Investigation of complex impedance and modulus properties of Nd doped 0.5BiFeO$_3$-0.5PbTiO$_3$ multiferroic Composites

Ajay Kumar Behera, Nilaya K Mohanty, Santosh K Satpathy, Banarji Behera*, Pratibindhya Nayak

Materials Research Laboratory, School of Physics, Sambalpur University, Jyoti Vihar, Burla - 768019, Odisha, India

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Abstract: 0.5BiNd$_{1-x}$Fe$_{1-x}$O$_3$ – 0.5PbTiO$_3$ (BN$_{1-x}$-PT) ($x = 0.05, 0.10, 0.15, 0.20$) composites were successfully synthesized by a solid state reaction technique. At room temperature, X-ray diffraction shows tetragonal structure for all concentrations of Nd doped 0.5BiFeO$_3$ – 0.5PbTiO$_3$ composites. The nature of Nyquist plot confirms the presence of bulk effects only for BN$_{1-x}$-PT ($x = 0.05, 0.10, 0.15, 0.20$) composites. The bulk resistance is found to decreases with the increasing temperature as well as Nd concentration and exhibits a typical negative temperature coefficient of resistance (NTCR) behavior. Both the complex impedance and modulus studies have suggested the presence of non-Debye type of relaxation in the composites. Conductivity spectra reveal the presence of hopping mechanism in the electrical transport process of the composites. The activation energy calculated from impedance plot of the composite decreases with increasing Nd concentration and found to be 0.89, 0.76, 0.71 and 0.70 eV for $x=0.05, 0.10, 0.15$ and 0.20 respectively.

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1. Introduction

Recently, there has been an extensive study in the direction of newer materials possessing ferromagnetic as well as the ferroelectric properties because of the richness of physics involved in the system as well as their potential applications in memory and functional devices [1–6]. These materials can be considered as multiferroic with the interaction of electric and magnetoelectric effects, and the effect of mutual influence of the polarization and magnetization [3, 4, 7]. BiFeO$_3$(BFO) is one of the natural ferroelectromagnets which exhibit both ferroelectric and ferromagnetic ($T_c = 1103$ K and $T_I = 1103$ K) and shows a rhombohedrally distorted perovskite crystal structure with space group of R$3c$ [8–10], which makes it a promising material for application at room temperature. However, its low remanent magnetization and relatively leakage current are the main limitation for possible device application. Hence, the special attention has been laid on synthesis of BFO with some rare earth or other...
group element as reviewed by Silva et al. [11] for improved multiferroic properties. However, there are reports of coexistence of ferroelectricity and magnetism in BFO which is induced by the rare earth substitution in the A and B site of the perovskite structure. Rare earth ions such as La$^{3+}$, Sm$^{3+}$ and Nd$^{3+}$ have been added at the A-site and Nb$^{5+}$, Mn$^{4+}$, Ti$^{4+}$ or Cr$^{3+}$ at the B-site for enhancing the electrical and magnetic properties of BFO [12–17]. The series of (BiFeO$_3$)$_{1-x}$-(PbTiO$_3$)$_x$ solid solution was first reported on by Venevstev et al. in 1960 [18]. They observed that the tetragonal phase persists in the series from the PbTiO$_3$ end member up to 70 wt% BFO and remarked upon the large c/a ratio present in the tetragonal structure at 60 wt% BFO. Moreover, Wang et al. [19] reported an enhancement of ME properties in the (1–x)BiFeO$_3$–xPbTiO$_3$ solid solutions. Zhu et al. [20] proposed a phase diagram for the (1–x)BFO–xPT. Their results reveal the existence of a morphotropic phase boundary (MPB) in this system, at which rhombohedral ($x \leq 0.20$), orthorhombic ($0.20 < x < 0.28$) and tetragonal ($x \geq 0.31$) phases exist with a large tetragonality in the tetragonal phase region. Further the impedance, modulus, relaxation time, ac and dc conductivity properties have not been investigated so far at elevated temperatures. In this work, we report the complex impedance and modulus properties of 0.5BiNd$_x$Fe$_{1-x}$O$_3$–0.5PbTiO$_3$ ($x=0.05, 0.10, 0.15, 0.20$) multiferroic composites.

