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# Dielectric and electrical properties of different inorganic nanoparticles dispersed phase separated polymeric nanocomposite bilayer films

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Phase separated polymer nanocomposite (PNC) bilayer films consisting of poly(vinyl alcohol) (PVA) and poly(methyl methacrylate) (PMMA) matrices (PVA/PMMA = 50/50 wt %) dispersed with 5 wt % amount of inorganic nanoparticles (i.e., $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) have been prepared by solution-cast method using the polymer blend binary solutions. Complex dielectric permittivity, electric modulus, alternating current (AC) electrical conductivity and impedance spectra of the PNC bilayer films have been investigated in the frequency range from 20 Hz to 1 MHz, at 27°C, by employing the dielectric relaxation spectroscopy. The values of real part of dielectric permittivity of all these films are found less than 2.5, and their dielectric loss part values are significantly low confirming the suitability of these PNC materials as high performance low-permittivity nanodielectrics. The complex dielectric permittivity values of these PNC bilayer films vary with the nanoparticles in the order $SnO_2 > ZnO > SiO_2 > Al_2O_3$. The complex dielectric permittivity values of $Al_2O_3$ and $SiO_2$ dispersed films are found less than $1.33–j0.035$ and also remain almost stable over the entire frequency range of measurements. The AC electrical conductivity and impedance values of these films have linear increase and decrease, respectively, with the increase of frequency which confirms their Ohmic behaviour. The direct current (DC) electrical conductivity values of the PNC bilayer films are found of the order of $10^{-14}$ S/cm, and these values have variation of about one order of magnitude with different nanofillers. The dependence of dielectric and electrical properties of the PNC films on the structural and dielectric permittivity of the nanofillers has been explored. The feasibility of these PNC bilayer films as polymer dielectrics, electrical insulator and dielectric substrate in the development of advanced microelectronic devices has been confirmed.

**Keywords:** Polymer nanocomposites, Binary film, Dielectric properties, Electrical conductivity, Nanodielectric

Among the synthetic polymers, poly(vinyl alcohol) (PVA) and poly(methyl methacrylate) (PMMA) have enormous technological applications in the pristine form and also the various nanoparticles (NPs) dispersed their polymer nanocomposites (PNCs)[1–12]. PVA is a biodegradable, non-toxic, hydrophilic and semicrystalline material, and it forms optically transparent and flexible-type film prepared by solution-cast method. The presence of hydroxyl groups in the carbon chain backbone of PVA macromolecules acts as functional group which establishes the intra- and inter-molecular hydrogen bonding in the pristine PVA film[13], and also forms interfacial interactions with inorganic nanofillers in the PNC materials[4-10]. The PMMA is hydrophobic and amorphous material, and its film prepared by solution-cast method has excellent optical transparency, but brittle nature under loaded force[14]. PMMA chain repeat units have carbonyl groups which behave as functional groups to form various interfacial interactions with the inorganic nanofiller. PVA and PMMA films are frequently used as gate insulator in organic thin film transistors (OTFTs) and organic field effect transistor (OFETs)[15-20] and in fabrication of thin film capacitors[21]. These polymers are also used in the preparation of PNC films dispersed with semiconducting nanoparticles of wide band-gap for their opto-electronic applications[7-10,12]. As compared to the PVA film, the PMMA film has relatively good thermal stability, weather resistance, chemical inertness and has low dielectric permittivity which makes PMMA a crucial material for its use as an polymer dielectric and flexible dielectric substrate for development of various microelectronic devices and sensors[16-18,20-23]. From the results on these polymers, it seems that the blend of PVA and PMMA can also serve as a better polymeric material of tailored properties for various electronic and electrical industrial uses.

