

# Comparative study of structure, dielectric and electrical behavior of Ba(Fe<sub>0.5</sub>Nb<sub>0.5</sub>)O<sub>3</sub> ceramics and their solid solutions with BaTiO<sub>3</sub>

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## ABSTRACT

Dielectric properties of (1-x)Ba(Fe<sub>0.5</sub>Nb<sub>0.5</sub>)O<sub>3</sub>-xBaTiO<sub>3</sub> (where x = 0.00, 0.05 and 0.10) solid solution ceramics at high temperature range of RT ~ 270 °C have been characterized in this paper. The above said polycrystalline ceramics with (x = 0.0, 0.05 and 0.10) have been produced via a mixed oxide route. The effects of BaTiO<sub>3</sub> substitution on the structure and on the electrical and ferroelectric properties of Ba(Fe<sub>0.5</sub>Nb<sub>0.5</sub>)O<sub>3</sub> samples have been studied by performing x-ray diffraction and dielectric measurements. The dielectric properties ( $\epsilon'$  and  $\tan\delta$ ) were investigated in the temperature range of 30-270 °C and in the frequency range of 100 Hz-5 MHz. The variation of relative dielectric permittivity ( $\epsilon'$ ) and tangent loss ( $\tan\delta$ ) has suggested a significant role of hopping of trapped charge carriers, which is resulted in an extra dielectric response in addition to the dipole response. It is observed that: (i) the relative dielectric permittivity( $\epsilon'$ ) and tangent loss ( $\tan\delta$ ) are dependent on frequency, (ii) the temperature of dielectric permittivity maximum shifts toward lower temperature side and (iii) dielectric permittivity and tangent loss rapidly increase by making solid solution of BFN with BaTiO<sub>3</sub>. X-ray diffraction analysis of the compound suggests the formation of single-phase compound with monoclinic structure. SEM photographs exhibit the uniform distribution of grains. The maximum ferroelectric transition temperature ( $T_c$ ) of this system was 250-270 °C with the dielectric constant peak of 72500 at 1.09 kHz for x = 0.05. Copyright © 2011 VBRI press.

**Keywords:** Ceramics; scanning electron microscopy; X-ray diffraction; dielectric properties.



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## Introduction

Recently, perovskite like ceramics have been at the center of research to study the ferroelectric and piezoelectric materials because of the diversity of their physical properties. These compounds have found applications in several industrial fields, such as high density capacitors, memory devices, actuators and so on [1]. Many ferroelectrics of the perovskite family of a general formula  $ABO_3$  ( $A =$  mono or divalent,  $B =$  triad to pentavalent ions) are very important because of their potential applications such as dynamical and volatile memory components, transducers, sensors and many other active and passive devices [2-4]. During the intervening years, a large amount a good understanding of the many inter-related properties and phenomena associated with these materials has been achieved. Most of dielectric materials with perovskite structures contain more or less lead, which pollutes the environment. So the, leadfree materials with high dielectric constants are becoming more attractive for research. Recently, high dielectric constant and dielectric relaxation in  $A(Fe_{0.5}B_{0.5}O_3)$ , ( $A =$  Ba, Sr and  $B =$  Nb, Ta) [5-11],  $CaCuTi_4O_{12}$  [12],  $CdCr_2S_4$  [13] etc have been pursued to understand the relaxation mechanism, which describes the dielectric relaxation (i.e., charge redistribution, structural frustration or polaron redistribution, ferroelectric relaxor, and Maxwell-Wagner space charge (pseudo relaxor)). The high value of dielectric constant over a very wide range of temperature interval is due to disorder in the distribution of B-site ions in the perovskite unit cell. Various relaxation processes seem to coexist in real perovskite crystals or ceramics, which contain number of different energy barriers due to point defects appearing during technological process. Therefore, the departure of the response from the ideal Debye model in the solid-state samples, resulting from the interaction between the dipoles, cannot be disregarded [14].  $BaTiO_3$ -based dielectrics are more focused in research field, due to its not detrimental to the environment both in synthesis and its application. The compositions of the majority dielectric materials used for ceramic capacitors are based on barium titanate [15-17].

