Electric Modulus and Impedance Study of $0.83Ba(Fe_{0.5}Nb_{0.5})O_3$-$0.17BaTiO_3$ Ceramics

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ABSTRACT: In the present paper, we report the preparation characterization and dielectric properties of $0.83Ba(Fe_{0.5}Nb_{0.5})O_3$-$0.17BaTiO_3$ (BFN-BT) ceramics were synthesized by the tradition solid-state reaction method. The X-ray diffractograms and scanning electron micrographs of the samples were obtained taken at room temperature. The average grain size of compounds was around ~2.8µm. Sintered pellets were investigated for its dielectric (Electric Modulus and Impedance) properties in the temperature range of 30°C-400°C and in the frequency range of 100Hz-5MHz. Modulus analysis has indicated the possibility of hopping mechanism for electrical transport processes in the system with non-exponential-type conductivity relaxation.

I. INTRODUCTION

The electrical properties are often represented in terms of some complex parameters like complex impedance (Z^*), complex dielectric modulus (M^*), and loss tangent (tan). They are related to each other as follows: $Z^* =$

$$Z - j Z = R_s - \frac{j}{\omega C_s}, M^* = M + j M = j \quad _0 Z^* \text{ and}$$

loss tangent, $\tan = \dfrac{M^{''}}{M'} = -\dfrac{Z^{'}}{Z''}$ where $j = \sqrt{-1}$, the

imaginary factor, R_s and C_s are the resistance and capacitance in series (s),   (=2 f) is the angular frequency and $_0$ is the permittivity in free space (8.854 x 10^{-12} F/m). The above three expressions offer a wide scope for graphical representation. In recent years, high dielectric materials play an important role in microelectronics and have been utilized in the wide range of technological applications. For the development of capacitors, many investigators have tried to synthesize and study the materials that exhibit dielectric constant [1-4]. Being a lead free complex perovskite material, $BaTiO_3$ doped $Ba(Fe_{0.5}Nb_{0.5})O_3$ ie. $0.83Ba(Fe_{0.5}Nb_{0.5})O_3$-$0.17BaTiO_3$ (BFN-BT) is an environmental friendly material, thus making it a good substitute for Pb containing compounds for various applications. In recent years, many workers have studied BFN including Yokosuka [5], Tezuka *et al.* [6], Raevski *et al.* [7], Saha and Sinha [8, 9].

They have reported that the BFN-based Electroceramics exhibit a relaxor behavior by showing very attractive dielectric and electric properties over a wide range of temperatures.

Complex modulus formalism is a very important and convenient tool to determine, analyze and interpret the dynamical aspects of electrical transport phenomena, i.e., parameters such as carrier/ion hopping rate, conductivity relaxation time, etc. We have adopted the modulus formalism to study the relaxation mechanism in BFN-BT ceramics. Complex electric modulus formalism/analysis gives information on the nature of the polycrystalline samples, which can be probed into bulk and grain boundary effects. The usefulness of the modulus representation in the analysis of the relaxation properties has been demonstrated for polycrystalline ceramic. In the modulus formalism, an electric modulus $M^*(\omega)$ is defined in terms of complex dielectric permittivity $\varepsilon^*(\omega)$,

$$M^*(\omega) = 1 / \varepsilon^* = M'(\omega) + M''(\omega) \quad \dots(1)$$

II. EXPERIMENTAL DETAILS

Powders of $BaCO_3$ (reagent grade), TiO_2, Fe_2O_3 and Nb_2O_5 (reagent grade) were taken in stoichiometric ratio, and mixed in the presence of acetone for 5 h. The mixture was calcined at 1200ºC in air for 6 h and pellets of sample were sintered at 1250ºC for 8 h.

From the measurement, we have obtained capacitance (C) and tangent loss (tan) of the samples as a function of frequency (100 Hz to 5 MHz) at different temperature (30°C–400°C) using a PSM (Comm.1735). The X-ray powder diffraction pattern of the sample is taken at room temperature using a X-ray powder diffractometer (Rigaku Miniflex, Japan) using Cuk radiation (= 1.5418 Å) in a wide range of Bragg angles 2 (20° 2 80°) with scanning rate 2°/min. The micrographs are recorded using scanning electron microscopy JEOL-JSM-5800 to study the surface morphology/microstructure of the sintered pellets.

