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Effective complex permittivity and AC conductivity of $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}$ TiO_3-PVDF 0-3 composite

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ABSTRACT

The present study addresses the problem of quantitative prediction of effective complex relative permittivity and ac conductivity of $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO_3$-Polyvenylidene Fluoride (BNBT06-PVDF) 0-3 composite samples (prepared by solution cast method at an elevated temperature) having 10, 20 and 30 volume percentage of BNBT06 powder. SEM micrographs of the fractured surfaces showed that the particle distribution in the grains is not strictly homogeneous. Some areas of agglomeration of particles in the grains are also seen in the micrographs. EDAX patterns confirmed the presence of different constituent elements of the composite samples. The resulting data for room temperature real and imaginary parts of relative permittivity as well as real part of ac conductivity showed an increasing trend with increasing volume fraction of the ceramic filler. The 30 vol. % of BNBT06-PVDF composite had the highest dielectric constant of 75.3 and dielectric loss of ~6.09 *i.e.*, loss tangent ~0.08. Among the dielectric mixing models presented, Rother-Lichtenecker model showed the best fit to the experimental data for the test composite. Similar equations for effective ac conductivity in terms of conductivity of the constituent phases of the composite were proposed in the present work to be fitted to find that none except Rother-Lichtenecker equation fitted the experimental data well. First order exponential growth type of equation applicable to all the three properties fitting the experimental data excellently is also proposed in the present work. Copyright © 2014 VBRI press.

Keywords: Ceramic-polymer 0-3 composites; complex permittivity; Rother-Lichtenecker model; AC conductivity; effective medium theory.

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Introduction

Ceramic-polymer composites form a new class of construction and functional materials of great potential application in having combined hardness and stiffness of ceramics and flexibility, elasticity, low density, and high breakdown strength of polymers. Consequently, ceramic-polymer composites are being increasingly utilized for their specific dielectric, ferroelectric, piezoelectric, pyroelectric, electro-optic as well as superconducting properties in micro-devices [**1**]. Piezoelectric ceramics have, in general, large dielectric constant, high piezoelectric charge coefficient as well as acoustic impedance whereas polymers have generally low dielectric constant and acoustic impedance close to that of water as well as human body tissue. This low value of acoustic impedance combined with higher values of piezoelectric voltage coefficients for piezoelectric polymers makes them suitable candidates for mixing with piezoelectric ceramics in order to make good strain-monitoring and hydrostatic sonar sensors [**2**]. Further, these ceramic-polymer composite materials have been suggested to be viable alternative tools in piezoelectric and pyroelectric transducer applications [**3**]. In short, these systems form the current area of active research activities and have received intensive global attention [**4-6**] because these composites can be prepared with excellent dielectric and mechanical properties at low temperature conditions and can be designed according to specific requirements by tailoring the relative fraction of the starting materials. One of the problems inherent in composite systems has been to predict their macroscopic properties using the properties of the constituents. Many theoretical investigations on binary systems have been performed in regard to the dielectric constant, piezoelectric constant, and the elastic constant [**7-10**]. Strong piezoelectricity for the composite medium is a prerequisite for it to be a useful one. Towards a step forward in providing a lead-free perovskite ceramic material having optimal dielectric and piezoelectric properties, Wang et al [**11**], Chen et. al. [**12**], Lam et. al. [**13**], Rawat et. al. [**14**] as well as the present group of workers [**15**] reported that the composition $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO_3$ (BNBT06), which is near the morphotropic phase boundary (MPB) composition, is one of the most suited materials. As far as the polymer phase of the bi-phasic composite(s) is concerned, piezoelectric polymer PVDF, despite many of its deficient properties, has appeared appealing to numerous industries for its inexpensive, lightweight, biologically compatible and mechanically stable structures. It can undertake large amount of deformation while sustaining large forces. It has expeditious response time, very low density, and distinguished flexibility when compared to those of electro-active ceramics and shape memory alloys. The piezoelectric PVDF and its copolymers are widely applied materials in both actuation and sensing mechanisms. They can be utilized as fibers and films mostly in linear movement requirements in various engineering applications such as active micro air vehicle wings, piezo-laminated columns [**16**], and shape correction films in space applications [**17**]. Other applications include Proton Exchange Membranes [**18**], filtration membranes [**19**], structural health monitoring [**20**], endoscopic tactile sensors [**21**], and macro-fluidic control [**22**]. An extensive literature survey revealed that a systematic study of dielectric properties and electrical conductivity for BNBT06-PVDF 0-3 ceramic-polymer composites has not been undertaken in the recent past. In view of the aforesaid facts, the present work relating to the study of the complex relative permittivity and ac conductivity for BNBT06-PVDF 0-3 composite samples having 10, 20 and 30 volume percent of BNBT06 powder has been undertaken.