The high permittivity materials are considered for temperature stable multilayer capacitors, to show abnormally flat dielectric temperature curves [18]. Number of perovskite solid solutions, such as  $Ba_{0.6}Sr_{0.4}TiO_3$  [19], lead lanthanum zirconate titanate (PLZT) [20], and  $Ba(Ti_{1-y}Zr_y)O_3$  [ $0 \leq y \leq 0.16$ ] [21], illustrated the phase transition behavior in certain ferroelectric compositions [17]. Furukawa et al. [22] developed multilayer relaxor/  $BaTiO_3$  capacitor composites with alternating layers of low and high-Curie temperature dielectrics to meet X7R-type capacitor specifications.  $Ba(FeNb)_{1/2}O_3$  is a relaxor-type ferroelectric material which has a partially disordered perovskite structure [1,23-25]. The  $Fe^{+3}$  and  $Nb^{+5}$  ions are randomly distributed in the octahedral positions and have small lattice distortion.

In view of the above, we have made an attempt to study  $Ba(Fe_{0.5}Nb_{0.5})O_3$  (BFN) and its solid solution with  $BaTiO_3$ . In this paper we will discuss the structural, dielectric and electrical behavior of the synthesized ceramics.

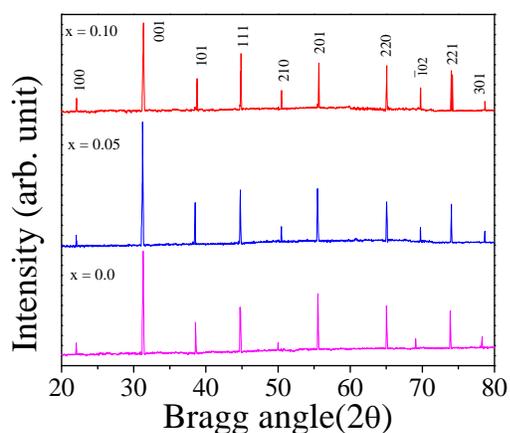
## Experimental

The compounds used in this investigation were prepared by a routine solid-state reaction technique. The starting materials were  $BaCO_3$  (M/S Burgoyne Buirbridger, India),  $Fe_2O_3$  (M/S Indian rare earth Ltd.),  $Nb_2O_5$  (M/s. Loba Chemie Pvt. Ltd., India) and  $TiO_2$  (M/S John Baker Inc., USA). The raw powders were based on the formula  $(1-x)Ba(Fe_{0.5}Nb_{0.5})O_3-xBaTiO_3$ . The doping concentration of was varied from 0.05 to 0.10 mole%. These materials were taken in a suitable stoichiometry ratio and mixed thoroughly with acetone in an agate-mortar for 4 h. The mixtures of ingredients of compounds were calcined in an alumina crucible at  $1200^\circ C$  for  $\sim 8$  hr in air atmosphere. The calcined powders were mixed with the binder (polyvinyl alcohol (PVA)) and compacted into cylindrical discs (pellets) of diameter  $\sim 10$  mm and thickness 1-2 mm at the pressure of  $5 \times 10^7$  N/m<sup>2</sup> using a hydraulic press. The pellets were sintered in air at  $1250^\circ C$  for 5 hr in high purity alumina crucibles. The formation and quality of the compounds were checked with X-ray diffraction (XRD) technique. The XRD patterns of the compounds were recorded at room temperature using x-ray powder diffractometer (Rigaku Miniflex, Japan) with  $CuK\alpha$  radiation ( $\lambda = 1.5405 \text{ \AA}$ ) in a wide range of Bragg angles  $2\theta$  ( $20^\circ \leq 2\theta \leq 60^\circ$ ) at a scanning rate of  $2^\circ \text{ min}^{-1}$ . SEM micrographs were obtained with a JEOL-JSM-5800 scanning electron microscope operated at 20 KeV. SEM images were taken from freshly broken surface of the specimens. The flat polished surfaces of the sintered samples were electroded with air-drying silver paste. Before taking electrical measurements, the pellets were kept at  $150^\circ C$  for two hours. The dielectric constants ( $\epsilon'$ ) and loss tangents ( $\tan\delta$ ) of samples were measured using a PSM (Comm.1735) while heating at the rate of  $0.5^\circ C \text{ min}^{-1}$  with a laboratory-made three-terminal sample holder.