2. Experimental

Neodymium (Nd) doped solid solutions of BFO–PT with the formula 0.5BiNd$_x$Fe$_{1-x}$O$_3$–0.5PbTiO$_3$ ($x=0.05, 0.10, 0.15, 0.20$) were prepared by the solid state reaction technique. The high purity precursors: Bi$_2$O$_3$ and Nd$_2$O$_3$ (99.99%, Spectrochem Pvt. Ltd., India), Fe$_2$O$_3$ (99%, M/s Loba Chemicals, Pvt. Ltd., India), PbO (95%, Qualigen Pvt. Ltd., India) and TiO$_2$ (99% Merck Specialities Pvt. Ltd., India) were carefully weighed in a suitable stoichiometric proportion. The polycrystalline materials were mixed thoroughly and ground in an agate mortar for 2 h and in methanol for 2 h. The mixed powders were calcined in a high purity alumina crucible at an optimized temperature of 800°C for 6 h in an air atmosphere. The process of grinding and calcinations was repeated several times till the formation of the compound was confirmed. Then calcined powder was mixed with polyvinyl alcohol (PVA) which acts as a binder to reduce the brittleness of the pellet and burnt out during high temperature sintering. The fine homogenous powder was cold pressed into cylindrical pellets of 10 mm diameter and 1–2 mm of thickness at pressure of $4 \times 10^6$ N/m$^2$ using a hydraulic press. These pellets were sintered at 800°C for 6 h in an air atmosphere. The sintered pellets were polished by fine emery paper to make both the surfaces flat and parallel. To study the electrical properties of the composites, both flat surfaces of the pellets were electroded with air-drying conducting silver paste. After electroding, the pellets were dried at 150°C for 4 h to remove moisture, if any, and then cooled to room temperature before taking any electrical measurement. The formation of composites were studied by an X-ray diffraction technique at room temperature with a powder diffractometer (Rigaku Miniflex, Japan) using CuK$_\alpha$ radiation ($\lambda = 1.5405$ Å) in a wide range of Bragg’s angles $2\theta (20° \leq \theta \leq 80°)$ with a scanning rate of 3°/min. The electrical parameters (impedance, modulus and capacitance) of the composites were measured using an LCR meter (HIOKI, Model-3532) in the frequency range of $10^2$ – $10^6$ Hz from 25 – 450°C.

3. Results and Discussion

3.1. Structural analysis

Figure 1 shows the XRD patterns of 0.5BiNd$_x$Fe$_{1-x}$O$_3$–0.5PbTiO$_3$ (BNxF$_{1-x}$–PT) with $x=0.05, 0.10, 0.15, 0.20$ at room temperature. The tetragonal crystal structure has been confirmed for all concentrations ranging from $x=0.05$ - 0.20. The composites show large tetragonality with increase in Nd concentration from 0.05-0.20 as it is expected from pure BiFeO$_3$–PbTiO$_3$ multiferroics composites [21]. It was found to be in good agreement between observed (obs) and calculated (cal) interplanar spacing $d = d_{obs} - d_{cal} = minimum$. The lat-
angular frequency and admittance and permittivity respectively, imaginary components of impedance, electrical modulus, \( Y^* \), complex permittivity \( \varepsilon^* \), complex electric modulus \( M^* \), which are related to each other:

\[
Z^* = Z' - jZ'' = \frac{1}{j\omega \varepsilon^*},
\]

\[
M^* = M' + jM'' = \frac{1}{\varepsilon^*} = j\omega C_0 Z^*,
\]

\[
Y^* = Y' + jY'' = j\omega C_0 \varepsilon^*,
\]

\[
\varepsilon^* = \varepsilon' - j\varepsilon''
\]

\[
\tan \delta = \frac{\varepsilon''}{\varepsilon'} = \frac{M''}{M'} = \frac{Z''}{Z'} = \frac{Y''}{Y'},
\]

where \( (Z', M', Y', \varepsilon') \) and \( (Z'', M'', Y'', \varepsilon'') \) are real and imaginary components of impedance, electrical modulus, admittance and permittivity respectively, \( \omega = 2\pi f \) is the angular frequency and \( j = \sqrt{-1} \) the imaginary factor. The complex impedance of "electrode/sample/electrode" configuration can be explained as the sum of a single with a parallel combination of RC (R=resistance, C=capacity) circuit. Thus, the result obtained using impedance analysis is an unambiguous, and hence provide a true picture of the electrical behavior of the material.