III. RESULTS & DISCUSSIONS

The X-ray diffraction (XRD) data indicate that single phase with monoclinic structure of BFN-BT ceramics is shown in Fig. 1. The lattice parameters of the selected unit cell were refined using the least-squares subroutine of a standard computer program package "POWD" [10].
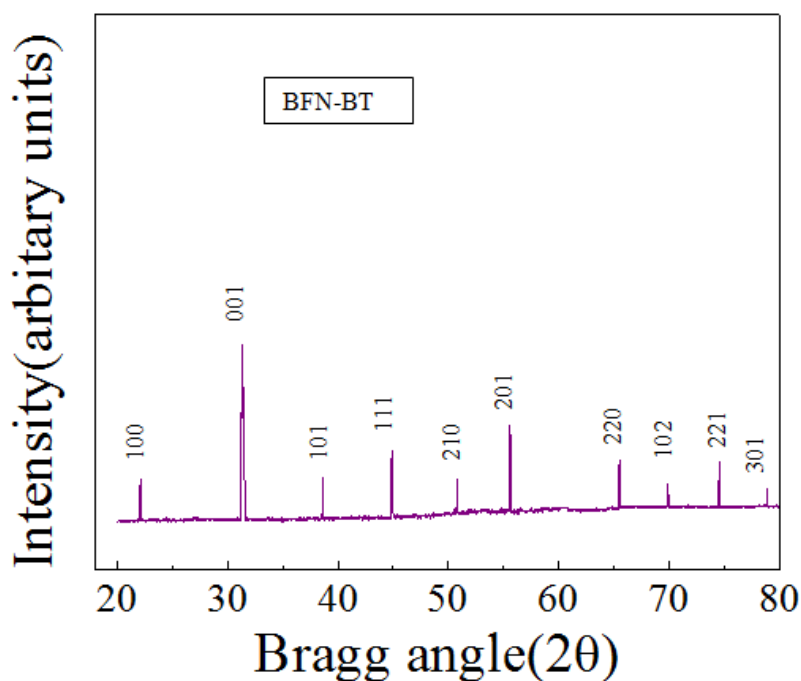

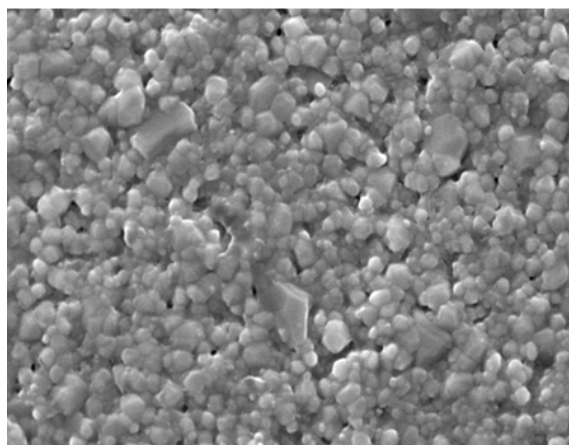**Fig. 1.** XRD of BFN-BT ceramics at room temperature.

Fig. 2. SEM micrographs of the BFN-BT ceramics, at room temperature.

In the Fig. 2 SEM micrographs of sintered pellets were showing a few pores at grain boundaries, but cracks or microcracks were not detected. The average grain size of compounds was around ~2.8µm. The logarithmic angular frequency dependence of imaginary Z and real Z' part of complex impedance (Z^*) for $0.83Ba(Fe_{0.5}Nb_{0.5})O_3-0.17BaTiO_3$ samples at several temperatures between 70°C and 230°C is plotted in Fig. 3. The value of Z' is larger in the low frequency range and gets a monotonous decrease with rise in frequency. It may be caused due to the effect of polarization in the samples. Interestingly ,the magnitude of Z' is found to decrease with the rise in temperature which suggests the typical negative temperature coefficient of resistance type behavior of above materials, usually observed in semiconductors. In the high frequency range, all the curves merge at a very low value so that Z' appears to become independent of both the frequency and temperature. It indicates a possible release of space charge [11–14].