## Results and discussion

### Structural analysis

Room temperature XRD patterns of the  $(1-x)Ba(Fe_{0.5}Nb_{0.5})O_3-xBaTiO_3$  (where  $x = 0.00, 0.05$  and  $0.10$ ) perovskite ceramics are shown in Fig. 1. All the reflection peaks were indexed using observed inter-planar spacing  $d$ , and lattice parameters of all the samples were determined by using least-squares refinement method. The calculated and observed  $d$  values of all diffraction lines (reflections) of above compounds suggest that there is a no change in the basic crystal structure from monoclinic as  $x$  changes from 0.00 to 0.10. A good agreement between the observed (obs) and calculated (cal)  $d$ -values (Table 1), suggests the correctness of the selection of crystal system and cell parameters. Our results are in good agreement with those by Saha and Sinha [7] and Shanming Ke et al. [8] deduced BFN having a monoclinic structure by using a standard computer program (POWD) analysis. The lattice parameters of different  $(1-x)Ba(Fe_{0.5}Nb_{0.5})O_3-xBaTiO_3$  compounds are given in Table 1.



**Fig. 1.** Room temperature XRD patterns of different  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples with different concentration ( $x = 0, 0.05, 0.10$ ).

### Microstructural analysis

Microstructure development during sintering was investigated by scanning electron microscopy (SEM). The surface micrographs of all compounds at magnification of  $2\mu\text{m}$  at room temperature are shown in **Fig. 2**. The SEM micrographs of as-sintered ceramics show a few pores at grain boundaries, but cracks or micro cracks were not detected. The average grain size of compounds was around  $2 - 5\mu\text{m}$ . Grains are distributed randomly in the samples surface. As in the **Fig. 2** for the solid solution  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$ , ( $x = 0.00, 0.05, 0.10$ ), the grain sizes are reduced to favor the matter transport mechanism during the sintering process. When the powders are submitted to the sintering process performed at  $1250^\circ\text{C}$  for 5 h, the movement of atoms or molecules is driven by differences in curvature between the particles in contact. In order to reduce surface free energy, atoms supposedly move from particles with smaller radius to those with larger radius. Particularly, for the  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  ceramics with concentrations of  $x = 0.00, 0.05$  and  $0.10$ , it is possible that the matter transport between several aggregated particles and the high anisotropy in the grain boundary energies induced the formation of compact and irregular particles.

### Dielectric analysis

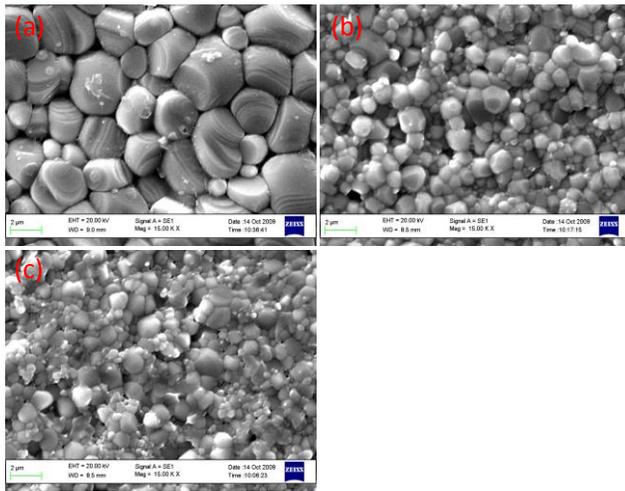
The logarithmic angular frequency dependence of the dielectric constant ( $\epsilon'$ ) and loss tangent ( $\tan\delta$ ) of all compounds at  $30^\circ\text{C}$  and  $130^\circ\text{C}$  are plotted in **Fig. 3(a-b)**. The dielectric constant ( $\epsilon'$ ) is decreases with increase of frequency at  $30^\circ\text{C}$ . But at  $130^\circ\text{C}$ , the  $\epsilon'$  value in low frequency region (below 1 kHz) increases significantly. This behavior is consistent with that of common ferroelectrics. Higher values of  $\epsilon'$  at lower frequencies are due to the presence of all different types of polarizations (i.e., dipolar, atomic, ionic, electronic contribution) in the material. At high frequencies, however, some of the above-mentioned polarizations may have less contribution in  $\epsilon'$ . The high value of  $\epsilon'$  in the low frequency region has been explained using Maxwell–Wagner (MW) polarization