In search of advanced polymeric materials of required technological properties, much effort has been directed towards the characterization of polymers blends to take advantages of the inherent merits of pristine polymers[24-28]. The polymer blending comes under green chemistry technique which is simple, effective and inexpensive. The miscibility is one of the most important factors which governs the various properties of the polymer blend matrix[26,27]. It is strongly dependent on the method of blend preparation, and also quite dependent on the polymers composition used in the preparation of blend. Characterization of PVA–PMMA blend miscibility, thermal properties and structural phases with varying composition has been intense topic of research for the enhancement of various applications of these polymers[29-34]. The atomistic simulation and thermodynamic study of the PVA–PMMA blend confirmed that this blend has compatibility for compositions[29] containing greater than 60 wt% of PVA. The dynamic mechanical analysis of PVA–PMMA blend prepared by melt kneading was also confirmed as incompatible for the PMMA-rich phase[31]. But the blend of PVA–PMMA forms good membrane at 5 wt% of PMMA through cross-linkage via the intermolecular interaction[30] of hydroxyl (OH) groups in PVA with the carbonyl (C=O) groups in PMMA. The doped lithium ions in the PVA–PMMA blend form good compatibility which is due to cross-linkage of PVA and PMMA chain segments through ion-dipolar coordinations, and such materials have good ionic conductivity confirming their suitability as electrolyte material for lithium ion batteries[35]. The PVA–PMMA blend compatibility is also reported using boric acid as a cross-linking agent for their biomedical applications[33]. The PVA–PMMA bilayer dielectric had been realized in fabrication[18] of OFETs. The dielectric properties of tin micro powder doped PVA–PMMA blend composite films have also been investigated for their various applications in microelectronic industries[36]. Further, the properties of PVA–PMMA blend can be enhanced by proper dispersion of inorganic nanofillers in the blend matrices.

The inorganic nanoparticles (NPs) (i.e., silica $SiO_2$, alumina $Al_2O_3$, zinc oxide ZnO and tin oxide $SnO_2$) dispersed PVA[5–8] and PMMA[12,16,17,20-23,37-39] matrices and also PVA–PMMA comb-polymer[40] based PNCs had been extensively investigated for their polymer dielectrics and opto-electronics applications. Among these NPs, the ZnO and $SnO_2$ are semiconductor materials of wide band-gap, whereas $SiO_2$ and $Al_2O_3$ are electrical insulating ceramic materials. The dielectric properties of the PNC materials vary significantly when these NPs are used as nanofiller in the polymers matrices. The interfacial interactions formed between the NPs and the dipolar group/s (functional group/s) of the polymer chain establish the various useful properties of the PNC materials especially as polymeric nanodielectrics[41,42]. Survey of literature reveals that the dielectric and electrical properties of $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$ nanoparticles dispersed PVA–PMMA blend based PNC films have not been attempted so far.

In order to understand the effect of various inorganic NPs on the PVA–PMMA blend compatibility and dielectric and electrical properties, the binary solution of these polymers (50/50 wt %) dispersed with 5 wt % NPs (i.e., $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) were used for the preparation of polymer blend based nanocomposites (i.e., PVA–PMMA–5 wt % NP). It has been found that these binary polymeric solutions form the phase separated PNC bilayer films. The detailed dielectric and electrical properties of these PNC films have been investigated to explore their suitability as flexible-type polymeric nanodielectrics and also as electrical insulators for advancement of audio and radio frequency voltage operated microelectronic devices.

## Experimental Section

### Sample preparation

PVA ($M_w$ = $77\times10^3$ g $mol^{-1}$) of Loba Chemie, India, and PMMA ($M_w$ = $3.5\times10^5$ g $mol^{-1}$) and $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$ nano powder of different particle sizes were obtained from Sigma-Aldrich, USA. The structural properties and dielectric permittivity values of these nanopaticle materials are given in Table 1. Anhydrous tetrahydrofuran (Loba Chemie, India) and double distilled deionized water of Millipore have