At lower temperatures Z decreases monotonically suggesting that the relaxation is absent. This means that relaxation species are immobile defects and the orientation effects may be associated. As the temperature increases, the peak of Z" starts appearing. The peak shifts towards higher frequency with increasing temperature showing that the resistance of the bulk material is decreasing. Also the magnitude of Z" decreases with increasing frequencies. Corresponding figure indicates the spreading of the relaxation time.
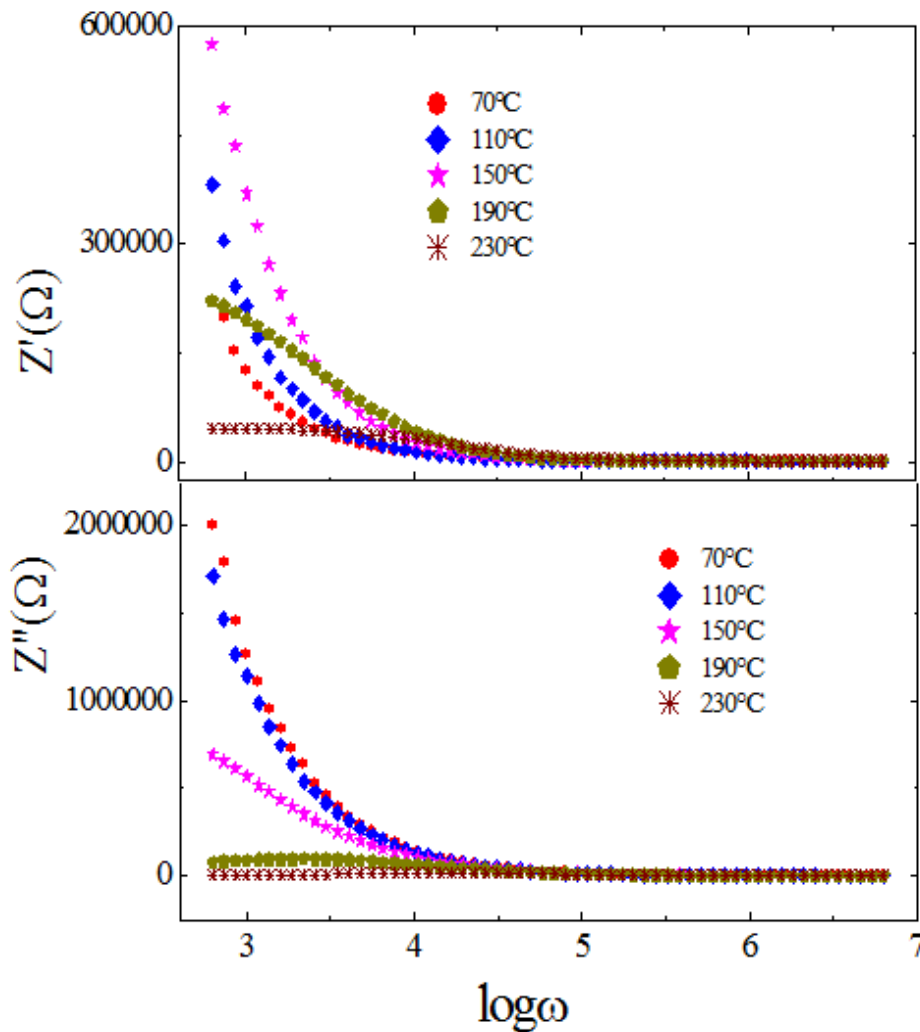