effect. Thus high values of permittivity are not usually intrinsic, but rather associated with heterogeneous conduction in the grain and grain boundary of the compounds. The grains of the sample are separated by more insulating intergrain barriers, as shown in a boundary layer capacitor [23]. It is observed that  $\tan\delta$  maximum increases as temperature increases from  $30^\circ\text{C}$  to  $130^\circ\text{C}$  for  $x = 0.00, 0.05$  and  $0.10$ , indicating that the concentration of conduction electrons increases as temperature due to thermal activation [24].

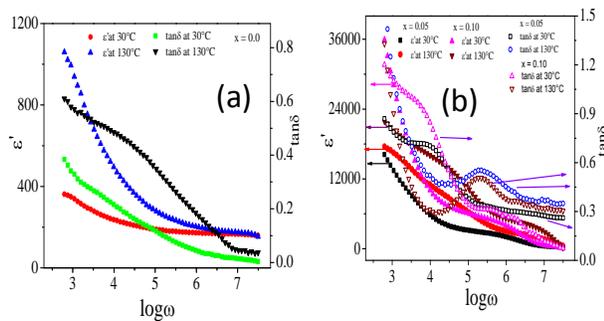
**Table 1.** Comparison of observed (obs) and calculated (cal) d-values (in nm) of reflections of  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples with different concentration ( $x = 0, 0.05, 0.10$ ) compounds at room temperature. The estimated error in d is  $\pm 0.00010$  nm.

For $x = 0.0$				
h	k	l	$d_{\text{obs}}$	$d_{\text{cal}}$
1	0	1	3.0328	3.0328
0	2	0	2.8564	2.8564
1	1	0	2.3338	2.3338
$\bar{1}$	2	1	2.0221	2.0221
0	3	1	1.6537	1.6534
For $x = 0.05$				
h	k	l	$d_{\text{obs}}$	$d_{\text{cal}}$
1	0	0	3.0233	3.0233
1	1	0	2.8609	2.8609
0	0	2	2.3386	2.3386
1	1	1	2.0247	2.0247
0	5	1	1.6548	1.6548
For $x = 0.10$				
h	k	l	$d_{\text{obs}}$	$d_{\text{cal}}$
1	1	0	3.0561	3.0561
0	0	1	2.8518	2.8518
$\bar{1}$	0	1	2.3334	2.3334
1	2	0	2.0230	2.0230
$\bar{1}$	2	1	1.6513	1.6513

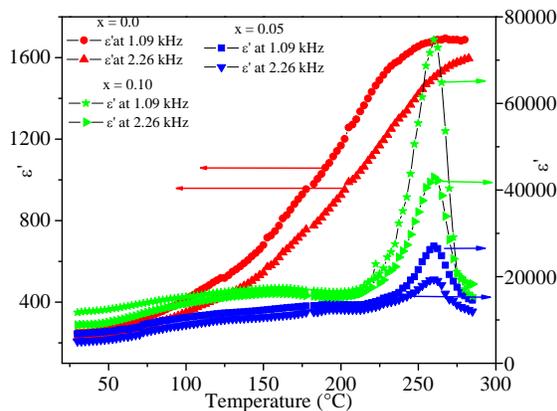
The variation of dielectric constant ( $\epsilon'$ ) as a function of temperature ( $30^\circ\text{C}$ – $270^\circ\text{C}$ ) at 1.09 kHz and 2.26 kHz frequencies of  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  ceramics is shown in **Fig. 4**. For  $x = 0.00$  the dielectric constant ( $\epsilon'$ ) is found to increase with rise in temperature for a given frequency. There are two possible explanations for this behavior. One possibility is that it is due to the increased conductivity in the samples caused by the presence of  $\text{Fe}^{2+}$  in sintered BFN ceramics, as suggested by Ananta and Thomas [25]. The concentration of  $\text{Fe}^{2+}$  ions is known to be very sensitive to temperature, and it increases as temperature increases [26]. It is known that the co existence of  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions on equivalent crystallographic sites



