

ELECTRICAL PROPERTIES OF BiFeO₃ - BaTiO₃ CERAMICS

R.K. Sharma,

Department of Physics, D.A.V. (P.G.) College, Dehradun

ABSTRACT

BiFeO₃- BaTiO₃ ceramics have been synthesized through solid state reaction method. In this paper, electrical properties for different composition $x= 0, 0.02, 0.04,$ and 0.06 wt% in BaTiO₃/xBFO ceramics were studied in the frequency range of 102 Hz to 105 Hz. The result shows perfect solubility of BiFeO₃ in BaTiO₃. The maximum dielectric constant was found to increase and transition temperature decreased with the increasing percentage of BFO in BaTiO₃ with diffuse phase transition.

Key words : Dielectric constant, Curie temperature, X-ray diffraction, SEM

INTRODUCTION

To obtain high quality ferroelectrics for a specific use it is necessary to understand various phenomena concerning structural and electrical properties of the materials. For this purpose it is desired to make available as many experimental data as possible by using various techniques. Hence the aim of the research described in this paper is focused on the preparation and characterization of $x\text{BiFeO}_3 - (1-x)\text{BaTiO}_3$, where $x=0, 0.02, 0.04$ and 0.06 . The effect of various-dopant-induced changes in structural, dielectric, mechanism of the dielectric peak broadening and frequency dispersion has been addressed. The basic idea is to enhance the dielectric and ferroelectric properties of these ceramics.

METHODOLOGY

The samples were characterized by powder X-ray diffraction. Stoichiometric amounts of the powders were mixed and milled in an agate mortar for about 20 minutes, with ethanol added as a milling aid. The powders were calcined for 10 hours in alumina crucibles at temperatures between 600 and 800°C.

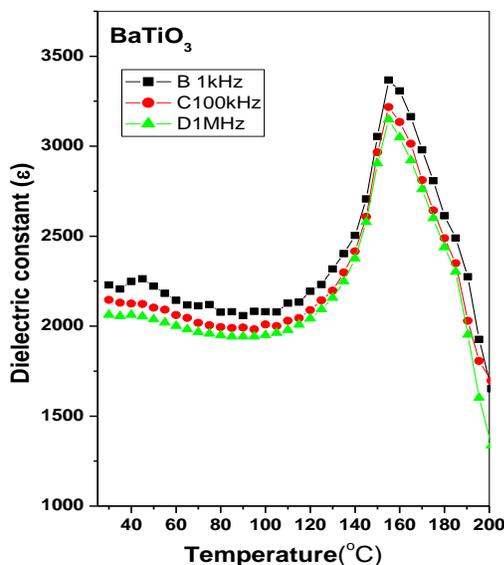
Subsequently, the powders were re-milled and pressed into pellets. The pellets were heated for 40 hours and then the cycle was repeated with milling, pelletizing and firing at raised temperatures. X-ray powder diffraction measurements at room temperature were performed with a Bruker advance D8 diffractometer, using CuK α radiation ($\lambda = 1.54056$ Å) in a range of Bragg's angle 2θ (20°-60°) at a scanning rate of 1°min⁻¹. In the present study **LEO-435VP** (operating voltage: 15 to 30 kV) scanning electron microscope has been used. The electrical properties were made using High Performance Frequency Analyzer at room temperature in the frequency range 1kHz-1MHz.

RESULTS AND DISCUSSION

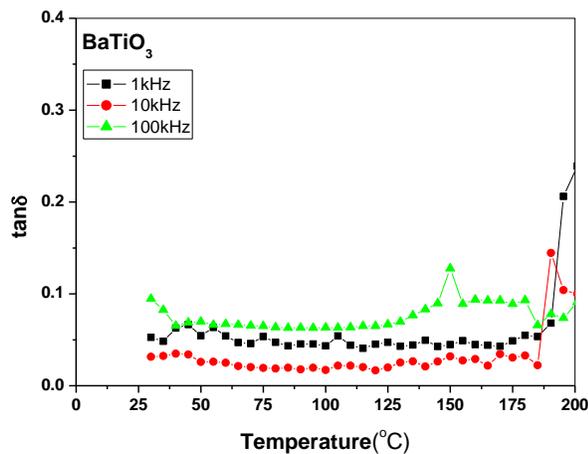
The variation of dielectric constant (ϵ) and $\tan\delta$ (dielectric loss) with frequency of all the samples at room temperature were measured in the frequency range of 10² Hz to 10⁵ Hz. From the nature of the variation of ϵ with frequency, it is possible to find out which contributions are predominately present in the compound in a particular frequency range.