Fig. 3. Logarithmic angular frequency dependence of Z' and Z of BFN-BT ceramics at various temperatures.
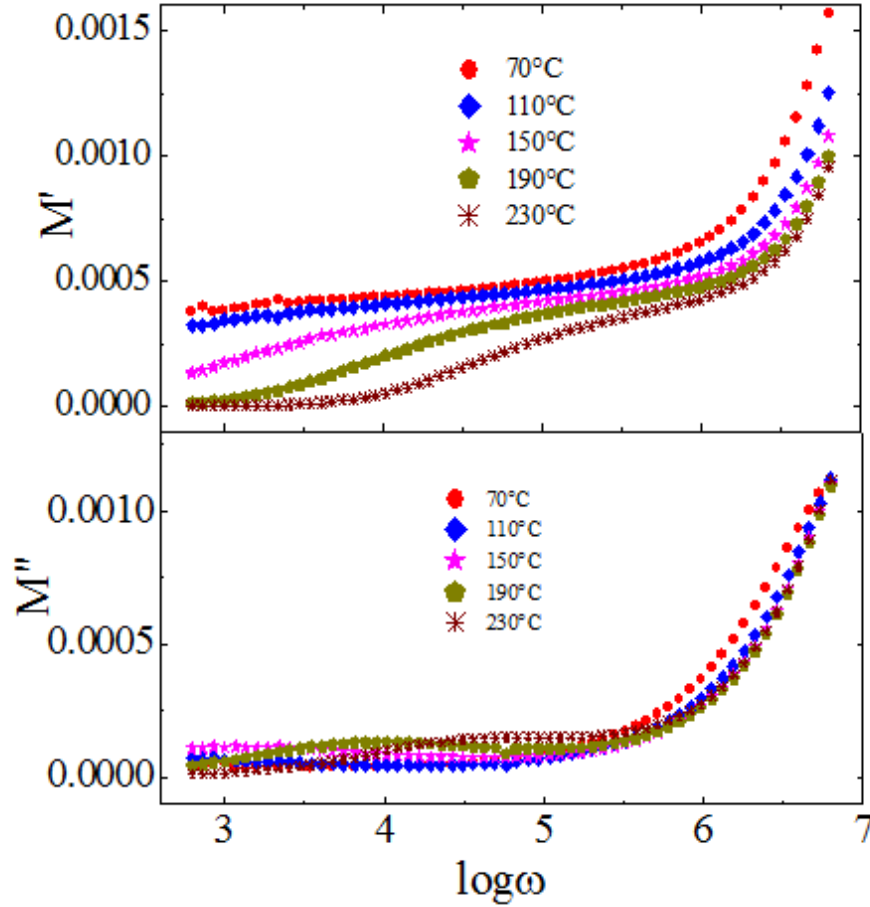
Fig. 4. Logarithmic angular frequency dependence of and M' and M of BFN-BT ceramics at various temperatures.

The logarithmic angular frequency dependence of the real (M′) and imaginary (M) part of complex electric modulus (M^*) over the temperature range from 70 to 230°C for BFN-BT ceramics is shown in Fig. 4. It shows a very low value (nearly zero) in the low frequency region and a higher value in the high frequency region arising due to continuous dispersion on increasing frequency. The value of M' increases with the increase in frequency and decreases with rise in temperature. It may be contributed to the conduction phenomena due to short range mobility of charge carriers. This implies the lack of a restoring force for flow of charge under the influence of a steady electric field [15]. The asymmetric modulus peaks shifts towards higher frequency side exhibiting the correlation between motions of mobile charge carriers [16]. The asymmetry in peak broadening shows the spread of relaxation times with different time constant, and hence the relaxation is of non-Debye type.

The existence of low frequency peaks suggests that the ions can move over long distances where as high frequency peaks suggest about the confinement of ions in their potential well. The nature of modulus spectrum confirms the existence of hopping mechanism in the electrical conduction of the materials.

IV. CONCLUSIONS

Complex perovskite oxides $0.83Ba(Fe_{0.5}Nb_{0.5})O_3$-$0.17BaTiO_3$ (BFN-BT) were synthesized by the tradition solid-state reaction method. The XRD patterns of the BFN-BT at room temperature show a monoclinic phase. The microstructure of the ceramics was examined by the scanning electron microscopy (SEM). The nature of modulus spectrums confirms the existence of hopping mechanism in the electrical conduction of the materials.

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