**Fig. 2.** SEM micrographs of cracked surfaces for different (1-x)Ba(Fe<sub>0.5</sub>Nb<sub>0.5</sub>)O<sub>3</sub>-xBaTiO<sub>3</sub> samples for (a) x = 0.0, (b) x = 0.05 and (c) x = 0.10 compositions.

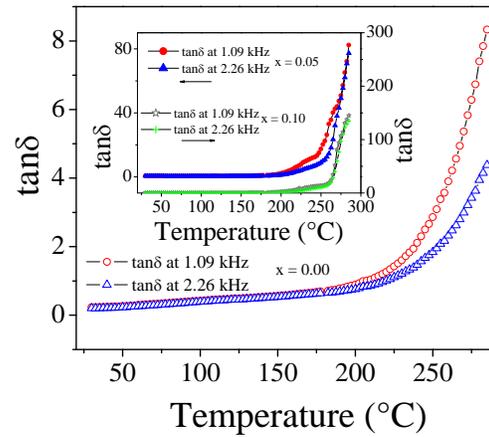


**Fig. 3.** (a) Variation of dielectric constant ( $\epsilon'$ ) and loss tangent ( $\tan \delta$ ) of compositions  $x = 0.0$  and (b) Variation of dielectric constant ( $\epsilon'$ ) and loss tangent ( $\tan \delta$ ) of compositions  $x = 0.05$  and  $0.10$  as a function of logarithmic angular frequency at various temperatures.



**Fig. 4.** Plots of dielectric constant vs. temperature with doping concentration ( $x = 0.0, 0.05$  and  $0.10$ ) at  $1.09$  kHz and  $2.26$  kHz.

can give rise to an electron hopping conduction mechanism. Due to the finite hopping (or jump) probability of electrons, this conduction mechanism tends to come into effect only at lower frequencies. An alternative explanation is related to disorder in the B-site cations of complex perovskites, as suggested by Majumdar et al. [27].

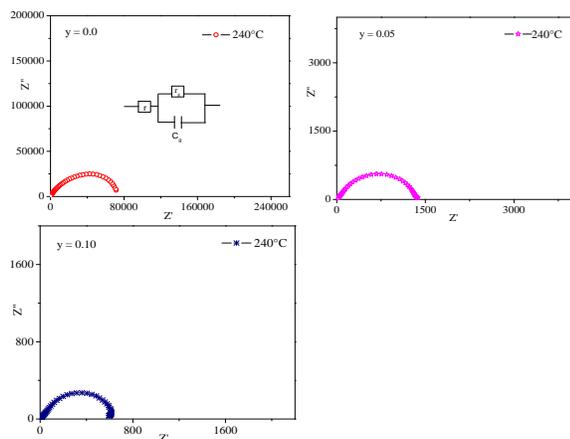


**Fig. 5.** Plots of loss tangent ( $\tan \delta$ ) vs. temperature with doping concentration ( $x = 0.0, 0.05$  and  $0.10$ ) at  $1.09$  kHz and  $2.26$  kHz.