Experimental

Synthesis of ceramic powder and composites

Polycrystalline ceramic samples of $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO_3$ were prepared by a standard high-temperature solid-state reaction technique using oxides/carbonates: Bi_2O_3, Na_2CO_3, $BaCO_3$ and TiO_2 (Hi-Media) (having purity > 99.5%) in a suitable stoichiometry. The above ingredients were mixed thoroughly, first in air and then in methanol medium, using an agate mortar and pestle. The mixtures were calcined at an optimized temperature of 1160ºC for about 3h. BNBT06-PVDF 0-3 ceramic-polymer composites having 10, 20 and 30 volume percent of BNBT06 powder were fabricated using solution cast method at an elevated temperature (~70^{o}C-80^{o}C) under constant stirring. Solvent cast disk-shaped samples were obtained by first dissolving PVDF in N-N Dimethyl formamide (DMF) at 70^{o}C on a hot plate of a magnetic stirrer for several minutes, until complete dissolution of the polymer. Then, the calcined BNBT06 powder was added slowly to the thoroughly stirred solution in order to avoid powder sedimentation or agglomeration. The composite samples in the molten (viscous) state were poured into shallow cylindrical as well as rectangular stainless steel dies and then were allowed to dry for a few days in open air. The fabricated composites were cut into the disk-shaped pieces of suitable thicknesses and were further allowed to dry before any measurements on them were made. The green samples were then polished and painted on both sides by the use of AR-grade silver paste (Ted Pele, USA).

Characterization

The XRD spectra were observed on calcined powder of BNBT06 with an X-ray diffractometer (X'pert-PRO, USA) at room temperature, using CuK_α radiation (λ=1.5405 Å) over a broad range of Bragg angles (20º≤2θ≤80º). The microstructure of the calcined ceramic powder was studied at room temperature from the micrographs obtained by using a scanning electron microscope (JEOL-JSM840A). BNBT06-PVDF 0-3 composite samples having 10, 20 and 30 volume percentage of BNBT06 powder were prepared by solution cast method (using DMF as the solvent for PVDF) at an elevated temperature, as detailed before. The room temperature dielectric constant (ε_r) and loss tangent (tanδ) (and hence dielectric loss) at 1kHz were measured using sample thickness and surface area along with the capacitance data provided by a computer interfaced LCR Hi-Tester (HIOKI 3532-50, Japan) on a symmetrical cell of type Ag|*Composite*|Ag, where Ag is a conductive paint coated on either side of the pellet.

Theoretical background

Real part of relative permittivity

Several predictive equations for effective relative permittivity have been proposed in one of our previous works [**23**]. In another work by other group on $BaTiO_3$–thermosetting epoxy resin composite, it was shown that Lichtenecker mixing rule or the logarithmic rule of mixing for random media provided the best fitting prediction of effective relative permittivity [**24**]. In a bid to obtain the more acceptable equation(s) for the given composite, a number of test equations chosen from the literature are given as follows:

1. Rother-Lichtenecker equation or Cuming equation [**25**]:

$$\varepsilon_{eff} = \exp(\Sigma f_i \ln \varepsilon_i) \qquad (1)$$

where ε_i is the relative permittivity of the i^{th} component of the mixture and f_i is the volume fraction of the component.