Fig. 1 to 4 shows the variation of dielectric constant and $\tan\delta$ as a function of temperature at different frequencies for $x\text{BiFeO}_3 - (1-x)(\text{BaTiO}_3)$ samples. As for normal ferroelectrics, dielectric constant increases with increase of temperature up to transition temperature (T_c) then it decreases. It is seen that the region around the peak is broadened the broadening of the transition has been attributed

to disorder in the arrangement of the cations developed in A-site which is occupied here by Ba^{2+} and Bi^{3+} and at B- site by Ti^{4+} and Fe^{3+} . This leads to a microscopic heterogeneity in the composition and thus in a distribution of different local curie points. It is also seen that T_c is independent of increase in frequency.

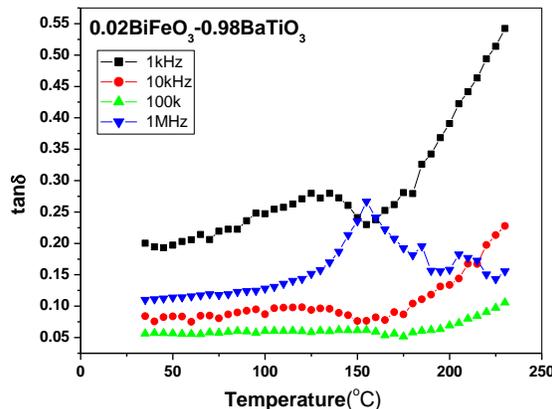
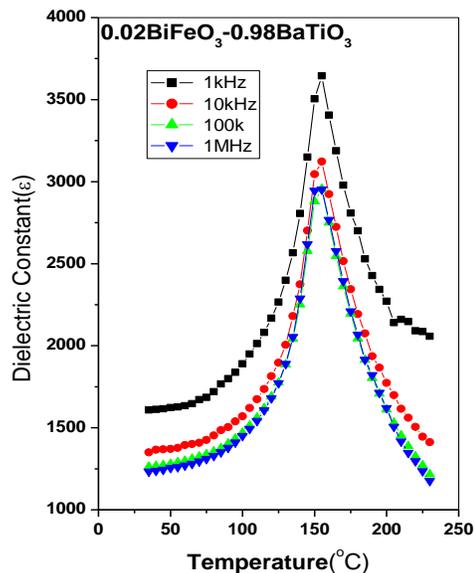


(a)



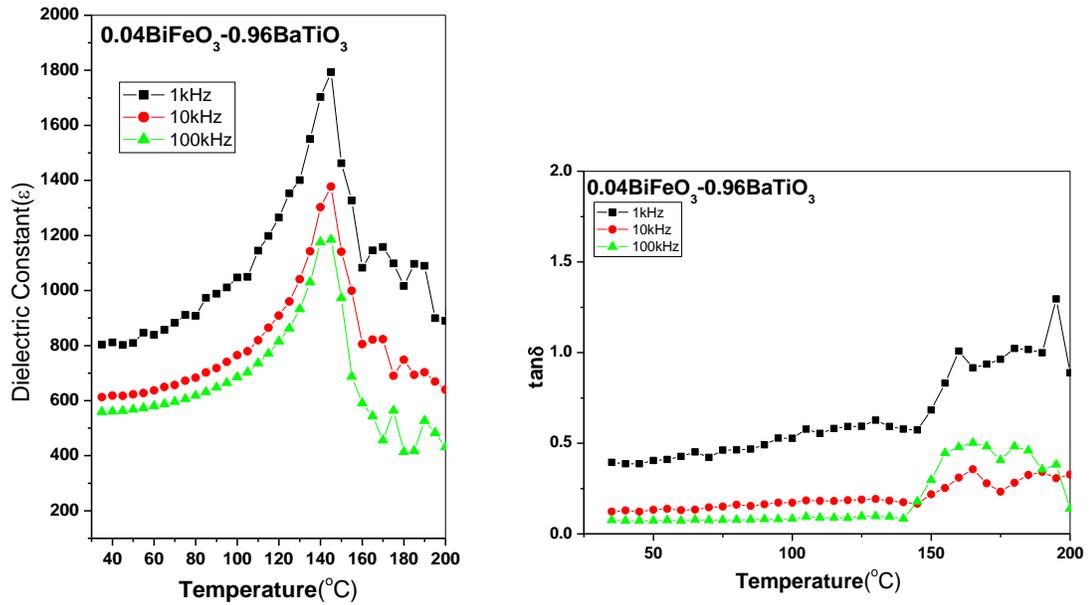
(b)

Figure 1 : Variation of (a) dielectric constant and (b) dielectric loss as a function of temperature of $x\text{BiFeO}_3 - (1-x)\text{BaTiO}_3$ for $x=0$.