Table 1 — Structural properties and dielectric permittivity of $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$ nanoparticles used in preparation of phase separated PNC bilayer films

| Nanoparticle (NP) | Structure | Particle size[a] (nm) | Dielectric permittivity $\varepsilon_r$ (at 1 MHz and 27°C) |
|---|---|---|---|
| $Al_2O_3$ | semicrystalline ($\gamma$-phase) | <50 | 9.7 [47] |
| $SiO_2$ | amorphous | <15 | 3.8 [48] |
| ZnO | crystalline (wurtzite) | <100 | 10.26 [49] |
| $SnO_2$ | crystalline (rutile-phase) | <100 | 34.5 [50] |

[a]Values are given by the Sigma-Aldrich, USA

been used as solvents. Initially, PVA and PMMA each of 0.5 g were dissolved in tetrahydrofuran and water, respectively, in separate glass bottles, and then these polymeric solutions were mixed to obtain the PVA–PMMA blend binary solution. The required amount for 5 wt % for each nanofiller was firstly suspended into water under magnetic stirring. This suspended nanoparticle solution was then homogeneously mixed with the polymer blend binary solution under continuous magnetic stirring and finally casted on to a poly propylene dish. The room temperature dried solution results in phase separated PNC bilayer film. The formation of phase separated PNC bilayer film reveals that the PVA and PMMA of 50/50 wt % composition are incompatible blend even on the dispersion of different nanoparticles. Further, a huge difference in rate of evaporation process of tetrahydrofuran and water solvents, the phase separated bilayer film of PVA and PMMA are formed one above another during drying of the casted polymer binary solution. The PNC bilayer films of all the nanoparticles dispersed PVA–PMMA were prepared by following the same procedure as mentioned above. These PNC bilayer films were dried 40°C for 24 h prior to their dielectric and electrical measurements.

### Characterizations

Dielectric and electrical properties of the PNC bilayer films were determined by employing the dielectric relaxation spectroscopy (DRS) over the frequency range from 20 Hz to 1 MHz, at 27°C. Frequency dependent values of capacitance $C_p$, resistance $R_p$ and loss tangent (tanδ = ε″/ε′ ) of the each PNC bilayer film sandwiched between the circular electrodes of dielectric test fixture were measured in parallel circuit operation mode using an Agilent technologies 4284A precision LCR meter and 16451B solid dielectric test fixture. A microprocessor-controlled oven was used for measurements at constant temperature by placing the PNC film loaded dielectric test fixture in the cavity of oven. Prior to sample measurements, the open circuit calibration of the cell was performed in order to eliminate the effect of stray capacitance of the cell leads. The spectra of complex dielectric permittivity $\varepsilon^*(\omega) = \varepsilon' - j\varepsilon''$, alternating current (AC) electrical conductivity $\sigma^*(\omega) = \sigma' + j\sigma''$, electric modulus $M^*(\omega) = M' + jM''$ and impedance $Z^*(\omega) = Z' - jZ''$ of the PNC bilayer films were evaluated using the expressions based on $C_p$ and $R_p$ as described in detail in the earlier publication[28].

## Results and Discussion

### Dielectric spectra

Figure 1 depicts the frequency dependent complex dielectric permittivity (real part ε′ and loss part ε″) and the loss tangent (tanδ = ε″/ε′) values of PVA–PMMA–5 wt % NP bilayer films (where NP is $Al_2O_3$, $SiO_2$, ZnO or $SnO_2$), at 27°C. The ε′ and ε″ values of a dielectric material represent the electrical energy storage ability and average energy loss in the material and therefore, the study of ε′ and ε″ values is very important for ultimate industrial applications of the material[41-44]. The ε′ and ε″ values of the investigated PNC bilayer films at some selective frequencies are