Table 1. Values of lattice parameters of 0.5BiNd,Fe_{1-x}O_{3−}0.5PbTiO_{3} \( x = 0.05 \) − 0.20

| Parameters Å | x=0.05 | x=0.10 | x=0.15 | x=0.20 |
|--------------|--------|--------|--------|--------|
| a            | 3.8805 | 3.9050 | 3.9076 | 3.9107 |
| c            | 4.2927 | 4.2049 | 4.1107 | 4.1029 |
| c/a          | 1.1062 | 1.0768 | 1.0520 | 1.0491 |

3.2. Impedance properties

Complex impedance spectroscopy (CIS) [24] is a unique and powerful technique to characterize the electrical behavior of a system. This analysis enables one to resolve the contributions of various processes such as the bulk, grain boundaries and electrode interface effects in the frequency domain. Generally, the data in the complex plane is represented in any of the four basic formalisms. These are complex impedance \( Z' \) , complex admittance \( Y' \) , complex permittivity \( \varepsilon' \) , complex electric modulus \( M' \) , which are related to each other:

\[
Z' = Z'' = \frac{1}{j\omega C_0 \varepsilon'},
\]

\[
M' = M'' = \frac{1}{\varepsilon'} = j\omega C_0 Z',
\]

\[
Y' = Y'' = j\omega C_0 \varepsilon',
\]

\[
\varepsilon' = \varepsilon' - j\varepsilon''
\]

\[
\tan \delta = \frac{\varepsilon''}{\varepsilon'} = \frac{M''}{M'} = \frac{Z''}{Z'} = \frac{Y''}{Y'},
\]

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where \( (Z', M', Y', \varepsilon') \) and \( (Z'', M'', Y'', \varepsilon'') \) are real and imaginary components of impedance, electrical modulus, admittance and permittivity respectively, \( \omega = 2\pi f \) is the angular frequency and \( j = \sqrt{-1} \) the imaginary factor. The complex impedance of "electrode/sample/electrode" configuration can be explained as the sum of a single with a parallel combination of RC (R=resistance, C=capacity) circuit. Thus, the result obtained using impedance analysis is an unambiguous, and hence provide a true picture of the electrical behavior of the material.

Figure 2 (a-d) shows the complex impedance spectrum \( Z' \) vs. \( Z'' \) of BN,Fe_{1-x}−PT with \( x = 0.05, 0.10, 0.15, 0.20 \) at different temperatures. Single semicircular arcs observed in a wide temperature \((200 - 275\degree C)\) region for different compositions. This confirms the presence of bulk effect in the composites even if increasing the percentage of Nd concentration. It is also observed that as the temperature increases the intercept point on the real axis shifts towards the origin which indicates that decrease in the resistive property i.e., called bulk resistance \( R_b \) of the composites [25, 26]. The electrical process taking place within the composites can be modeled (as an RC circuit) on the basis of the brick-layer model [24]. The impedance data did not fit well with single RC-combination, rather this fit well with equivalent circuits (insets of Fig. 2(a-d)) at 200\degree C for \( x = 0.05-0.20 \), and the fitting parameters of the circuit are shown in the table 2.