(a) (b)

Figure 2 : Variation of (a) dielectric constant and (b) dielectric loss as a function of temperature of $x\text{BiFeO}_3-(1-x)\text{BaTiO}_3$ for $x=0.02$.



(a) (b)

Figure 3 : Variation of (a) dielectric constant and (b) dielectric loss as a function of temperature of $x\text{BiFeO}_3-(1-x)\text{BaTiO}_3$ for $x=0.04$.

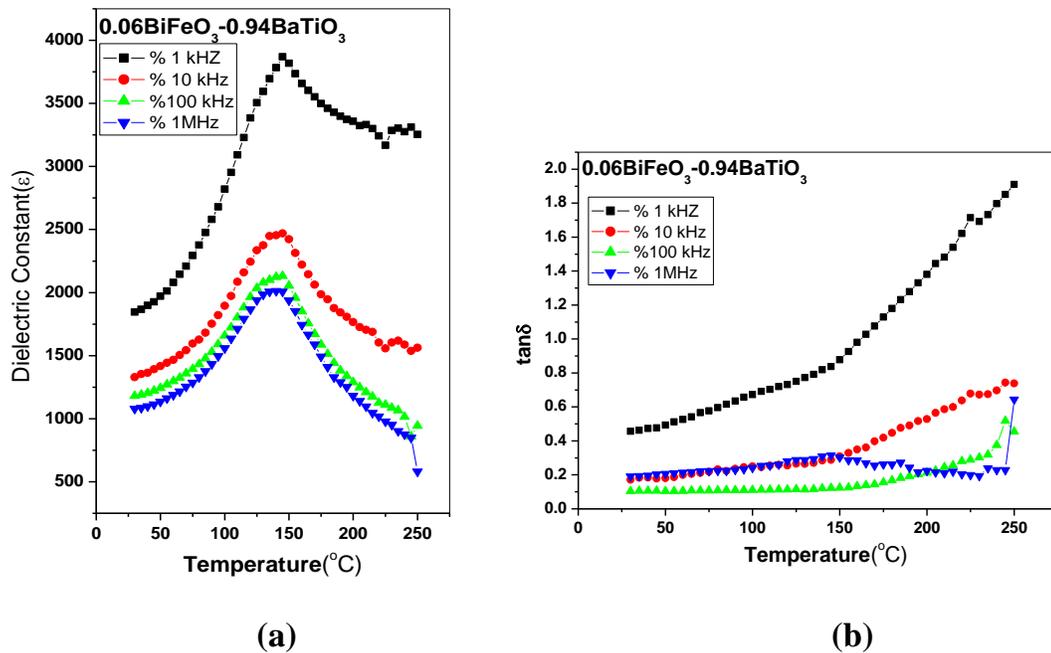


Figure 4 : Variation of (a) dielectric constant and (b) dielectric loss as a function of temperature of $x\text{BiFeO}_3 - (1-x)\text{BaTiO}_3$ for $x=0.06$.

The dielectric constant of all compositions was found to increase with increasing temperature, attaining a peak at a particular temperature called transition temperature (T_c), and there after it decreases. However, the magnitude of dielectric constant was found to be highly depending on both doping and measured frequency. The frequency dependence of dielectric constant shows strong dispersion at lower frequency range. This event is attributed to the low frequency space charge accumulation effect. Such strong dispersion in dielectric constant appears to be common feature in ferroelectrics associated with non-negligible ionic conductivity and is referred to as the low frequency dielectric dispersion (LFDD). The region around the dielectric peak is observed to be broadened, which is one of the most important characteristics of a disordered perovskite structure

with diffuse phase transition. This broadening is considered to be due to compositional fluctuations.