2. Modified Rother-Lichtenecker equation [**26**]:

$$\varepsilon_{eff} = \exp[\ln \varepsilon_1 + f_2(1-k)\ln(\varepsilon_2/\varepsilon_1)] \qquad (2)$$

Eqs. (1) and (2) are nothing but general representations of logarithmic law of mixing for a chaotic or statistical mixture. The Eq. (1) does not contain a shape-dependent parameter whereas Eq. (2) contains it as '*k*'.

3. Webman equation (WE) or Furukawa equation (FE) or equation from Effective Medium Theory (EMTE) [**27-28**] is given as:

$$\varepsilon_{eff} = \varepsilon_B \frac{1+2f_A\{(\varepsilon_A-\varepsilon_B)/(\varepsilon_A+2\varepsilon_B)\}}{1-f_A\{(\varepsilon_A-\varepsilon_B)/(\varepsilon_A+2\varepsilon_B)\}} \qquad (3a)$$

and Hashin-Shtrikman composite cylinder model, [**29**] as:

$$\varepsilon_{eff} = \frac{\varepsilon_1[(1+2f)\varepsilon_2 + 2\varepsilon_1(1-f)]}{\varepsilon_2(1-f)+(2+f)\varepsilon_1} \qquad (3b)$$

4. Bruggeman equation or modified EMT equation or Dias-Dasgupta equation using Yamada model or Rao equation, all commonly designated as MEMTE [**30-31**] is given as:

$$\varepsilon_{eff} = \varepsilon_B\left[1+\frac{f_A(\varepsilon_A-\varepsilon_B)}{\varepsilon_B+n(1-f_A)(\varepsilon_A-\varepsilon_B)}\right] \qquad (4a)$$

where *n* is the morphology factor. Interchanging the subscripts *A* and *B*, changing the notations, and rearranging the terms, one finally gets:

$$\varepsilon_{eff} = \varepsilon_1\left[1+\frac{f(\varepsilon_2-\varepsilon_1)}{\varepsilon_1+n(1-f)(\varepsilon_2-\varepsilon_1)}\right] \qquad (4b)$$

5. Jayasundere-Smith equation [**32**]:

$$\varepsilon_{eff} = \frac{[\{\varepsilon_1 f_1 + \varepsilon_2 f_2(3\varepsilon_1/(\varepsilon_2+2\varepsilon_1))\}\{1+3f_2(\varepsilon_2-\varepsilon_1)/(\varepsilon_2+2\varepsilon_1)\}]}{[\{f_1+f_2(3\varepsilon_1/(\varepsilon_2+2\varepsilon_1))\}\{1+3f_2(\varepsilon_2-\varepsilon_1)/(\varepsilon_2+2\varepsilon_1)\}]} \qquad (5)$$