Figure 3(a-d) shows the variation of \( Z' \) as a function of frequency \((10^2 - 10^6 \text{ Hz})\) of BN,Fe_{1-x}−PT with \( x = 0.05, 0.10, 0.15, 0.20 \) at different temperatures. It is observed that the magnitude of \( Z' \) (bulk resistance) decreases on increasing temperature as well as Nd concentration in the low frequency ranges (up to a certain frequency), and thereafter appears to merge in the high-frequency region. This is possible due to the release of space charge polarization with rise in temperatures and frequencies [27]. This behavior shows that the conduction mechanism increases with increasing temperature and frequency (i.e., negative temperature coefficient of behavior like that of a semiconductor). The coincidence of the value of \( Z' \) at higher frequencies at all the temperatures indicates a possible release of space charge [28, 29] and the frequency at which the release of space charge occurs also depends upon the Nd concentration. The space charge polarization occurs maximum at higher frequency side for \( x = 0.20 \) concentration as compared to all other concentration. This may be due to the reduction in barrier properties of the materials with rise in temperature which is responsible for the enhancement of conductivity of the materials [30, 31]. At a particular frequency, \( Z' \) becomes independent of frequency. This type behavior is similar to the other material reported by Singh et al. [32].

Figure 4(a-d) shows the frequency-temperature dependence of \( Z'' \) (usually called as loss spectrum) of BN,Fe_{1-x}−PT with \( x = 0.05-0.20 \). The magnitude of \( Z'' \) decreases with increase in frequency as well as Nd concentration at high...
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Figure 2. (a – d) Complex impedance plot ($Z'$ vs $Z''$) in the temperature range 200 – 275°C with equivalent circuit (inset).

Table 2. Summarizing of fitting parameters corresponding to equivalent circuits at 200°C of Fig. 2(a-d).

| Different Parameters | X=0.05 | X=0.10 | X=0.15 | X=0.20 |
|----------------------|--------|--------|--------|--------|
| $R_1$                | 16517  | 9.9E-8 | 1.001E-7 | 51.67  |
| $R_2$                | 1.225E-5 | 4.546E-4 | 2.83E-5 | 1.434E-5 |
| $R_3$                | 1.172E-7 | 2.06E-5 | 1.176E-5 | 8588  |
| $R_4$                | 1476   |        |        |        |
| $C_1$                | 4.364E-11 | 7.028E-9 | 8.211E-10 | 1.116E-10 |
| $C_2$                | 5.52E-10 |        |        |        |
| CPE                  | 1.155E-9 | 9.885E-10 | 3.228E-10 | 1.802E-9 |
| Frequency Power(n)   | 0.5493  | 0.8718 | 0.9089 | 0.7211 |
| Chi square           | 0.002373 | 0.0156 | 0.01227 | 0.01415 |
| Warburg($\gamma_0$) | 1.413E-6 |        |        |        |

temperature region. The appearance of peaks in the loss spectrum at the high temperature region suggests the existence of relaxation process of the different compositions. This may due to the immobile species at low temperatures and defect or vacancies at high temperatures [32, 33]. It shows that with increasing Nd concentration and temperature, the magnitude of $Z''$ decreases and all the peaks shift towards the higher frequency side and finally, all the curves merge in high-frequency region. At higher frequencies, the contribution from the grain predominates owing to the absence of the space charge effects of the different compositions [34]. Generally, the impedance data were used to evaluate the relaxation time ($\tau$) of the electrical phenomena in the different compositions using the relation $\tau = 1/\omega = 1/2\pi f$, $f$ is the relaxation frequency. The variation of $\tau$ as a function of inverse of absolute temper-
Figure 3. (a–d) Variation of $Z'$ with frequency at different temperatures.

ature shown in the figure 5 calculated from impedance plot and it appears to be linear which follows the Arrhenius relation $\tau = \tau_0 \exp(-E_a/K_B T)$, where $\tau_0$ is the pre-exponential factor, $E_a$ is the activation energy, $K_B$ is the Boltzmann constant and $T$ is the absolute temperature. The activation energy calculated from the slope of $\log(\tau)$ against $10^3/T(K^{-1})$ curves of BN$_x$F$_{1-x}$-PT are 0.89, 0.76, 0.71, 0.70 eV for $x=0.05, 0.10, 0.15$ and 0.20 respectively.