The dielectric loss was found to be very small and decreasing with frequency, also above 300 °C, a sharp increase in $\tan \delta$ was observed. This growth in $\tan \delta$ is brought by an increase in both conduction of residual current and the conduction of absorption current. In polar substances, apart from dipole losses, losses due to electrical conduction, which increases with increase in temperature, are present.

Room temperature magnetization versus magnetic field ($M-H$) curves (Fig. 5) of $(1-x)\text{BiFeO}_3 - x\text{BaTiO}_3$ ceramics was measured with magnetic field upto 70 kOe showing weak hysteresis loop.

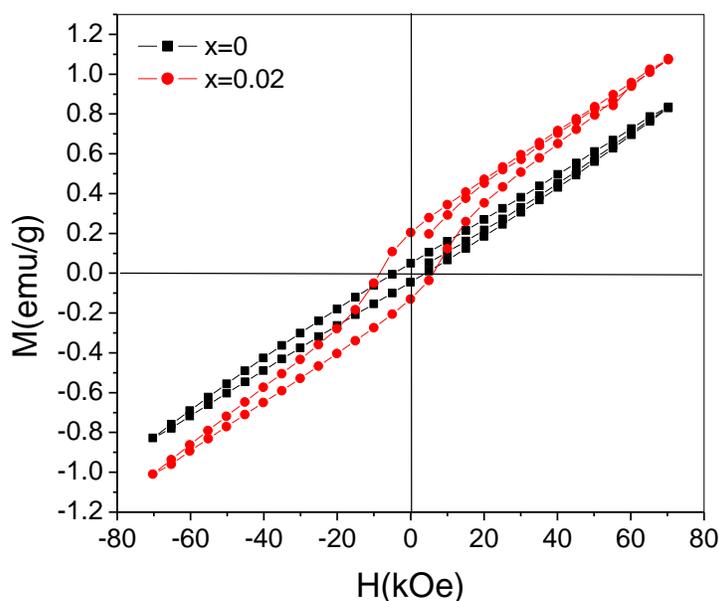


Figure 5 : Room temperature magnetic hysteresis plots for $(1-x)\text{BiFeO}_3 - x\text{BaTiO}_3$ samples.

The $M-H$ loops show a maximum magnetization value of 1.1 emu g^{-1} and no tendency to saturate up to the maximum applied field of 7 T. We obtain a

remnant magnetization of $2M_r = 0.0944 \text{ emu g}^{-1}$ and a coercivity of $2H_c = 8746 \text{ Oe}$ for BLPFO-1 sample. For $x=0.02$ samples, the $2M_r$ value increases to

0.3375emu g⁻¹ (3.57 times of BLPFO-1) and 2H_c increases to 14933Oe. Fig. 6 shows the room temperature VSM measurements of (1-x)BiFeO₃-xBaTiO₃, where x=0, x=0.02, x=0.04 and x=0.06. It is clear that a little enhancement in magnetization is seen for x=0.02 sample as compared to pure BiFeO₃. On the other hand the magnetization is increased to almost 4 times when Ba is co substituted at Bi site.

Hence Ba doping plays the dominant role towards the increase in magnetization. This increase in magnetization could be attributed to the substitution induced suppression of spiral spin structure. Also the magnetization behavior of our modified ceramic was found to be notably temperature dependent.