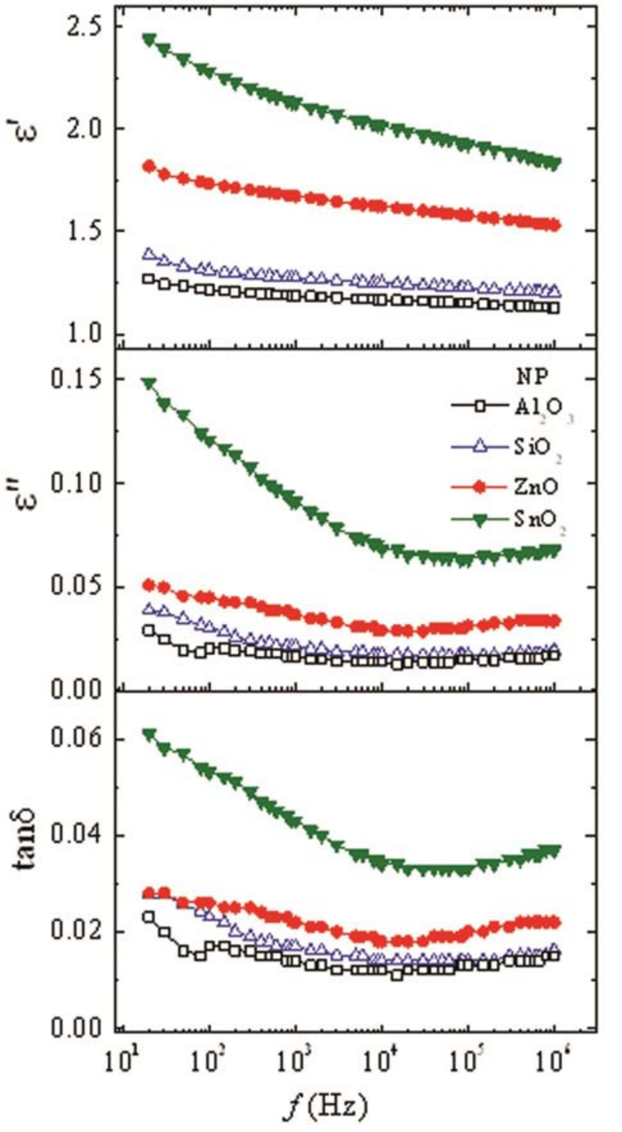

Fig. 1 — Complex dielectric permittivity (real part ε′ and loss part ε″) and dielectric loss tangent (tanδ) spectra of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at 27°C

recorded in Table 2 for the benefit of readers and also selection of the material for industrial uses. Figure 1 shows that the ε′ values decrease non-linearly with increase of frequency, but the rate of decrease is found relatively high for $SnO_2$ dispersed film. Table 2 shows that the decrease of complex permittivity values with increase of frequency in high frequency region (at radio frequencies) is insignificant for the $Al_2O_3$, $SiO_2$ and ZnO dispersed PNC films, and therefore, these materials can serve as stable and low permittivity polymer dielectrics at radio frequencies. The slight decrease in ε′ values with increase of frequency may be owing to the contribution of interfacial polarization (IP) effect which is also called Maxwell-Wagner-Sillars (MWS) effect and is commonly exhibited in PNC materials[4,11,28,45,46].