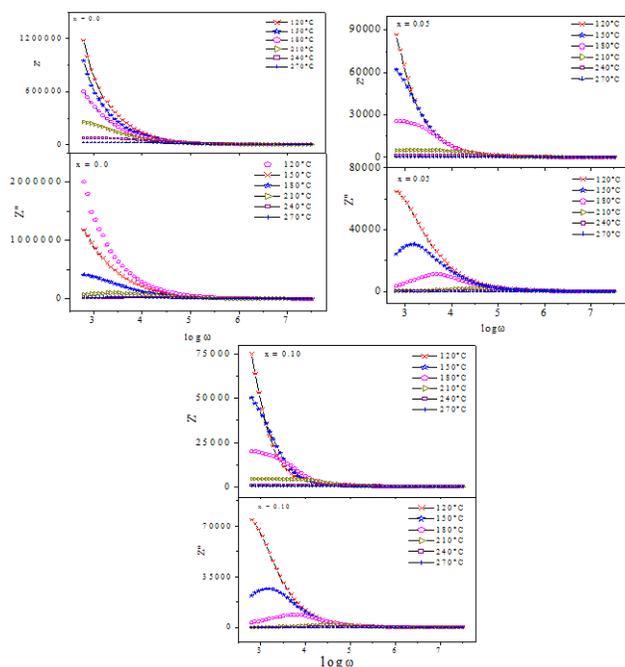
The dielectric constant ( $\epsilon'$ ) of  $x = 0.05$  and  $0.10$  ceramic showed a ferroelectric to paraelectric transition around  $260^\circ\text{C}$ . The peak position at the transition shows its dependence on the measured frequency. From **Fig. 5**, it is observed that  $\tan \delta$  remains almost constant up to  $215^\circ\text{C}$ , afterwards, it increases rapidly [28]. **Fig. 6** shows a complex-plane impedance plot ( $Z^*$ -plot) for  $x = 0.00, 0.05$  and  $0.10$  ceramics, between the imaginary part  $Z''$  against the real part  $Z'$  of complex impedance  $Z^* = Z' - jZ''$  at temperature  $240^\circ\text{C}$ . Generally, this type of plots can be characterized by three zones: (i) low-frequency zone where electrode polarization dominates, (ii) intermediate-frequency zone where polarization by migration of charges plays an important role, and (iii) high-frequency zone where dipole relaxation takes place. In all ceramics, a single semicircular arc is observed in the complex plane, and this can be explained by a RC equivalent circuit (**Fig. 6** (a), inset)). Dipole relaxation process occurs in all the materials since semicircular arc observed in the high-frequency zone. The resistance was obtained from the intersection of the semicircle and the  $Z'$ -axis. It is clear from the figure that the resistance decreases on increasing temperature. The semicircle has their center located away from the real axis, indicating the presence of relaxation species, and non-Debye type of relaxation process occurs in the materials.

The logarithmic angular frequency dependence of  $Z'$  and  $Z''$  of  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples, at several temperatures between  $120^\circ\text{C}$  and  $270^\circ\text{C}$  is plotted in **Fig. 7**. At lower temperature,  $Z'$  decreases monotonically with increasing frequency up to certain frequency and then becomes frequency independent in all samples. At higher temperatures,  $Z'$  is almost constant and for even higher frequencies decreases sharply. This also means that the resistive grain boundaries become conductive at these temperatures. This also shows that the grain boundaries are not relaxing even at very high frequencies even at higher temperatures. 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  $Z''$  for  $x = 0.05$  and  $0.10$  starts appearing. The peak shifts towards higher frequency with