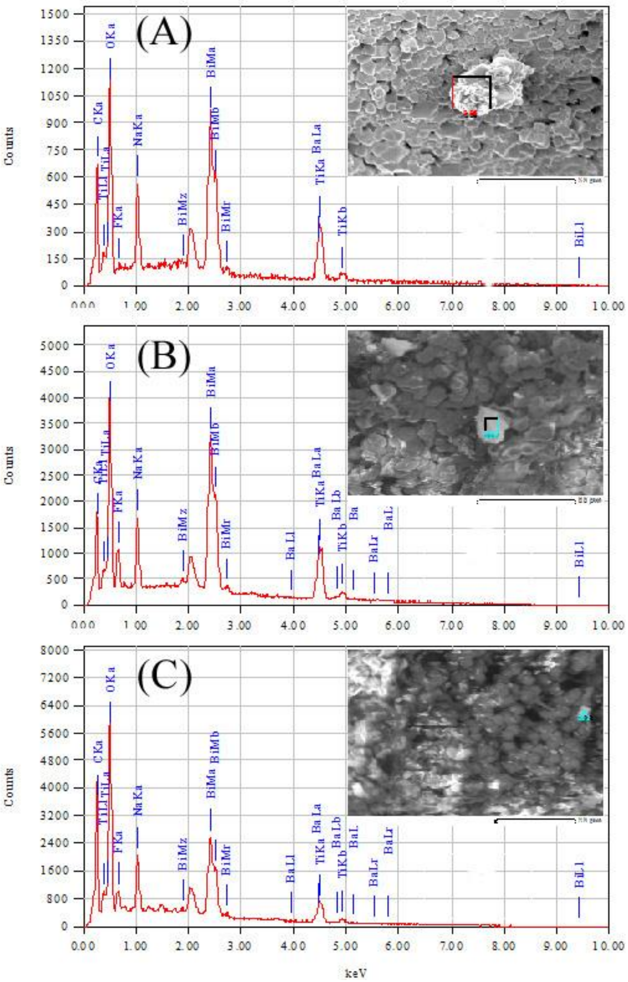

Fig. 1. EDAX pattern and SEM micrograph (inset) of BNBT06-PVDF 0-3 composites with (A) 10, (B) 20 and (C) 30 vol.% of BNBT06.

Imaginary part of relative permittivity

The equations governing the filler-concentration dependent variation of effective dielectric loss factor of composites, unlike those for dielectric constant, are fewer in number. However, an equation given by Bruggeman [**31**] reads as follows:

$$\varepsilon''_{eff} = \varepsilon'' = \frac{\varepsilon''_p[(\varepsilon'_c-\varepsilon')(\varepsilon'_c+2\varepsilon'_p)\varepsilon']}{[(\varepsilon'_c-\varepsilon'_p)(\varepsilon'_c+2\varepsilon')\varepsilon'_p]} + \varepsilon''_c\frac{3(\varepsilon'-\varepsilon'_p)\varepsilon'}{(\varepsilon'_c-\varepsilon'_p)(\varepsilon'_c+2\varepsilon')} \qquad (6)$$

where the subscripts '*c*' and '*p*' represent 'ceramic' and 'polymer' respectively, and the quantity without a subscript refers to the composite. The above sole equation paved the way for using all the above five equations (Eqs.1-5) for effective relative permittivity (= ε') to be put in Eq. (6) to get the corresponding five values of dielectric loss factor (= ε'') for their comparison with experimental results for effective loss factor of the composite to get the best fitting prediction equation(s).

Real part of ac conductivity

The real part of ac conductivity is given by:

$$\sigma'_{ac} = \omega\varepsilon_o\varepsilon'' \qquad (7)$$

where $\omega = 2\pi f$, (f being the frequency used); ε_o is the permittivity of free space (= 8.854 x 10^{-12} Fm^{-1}) and ε'' is the dielectric loss factor, as referred to earlier. Thus, it is directly related to the dielectric properties of the material.