Figure 1 and Table 2 show that the frequency dependent ε′ values of these PNC bilayer films depend on the structural and dielectric permittivity[47-50] $\varepsilon_r$ of the nanofillers as given in Table 1. $SnO_2$ and ZnO are the semiconducting and highly crystalline materials. The $\varepsilon_r$ values of $SnO_2$ and ZnO are 34.5 and 10.26, respectively (Table 1). The ε′ values of $SnO_2$ dispersed PNC bilayer film are found much high as compared to that of the ZnO dispersed PNC bilayer film which are owing to a large difference in the $\varepsilon_r$ values of these NPs since the particle sizes of these are same (Table 1). In case of $Al_2O_3$ and $SiO_2$ dispersed PNC bilayer films, the ε′ values for PVA–PMMA–5 wt % $Al_2O_3$ film is found slightly low as compared to the PVA–PMMA–5 wt % $SiO_2$ film although $\varepsilon_r$ of $Al_2O_3$ ($\varepsilon_r = 9.7$) is higher than that of the $SiO_2$ ($\varepsilon_r = 3.8$). From these results, it seems that amorphous/crystalline structures and the difference in particle sizes of $Al_2O_3$ and $SiO_2$ mask the effect of $\varepsilon_r$ values of these nanoparticles in governing the frequency dependent ε′ values of these nanoparticles dispersed PNC bilayer films. Further, the $\varepsilon_r$ values of ZnO and $Al_2O_3$ nanoparticles are nearly equal (Table 1), but the observed high ε′ values of ZnO dispersed PNC film as compared to $Al_2O_3$ dispersed PNC film is due to the difference in their particle sizes and also the structural and semiconducting/insulating behaviour of these nanofillers. Although, the ε′ values of these PNC bilayer films vary with the type of dispersed NP but these values are found significantly lower than that of the ε′ values of the films of pristine PVA[4] (ε′ = 3.15 at 1 MHz and 27°C) and PMMA[45] (ε′ = 3.82 at 1 MHz and 27°C). The lowering of ε′ values of the PNC bilayer films may be due to the existence of air layer between the bilayered structures of these PNC films. Further, it seems that the dispersed NPs in polymer matrices disturb the dipolar ordering of the functional groups as in the case of other PNC materials[45].

The ε″ spectra of these phase separated PNC bilayer films (Fig. 1) vary with the type of nanoparticles but their order of variation with the change of nanoparticles is according to the order of the ε′ values. The ε″ values of $SnO_2$ dispersed PNC bilayer film are found relatively high. The ε″ values of $Al_2O_3$, $SiO_2$ and ZnO dispersed PNC bilayer films have less variation with the increase of frequency confirming their technological applications over broad frequency range. The ε″ values of $Al_2O_3$ and $SiO_2$ dispersed PNC bilayer films are around 0.025 or less confirming their suitability as polymer dielectric in fabrication of high quality capacitors usable at audio and radio frequencies. The tanδ spectra of these materials did not exhibit dielectric relaxation peaks which confirm that over the audio and lower radio frequencies, there is no existence of any dynamical process in such composite polymeric dielectric materials which is also interesting for their insulation and dielectric applications point of view over the experimental frequency range.

### Electric modulus spectra

The electric modulus $M^*(\omega)$ of a dielectric material is taken equal to the reciprocal of its complex permittivity $\varepsilon^*(\omega)$ due to which the contribution of unwanted electrode polarization (EP) effect nullifies

Table 2 — Values of complex dielectric permittivity $\varepsilon' - j\varepsilon''$ of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at different frequencies and 27°C

| Nanoparticle (NP) | $\varepsilon' - j\varepsilon''$ | | | | | |
|---|---|---|---|---|---|---|
| | 50 Hz | 100 Hz | 1 kHz | 10 kHz | 100 kHz | 1 MHz |
| $Al_2O_3$ | $1.23 - j0.020$ | $1.22 - j0.021$ | $1.19 - j0.017$ | $1.17 - j0.014$ | $1.15 - j0.015$ | $1.13 - j0.017$ |
| $SiO_2$ | $1.33 - j0.035$ | $1.30 - j0.030$ | $1.27 - j0.022$ | $1.25 - j0.017$ | $1.23 - j0.017$ | $1.20 - j0.019$ |
| ZnO | $1.76 - j0.046$ | $1.73 - j0.045$ | $1.67 - j0.037$ | $1.62 - j0.029$ | $1.58 - j0.032$ | $1.53 - j0.034$ |
| $SnO_2$ | $2.34 - j0.133$ | $2.28 - j0.121$ | $2.13 - j0.091$ | $2.02 - j0.069$ | $1.93 - j0.064$ | $1.83 - j0.068$ |

