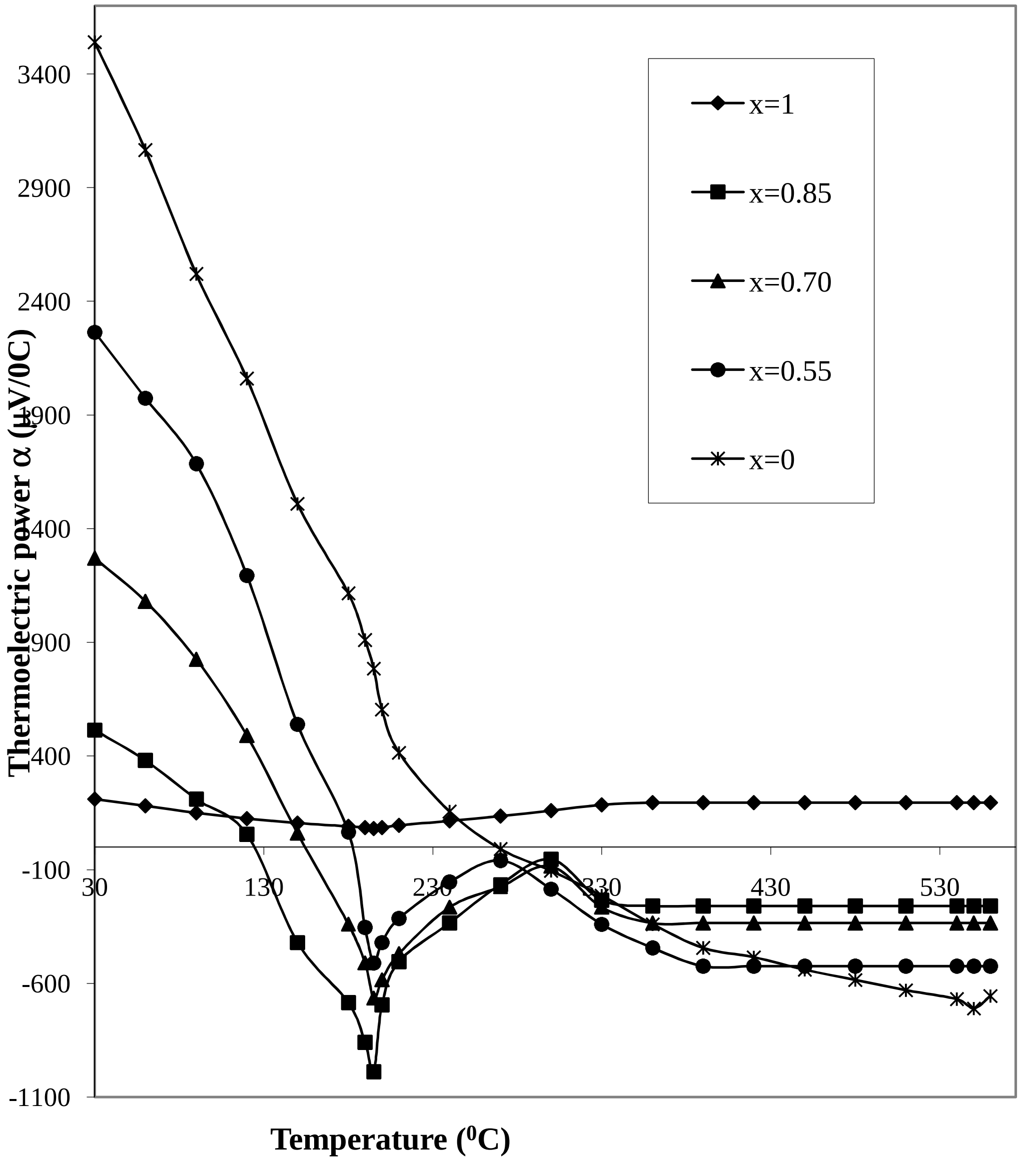

Fig. 2 **Variation of Thermoelectric Power with Temperature for $xBa_{0.7}Pb_{0.3}TiO_3 + (1\text{-}x)Ni_{0.4}Co_{0.6}Fe_2O_4$ Composites**