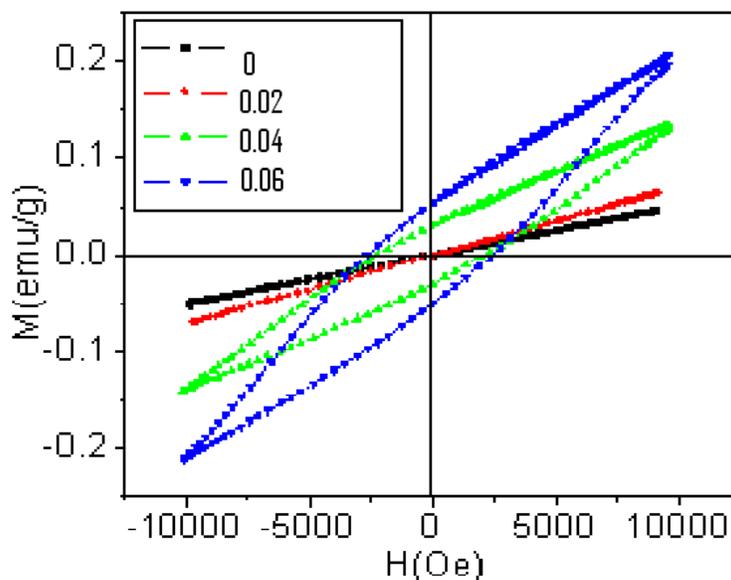
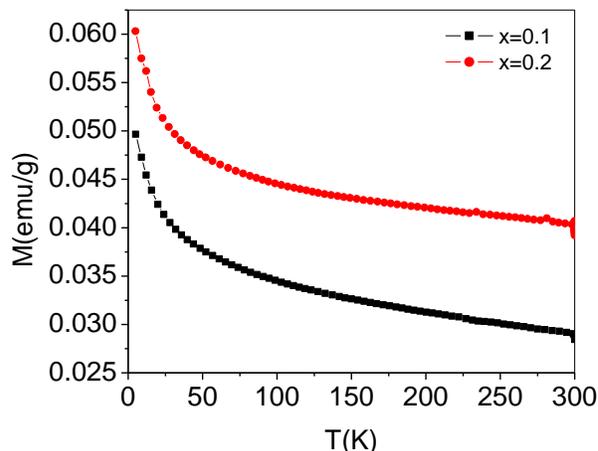
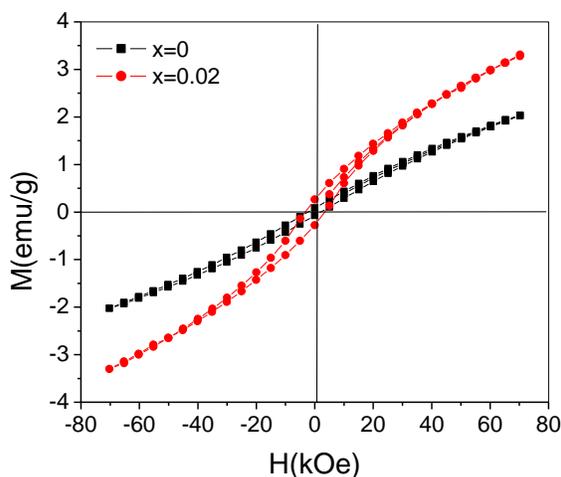


Figure 6 : Room temperature VSM measurements for (1-x)BiFeO₃-xBaTiO₃ samples.

Figure 7 (a) shows the M-H response for the (1-x)BiFeO₃-xBaTiO₃ ceramic taken at T=5K while 7(b) shows M-T response for (1-x)BiFeO₃-xBaTiO₃ ceramic and the inset shows the temperature

dependent magnetization for the moderate amount of applied field of 500Oe so that the external field might not overshadow the intrinsic behavior of spins.



(a)

(b)

Figure 7 : shows the (a) *M-H* and (b) *M-T* response for the $(1-x)\text{BiFeO}_3-x\text{BaTiO}_3$ samples.

The remnant magnetization was found to increase upto 1.63 times for both the samples on cooling to $T=5\text{ K}$; however, the coercive field was decreased to half of the room temperature H_c values. This behaviour is also observed in some high anisotropy magnetic materials. Similar decrease in coercivity at low temperature has also been reported earlier in Nd doped BiFeO_3 and $\text{BiFeO}_3-\text{BaTiO}_3$ solid solution. J F Scott et. al. have reported the magnetic anisotropy changes at low temperature in BiFeO_3 . Below room temperature it undergoes spin reorientation transitions at $T_2 = 200\text{K}$ and $T_1 = 140\text{K}$. Between T_1 and T_2 the magnetic symmetry is very low and hence the system shows linear magnetoelectric effect. These transitions are related to spin frustration. Hence this decrease in magnetic coercivity (H_c) could be attributed to magnetic anisotropy changes at low temperatures.

CONCLUSION

In the present work we have reported the $\text{BaTiO}_3/x\text{BFO}$ ($x= 0, 0.02, 0.04, \text{ and } 0.06\text{ wt\%}$) ceramic which was successfully synthesized by a solid-state mixed oxide method. The result also clearly shows that there are no secondary phases. It shows perfect solubility of BiFeO_3 in BaTiO_3 . The maximum dielectric constant was found to increase and transition temperature decreased with the increasing percentage of BFO in BaTiO_3 with diffuse phase transition. So a new $\text{BaTiO}_3/x\text{BFO}$ ($x= 0, 0.02, 0.04, \text{ and } 0.06\text{ wt\%}$) ceramic system provides useful information for material development.

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