and the bulk properties of a material can be characterized[44]. The spectra of real *M′* and imaginary *M″* parts of electric modulus for PVA–PMMA–5 wt % NP bilayer films, at 27°C, are depicted in Fig. 2. It is found that the *M′* values gradually increase with the increase of frequency over the entire frequency range, which reveals that there is no contribution of EP effect in the observed ε′ spectra of these materials, because the *M′* and *M″* values remain close to zero and are also frequency independent over the EP dominated frequency region of a composite dielectric material[51,52]. Mostly, in charge conducting dielectric material, the *M″* exhibits relaxation peak in high frequency region corresponding to the conductivity relaxation process[53,54], but for these phase separated PNC bilayer films, such process has not been observed which is due to poor electrical conductivity of these materials as discussed in the next section.

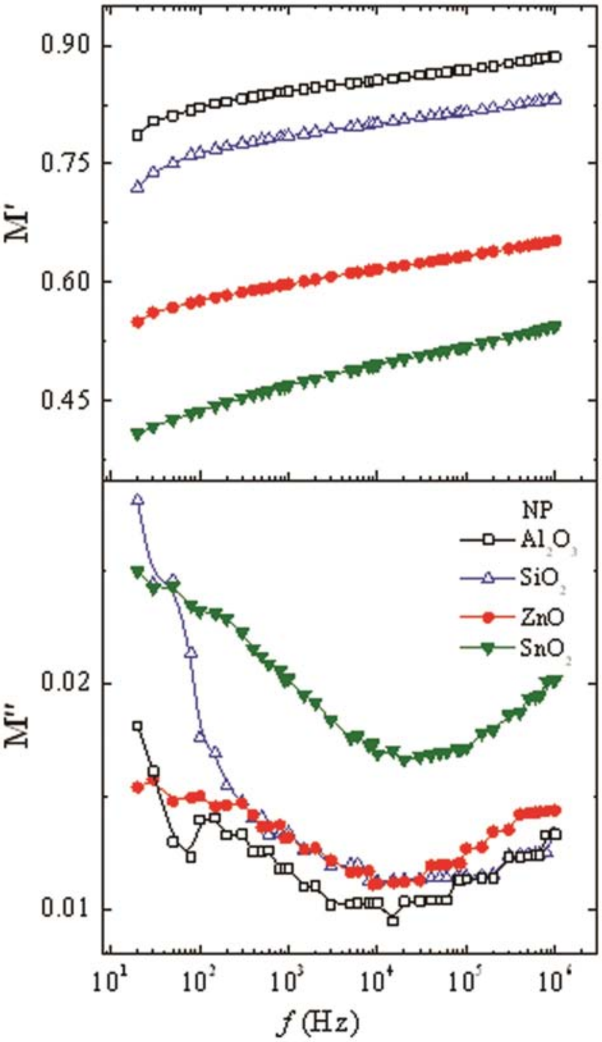

Fig. 2 — Complex electric modulus (real part *M′* and loss part *M″*) spectra of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at 27°C

### AC conductivity spectra

The frequency dependent values of real σ′ and imaginary σ″ parts of complex AC electrical conductivity σ*(ω) of the PVA–PMMA–5 wt % NP bilayer films, at 27°C, are shown in Fig. 3. On log-log scale, σ′ and σ″ values increase linearly with the increase of frequency having unity value of their slope confirming Ohmic conduction behaviour. The σ″ values of the materials are found more than one order of magnitude higher than that of the corresponding σ′ values. The σ′ and σ″ values at selective frequencies for these materials are given in Table 3 for their comparison in order to see the effect of various nanoparticles on the conductivity behaviour. The σ′ values of these materials at 20 Hz are of the order of $10^{-13}$ S/cm which increases up to $10^{-8}$ S/cm at 1 MHz. The direct current (DC) electrical conductivity $\sigma_{dc}$ values of these materials are determined from the intercept of linear fit of the σ′