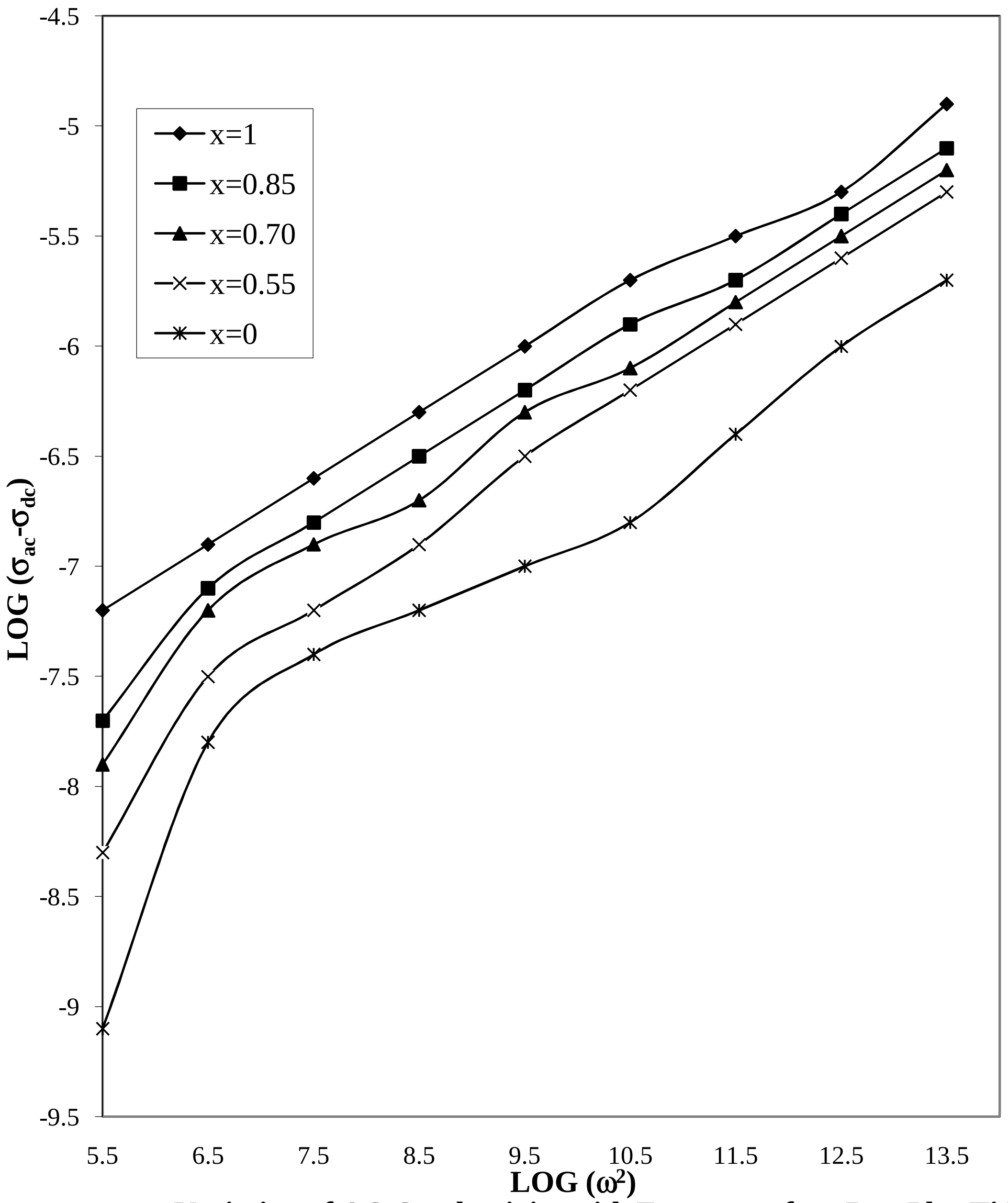

Fig.3 Variation of AC Conductivity with Frequency for $xBa_{0.7}Pb_{0.3}TiO_3$ + (1-x)$Ni_{0.4}Co_{0.6}Fe_2O_4$ Composites