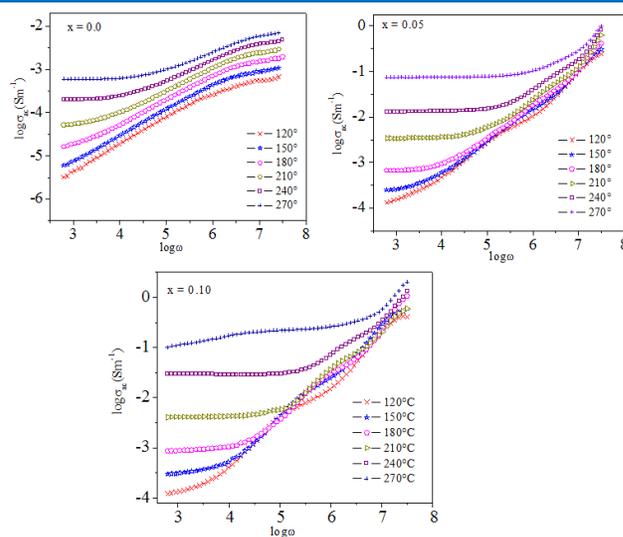
increasing temperature showing that the resistance of the bulk material is decreasing. Also, the magnitude of  $Z''$  decreases with increasing frequencies. Corresponding figure indicates the spreading of the relaxation time. This would imply that relaxation is temperature dependent, and there is apparently not a single relaxation time. There by relaxation processes involved with their own discrete relaxation time depending on the temperature. It is evident that with increasing temperature, there is a broadening of the peaks and at higher temperatures, the curves appear almost flat.



**Fig. 6.** Complex plane impedance plot of  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples with different concentration ( $x = 0, 0.05, 0.10$ ) at  $240^\circ\text{C}$  and equivalent circuit (inset).



**Fig. 7.** Plots of logarithmic angular frequency dependence of  $Z'$  and  $Z''$  of  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples of different concentration ( $x = 0, 0.05, 0.10$ ) at various temperatures.



**Fig. 8.** Logarithmic angular frequency dependence of ac conductivity ( $\sigma_{ac}$ ) for  $(1-x)\text{Ba}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3-x\text{BaTiO}_3$  samples of different concentration ( $x = 0, 0.05, 0.10$ ) at various temperatures.

The origin of frequency dependence of conductivity in the relaxation phenomenon arises due to mobile charge carriers. The ac conductivity provides the insight of the mobility of charge carriers. **Fig. 8** shows the ac conductivity of the all the compounds as a function of frequency at different temperature. The ac conductivity of the system depends on the dielectric properties and sample capacitance of the material. This behavior may be attributed to the presence of space charge in the material. The two plateaus separated by frequency region are observed in BFN( $x=0.00$ ) ceramic. The low-frequency plateau represents the total conductivity whereas the high-frequency plateau represents the contribution of grains to the total conductivity. The presence of both the high and low frequency plateaus in conductivity spectra suggests that the two processes are contributing to the bulk conduction behavior. One of these processes relaxes in the higher frequency region and contribution of the other process appears as a plateau in the higher frequency region. The investigated material followed universal Jonscher's power law [29] (i.e.,

$$\sigma(\omega) = \sigma(0) + A\omega^n, \quad (3)$$

where  $\sigma(\omega)$  is the total conductivity,  $\sigma(0)$  is the frequency independent conductivity (i.e.,  $\sigma_{dc}$  and  $A$  depends on temperature). The exponent  $n$  in the lower frequency region increases with the increase of temperature and is found to be in the range of 1.5-2. The conductivity shows dispersion which shifts to higher frequency side with the increase of temperature. The strong frequency dependent conductivity has been found above 10 kHz for  $x = 0.05$  and 0.10 with the frequency independent conductivity below 10 kHz which is a typical feature of perovskite at elevated temperature ( $>180^\circ\text{C}$ ).

## Conclusion

BFN-BT solid solution ceramics were prepared using solid state reaction method. BFN-BT ceramics have monoclinic

crystal structure at room temperature exhibiting better homogeneity and formation of compounds. BaTiO<sub>3</sub> doping in BFN exhibits many interesting feature. Studies of the dielectric constant ( $\epsilon'$ ) and tangent loss ( $\tan \delta$ ) of compounds suggest that the higher value of  $\epsilon'$  at the low frequency region has been explained using Maxwell–Wagner polarization effect. This is associated with heterogeneous conduction in the grains and grain boundaries of the compounds. The impedance plots showed only bulk contributions, and non-Debye type of relaxation process occurs in the material. The abnormal behavior of dielectric constant ( $\epsilon'$ ) of  $x = 0.05$  and  $0.10$  shows ferroelectric to paraelectric transition around  $260^\circ\text{C}$ .

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