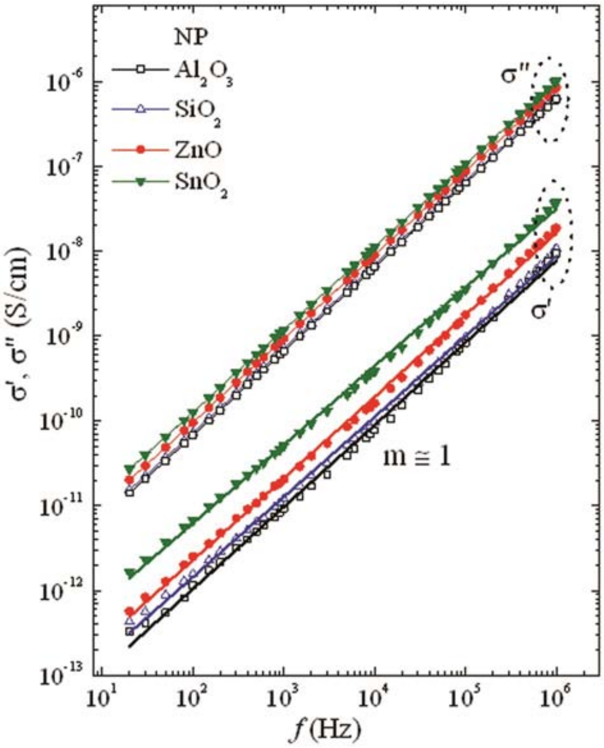

Fig. 3 — Complex AC electrical conductivity (real part σ′ and loss part σ″) spectra of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at 27°C. Solid lines are linear fit of experimental data

Table 3 — Values of dc ionic conductivity $\sigma_{dc}$, complex AC electrical conductivity $\sigma' + j\sigma''$ and complex impedance $Z' - jZ''$ of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at different frequencies and 27°C

| Nanoparticle (NP) | $\sigma_{dc}$ ($10^{-14}$ S/cm) | 100 Hz $\sigma' + j\sigma''$ ($10^{-12}$ S/cm) | 10 kHz $\sigma' + j\sigma''$ ($10^{-10}$ S/cm) | 1 MHz $\sigma' + j\sigma''$ ($10^{-8}$ S/cm) |
|---|---|---|---|---|
| $Al_2O_3$ | 1.21 | $1.15 + j67.7$ | $0.78 + j65.0$ | $0.94 + j62.8$ |
| $SiO_2$ | 1.77 | $1.56 + j72.8$ | $0.97 + j69.5$ | $1.07 + j66.9$ |
| ZnO | 2.72 | $2.51 + j96.4$ | $1.62 + j90.2$ | $1.87 + j85.2$ |
| $SnO_2$ | 8.73 | $6.72 + j126.9$ | $3.81 + j112.0$ | $3.78 + j102.0$ |
|  | 1 kHz $Z' - jZ''$ ($10^4$ Ω) | 10 kHz $Z' - jZ''$ ($10^3$ Ω) | 100 kHz $Z' - jZ''$ ($10^2$ Ω) | 1 MHz $Z' - jZ''$ ($10^1$ Ω) |
| $Al_2O_3$ | $2.89 - j234$ | $2.65 - j230$ | $2.92 - j234$ | $3.49 - j238$ |
| $SiO_2$ | $5.47 - j343$ | $4.69 - j330$ | $4.72 - j336$ | $5.32 - j342$ |
| ZnO | $4.17 - j188$ | $3.41 - j176$ | $3.58 - j181$ | $4.18 - j186$ |
| $SnO_2$ | $5.09 - j119$ | $4.30 - j126$ | $4.33 - j131$ | $5.09 - j138$ |

plots and the observed $\sigma_{dc}$ are given in Table 3. These $\sigma_{dc}$ values are found of the order of $10^{-14}$ S/cm for the investigated PNC bilayer films and these vary by one order of magnitude with various nanofillers. It is found that $\sigma'$ values for the various NP dispersed PNC bilayer films vary in the order $SnO_2$ > ZnO > $SiO_2$ > $Al_2O_3$, and in comparison to $Al_2O_3$ dispersed PNC film the $\sigma'$ values of $SnO_2$ dispersed PNC bilayer film are found nearly one order of magnitude high. Comparatively low conductivity values and their linear variation with frequency confirm the suitability of these materials as polymeric dielectric for DC voltage operated microelectronic devices especially for gate insulator in OTFTs and in the fabrication of OFETs.