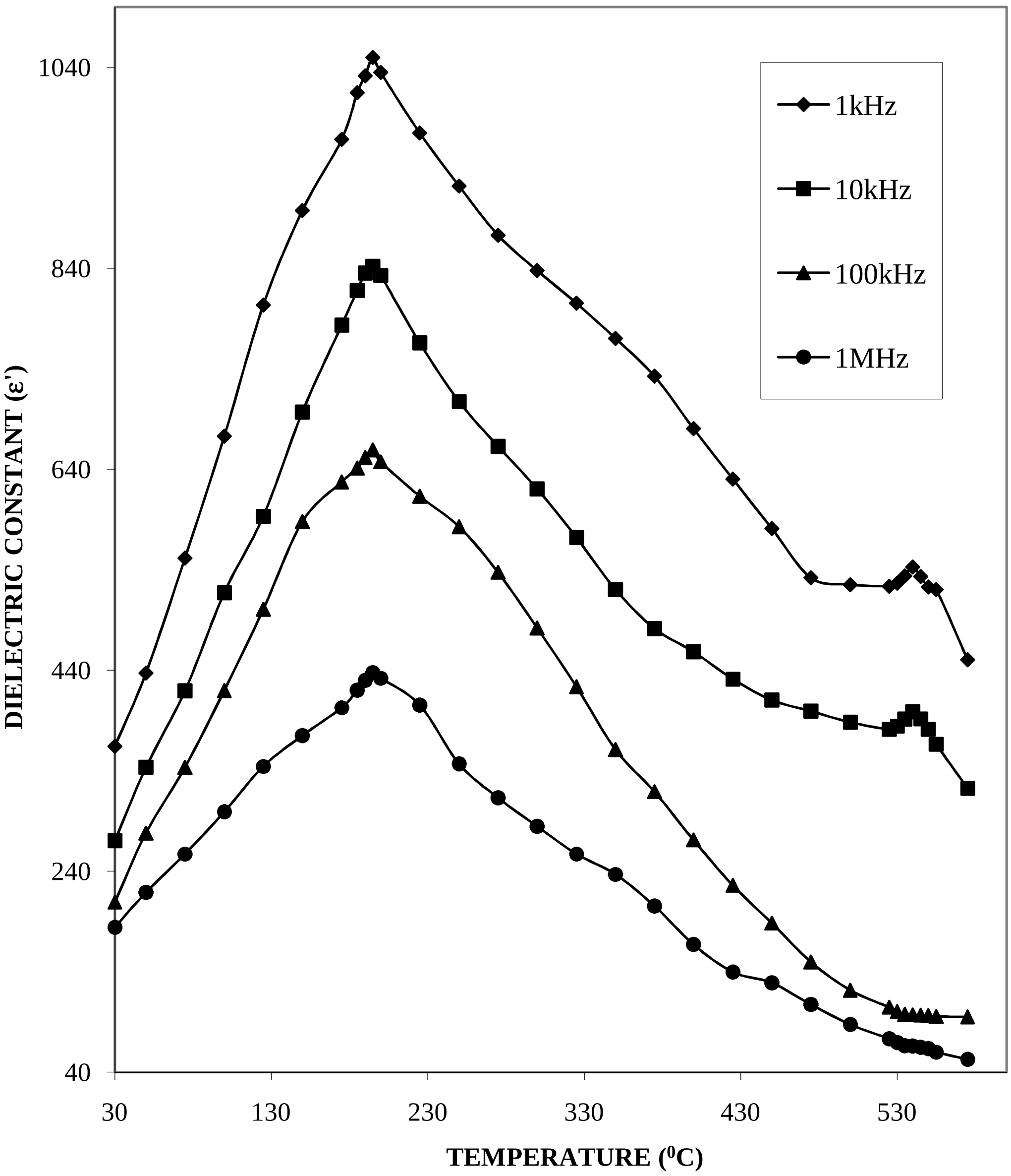

Fig.4 Variation of Dielectric Constant with Temperature for 70% $Ba_{0.7}Pb_{0.3}TiO_3$ + 30% $Ni_{0.4}Co_{0.6}Fe_2O_4$ Composites