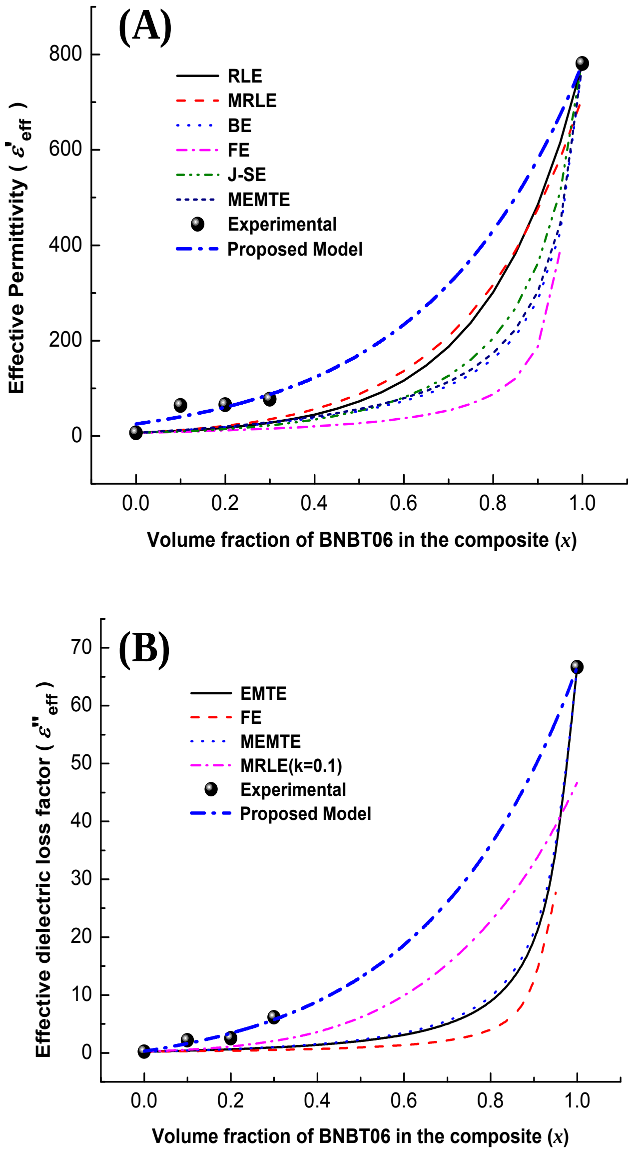

Fig. 2. Filler (BNBT06) concentration dependence of experimental and theoretical (A) effective dielectric constant and (B) effective dielectric loss factor of BNBT06-PVDF 0-3 composite measured at 1kHz.

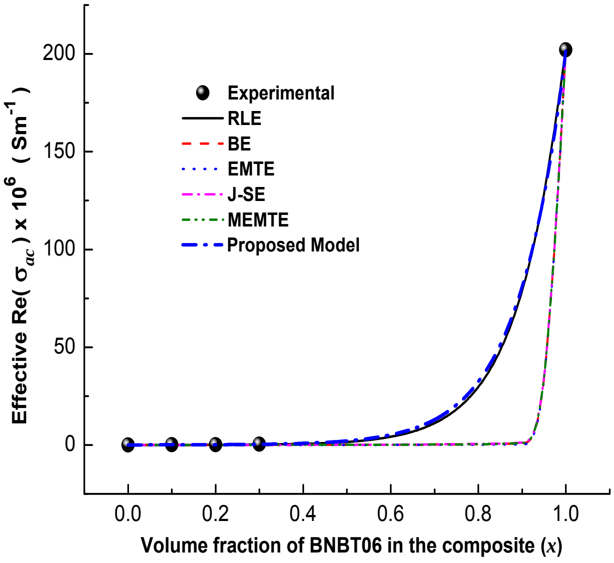

Fig. 3. Filler (BNBT06) concentration dependence of experimental and theoretical effective real part of ac conductivity of BNBT06-PVDF 0-3 composite measured at 1 kHz.

Results and discussion

Fig.1 shows the EDAX pattern and SEM micrograph (inset) for BNBT06-PVDF 0-3 composites with (a) 10, (b) 20 and (c) 30 vol. % of BNBT06 ceramic fillers. **Fig. 2** (a) and (b), respectively, show the filler concentration dependence of experimental as well as theoretically predicted acceptable results for effective relative permittivity and dielectric loss factor of the same composite. Comparison of theoretically derived results for filler concentration dependent variation of effective relative permittivity of the composite with experimental results showed that none out of at least five independent test equations proven themselves to be treated as acceptable. For dielectric loss factor, it was observed that RLE, J-SE, EMTE, and MEMTE (n = 0.1) did not show their acceptability for this variation. On the other hand, only MRLE (k = 0.1) showed tolerably good acceptability.