4. Modulus properties

Figure 6(a–d) shows the complex modulus spectrum ($M'\text{ vs } M''$) of BN$_x$F$_{1-x}$-PT with $x=0.05, 0.10, 0.15, 0.20$ at different temperatures. Complex impedance spectrum gives more emphasis to elements with larger resistance whereas complex electric modulus plots highlight those with smaller capacitance. Using the complex electric modulus formalism, the inhomogeneous nature of the polycrystalline sample can be probed into bulk and grain boundary effects, which may not be distinguished from complex impedance plots. The complex electric modulus spectrum of the samples does not form semicircles. But slightly they exhibit deformed arcs with their centers positioned below the x-axis. The scaling behavior in the sample was studied by normalizing $M'/M'_{\text{max}}$ versus $\log_{10} f/f_{\text{max}}$ at different temperatures for different compositions as shown in the figure 6(a–d) (inset). This is called the master modulus curve. It gives an insight into the dielectric processes occurring inside the material. It is observed that there is a slight shift in the peak pattern and similar shape of spectrum with a slight variation in the full-width at half maximum (FWHM) with rise in temperature for the concentration $x=0.05-0.10$, whereas for the concentration 0.15 to 0.20, it is found that all the peaks merge in to one master curve at different temperature ranging from $200^\circ - 275^\circ$C. This suggest that the distribution of relaxation times is temperature independent [35, 36].

Figure 7(a–d) shows the variation of $M'$ as a function of frequency for BN$_x$F$_{1-x}$-PT with $x=0.05, 0.10, 0.15, 0.20$ at selected temperatures. For all the concentrations, it shows that $M'$ approaches to zero in the low frequency
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Figure 4. (a-d) Variation of $Z''$ with frequency at different temperatures.

Figure 5. Variation of relaxation time with inverse of temperature ($10^3/T$) calculated from impedance plot.

region, and a continuous dispersion on increasing frequency 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 [37]. This confirms elimination of electrode effect in the composites.

Figure 8(a-d) shows the variation of imaginary part of electric modulus with frequency for BN$_x$F$_{1-x}$ – PT (x=0.05-0.20) at selected temperatures. The maxima $M''_{\text{max}}$ shifts towards higher frequencies side with rise in temperature as well as Nd concentration ascribing correlation between motions of mobile ions [38] and suggests that the dielectric relaxation is thermally activated process. The asymmetric peak broadening indicates the spread of relaxation times with different time constant, and hence relaxation is of non-Debye type. The low frequency peaks show that the ions can move over long distances whereas high-frequency peaks merge to spatial confinement of ions in their potential well. The nature of modulus spectrum suggests the existence of hopping mechanism of electrical conduction in the materials. The variation of relaxation time ($\tau$) as a function of reciprocal of temperature ($1/T$) of BN$_x$F$_{1-x}$ – PT (x=0.05–0.20) at high temperature region is shown in figure 9 (calculated from modulus spectrum). This graph follows the Arrhenius relation, $\tau = \tau_0 \exp(-E_a/K_B T)$ where the symbols have their usual meanings and thermally activated process. The activation energy of the composites is found to be 0.81, 0.69, 0.68.
and 0.67 eV for x=0.05, 0.10, 0.15 and 0.20 respectively. The activation energies calculated from both impedance and modulus plot found to similar.

5. Electrical conductivity

The ac electrical conductivity ($\sigma_{ac}$) was calculated from the dielectric data using the empirical relation: $\sigma_{ac} = \omega \varepsilon_0 \tan \delta$, where $\varepsilon_0$ is the vacuum dielectric constant and $\omega (= 2\pi f)$ is the angular frequency.

Figure 10 shows the variation of $\sigma_{ac}$ with inverse of absolute temperature ($10^3/T$) of BN$_x$F$_{1−x}$ − PT (x=0.05-0.20) at a particular frequency (1 MHz).