### Impedance spectra

The resistive part $Z'$ and capacitive reactance part $-Z''$ of the complex impedance of the PVA–PMMA–5 wt % NP films, at 27°C, are shown in Fig. 4. On log-log scale, it is found that the magnitude of both $Z'$ and $Z''$ decrease linearly with the increase of frequency confirming their Ohmic behaviour because the slope is close to unity. The values of $Z'$ and $Z''$ at some selective frequencies are given in Table 3, which shows that these values also vary with the types of NPs dispersed in the PNC bilayer films. Further, it is found that the values of $Z''$ are about two orders of magnitude high as compared to the $Z'$ values at the respective frequencies. The high magnitude of $Z''$ suggests the suitability of these materials as dielectric in the fabrication of flexible-type capacitors workable at audio and radio frequencies.

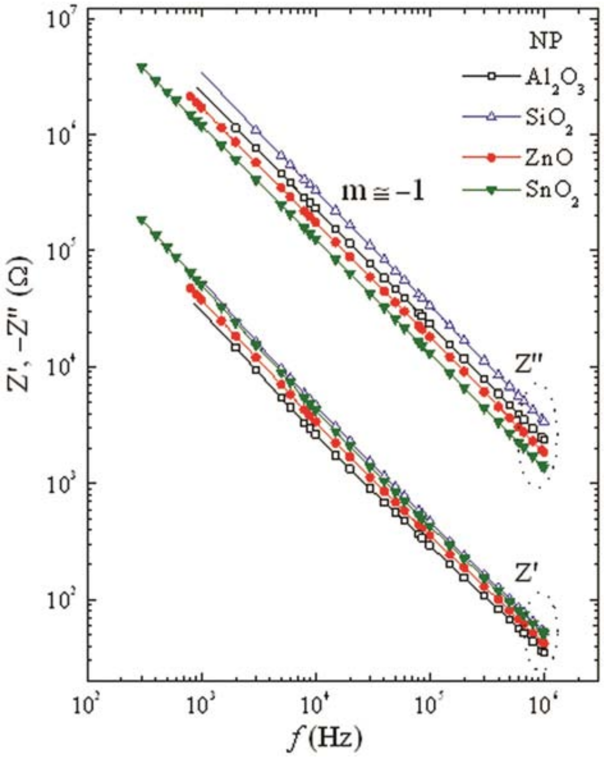

Fig. 4 — Complex impedance (real part $Z'$ and loss part $Z''$) spectra of PVA–PMMA–5 wt % NP (NP; $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$) phase separated PNC bilayer films at 27°C. Solid lines are linear fit of experimental data

## Conclusion

The complex dielectric permittivity, electric modulus, AC electrical conductivity and the impedance properties of the PVA–PMMA–5 wt % NP films, where NPs are $Al_2O_3$, $SiO_2$, ZnO and $SnO_2$, were reported over the audio frequency range of 20 Hz–20 kHz and also at lower radio frequency range of

20 kHz–1 MHz, at 27°C. The dielectric permittivity values of these PNC bilayer films are found low and relatively stable over the experimental frequency range confirming their suitability as low-permittivity high performance polymeric nanodielectrics. The very low electrical conductivity and high impedance values of these materials reveal their use as electrical insulator and polymeric dielectric substrate in design and fabrication of advanced microelectronic devices operated at low voltage.

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