In **Fig. 3** the filler concentration dependence of real part of complex ac conductivity for the BNBT06-PVDF composite is shown in which it was observed that RLE, J-SE, EMTE and MEMTE (n = 0.1) did show their acceptability up to the experimental range for this variation i.e., up to 30 vol. %, while only RLE showed tolerably good acceptability over the entire range of volume fraction. On the basis of the above analyses it transpired that mixture equations for effective complex permittivity did not agree well with the experimental results for the test ceramic-polymer composite material for the present study. In a bid to provide new acceptable model for the ceramic (filler) concentration dependent variation of complex permittivity and real part of ac conductivity of the test composite material under the present study the first order exponential growth type of mathematical model in the form $Y = Y_o + A\exp(x/t)$ (where Y_o, A and t or β (= $1/t$) are the model parameters and x is the volume fraction of ceramic in the composite) was found suitable. As shown in **Table 1**, the values of r^2 are approaching unity for the test material and for all the three material properties *viz*. real and imaginary parts of complex permittivity and real part of complex ac conductivity, thereby showing good agreement between theoretical and experimental results.

Table 1. Fitting parameters for the first order exponential growth regression equation applied to experimental data of effective relative permittivity, dielectric loss and real part of ac conductivity of BNBT06-PVDF 0-3 ceramic-polymer composites at 1 kHz.

Equation → *Parameters →*	*Exponential growth: $Y=Y_0+A\exp(x/t)$* Y_0	 A	 t	 r^2
ε′, BNBT06-PVDF 0-3 composite	-20.09775	45.34509	0.34825	0.99518
ε″, BNBT06-PVDF 0-3 composite	-3.72271	4.00656	0.34894	0.99926
Re(σ_{ac}), BNBT06-PVDF 0-3 composite	1.52127E-7	1.69222E-10	0.07147	1.00

In the present context, the term (Y_o+A) corresponds to the value of three material properties at x = 0 *i.e*. for the polymer matrix, while the term [$Y_o+A\exp(\beta)$] for x = 1 corresponds to the values of the same properties for the ceramic filler. Here β may be designated as the filler concentration dependent "permittivity/ac conductivity growth parameter". It is observed that the experimental data fit excellently well (r^2 > 0.99, Table 1) with the proposed theoretical model. Further, almost the same order of values of t ($=1/\beta$) ≈ 0.349 for real as well as imaginary parts of relative permittivity indicate the same nature of growth for them and different in case of real part of ac conductivity in having a value of 0.071. From the study it transpired that if quite different types of ceramic fillers as well as polymer matrices were chosen as the test materials, the filler concentration dependent permittivity/ac conductivity growth parameter (β) would certainly assume different values in view of the fact that at least due to the different nature of densification of the samples (lighter polymer particles being replaced by denser ceramic particles) as the ceramic concentration is increased. The growth may also be ascribed to the different ratios of the permittivity and ac conductivity values for ceramic and polymer phases of the composite (which is generally greater than 1). Thus, it is expected that the values of β for different test composite materials might throw some light on the nature (sharp or flat) of such type of variation and could thus assume a type of calibration parameter, which may be useful for further study.

Conclusion

The effective complex relative permittivity and real part of complex ac conductivity of BNBT06-PVDF 0-3 ceramic-polymer biphasic composite system as functions of broad compositional range have been investigated and the experimental results have been compared with the theoretical results derived from various predictive equations. The study revealed that only the Rother-Lichtenecker model or the logarithmic law of mixing for chaotic mixtures did fit the experimental data for real part of complex ac conductivity well, whereas for complex permittivity none of the test mixture equations could provide acceptable results. The first order exponential growth type of model provided the most acceptable fits with experimental results for effective complex permittivity as well as for real part of complex ac conductivity of BNBT06-PVDF 0-3 ceramic-polymer composite system. From the study it may be opined that the permittivity/ac conductivity growth parameter, β, used in first order exponential growth type of model might throw some light on the nature of ceramic filler used in the composite.

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