The nature of variation of $\sigma_{ac}$ over a wide temperature range supports the thermally activated transport properties of the materials obeying Arrhenius equation $\sigma_{ac} = \sigma_0 \exp(-E_a/K_B T)$, where $\sigma_0$ is the pre-exponential factor and other symbols have their usual meaning. It is observed that the ac conductivity of the material increases with rise in temperature and show the NTCR behavior. This behavior suggests that the conduction mechanism of the compounds may be due to the hopping of charge carrier. The values of activation energy of the composites are found to be 0.28, 0.27, 0.31 and 0.32 eV for different compositions x=0.05-0.20 respectively.

The frequency–temperature dependence of ac conductivity is shown in Figure 11(a-d). This figure shows that the conductivity for the concentration x=0.10, 0.15, 0.20 exhibit step like decrease in low frequency region suggest low frequency polarization which may be due to the transition from the bulk to contact resistance [28]. There is a deviation in the slope at a particular frequency usually called hopping frequency. This typical behavior suggests the presence of hopping mechanism in the composites. This type of conducting behavior is well described by Jonscher’s universal power law [39]: $\sigma(\omega) = \sigma_{dc} + A\omega^n$ where n is the frequency exponent with 0 < n < 1 and A is the temperature dependent pre-exponential factor. The term $A\omega^n$ comprises the ac dependence and characterizes all dispersion phenomena. The exponent n can vary differently from material to material, depending on temperature. This suggests that the electrical conduction in BN$_x$F$_{1−x}$ − PT is a thermally activated process. The non linear curve well fit to Jonscher’s power law for all the composites with x=0.05, 0.10, 0.15, 0.20 at different tem-
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Figure 7. (a–d) Variation of $M'$ as a function of frequency for BN$_x$F$_{1-x}$ – PT with $x = 0.05 – 0.20$ at selected temperatures.

Figure 8. (a–d) Variation of imaginary part of electric modulus with frequency for BN$_x$F$_{1-x}$ – PT ($x = 0.05 – 0.20$) at selected temperatures.
Table 3. Fitting parameters obtained from the Jonscher’s power law at different temperatures.

| T (°C) | x=0.05   | x=0.10   | x=0.15   | x=0.20   |
|--------|-----------|-----------|-----------|-----------|
|        | A (×10⁻⁹) | A (×10⁻⁹) | A (×10⁻⁹) | A (×10⁻⁹) |
| 200    | 3.94      | 0.84      | 1.93      | 0.86      |
| 225    | 2.80      | 0.88      | 6.5       | 0.96      |
| 250    | 3.18      | 0.88      | 1.71      | 1.07      |
| 275    | 2.16      | 0.92      | 1.00      | 1.12      |

Figure 9. Variation of relaxation time (τ) as a function of reciprocal of temperature (1/°C) of BN$_x$F$_{1-x}$–PT (x=0.05–0.20) at different temperature region calculated from modulus spectrum.

Figure 10. Variation of ac conductivity with inverse of temperature (10⁴/T) of BN$_x$F$_{1-x}$–PT (x=0.05–0.20) at different temperature region shown in figure 11(a-d). The fitting parameter A, n are calculated from the non-linear fitting shown in table 3.

6. Conclusions

The BN$_x$F$_{1-x}$–PT (x = 0.05, 0.10, 0.15, 0.20) composites were prepared by a high-temperature solid state reaction technique. X-ray structural analysis confirmed the composites show tetragonal structure at room temperature. The crystallite size of the composites are found to be in the range of 10-14 nm. Complex impedance spectroscopy was used to characterize the electrical properties of the materials. The electrical conduction in the composites is due to the bulk effects only. The bulk resistance decreases with rise in temperature and exhibit a NTCR behavior. The electrical modulus has confirmed the presence of a hopping mechanism in the composites. The scaling behavior of modulus spectra demonstrates that the distribution of relaxation time is temperature independent. The dc conductivity shows a typical Arrhenius type of electrical conductivity. The activation energy calculated both from impedance and modulus spectra are comparable and is of the same order found to be in the range 0.70-0.89 eV. The ac conductivity spectrum was found to obey Jonscher’s universal power law.
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Figure 11. (a – d) Variation of ac conductivity with frequency at different temperature.

Figure 12. Variation of dc conductivity with inverse of temperature.

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