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# Impedance-spectroscopy analysis of oriented and mercerized bamboo fiber-reinforced epoxy composite

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**Abstract** Bamboo fiber-reinforced epoxy composites were fabricated with untreated and alkali treated bamboo fibers. Dielectric, electric modulus, ac, and dc conductivity studies were carried out to rationalize the dielectric behavior of bamboo/epoxy composites. Composites of two fiber orientation parallel and perpendicular to the electric field were prepared. The dielectric behavior and electric modulus spectra of the composites were characterized using standard impedance analyzer. Dielectric properties were analyzed as a function of frequency (95 Hz–2 MHz) for temperatures in the range from 30 to 180 °C. Real part of dielectric constant ($\varepsilon'$), conductivity, and dielectric dissipation factor (tan $\delta$) of 0° oriented bamboo/epoxy composites were higher than that of 90° oriented composites. Conductivity activation energy, tan $\delta$, $\varepsilon'$, and volume resistivity decreased with increase in frequency at all the temperatures under study. Mercerization reduces the water absorption in bamboo fibers and thus improves corresponding dielectric properties of composites. Relaxation times 39.80 μs and 258.5 μs for 0° and 90° oriented bamboo/epoxy composites were calculated respectively from the relaxation peaks observed in electric modulus spectra at 180 °C.

## Introduction

The benefits of natural fiber-reinforced plastic (NFRP) composites, as compared to wood, plastic, and synthetic fiber composites have been well recognized due to a number of properties such as raw material utilization, biodegradability [1] flexibility, modulus, impact resistance [2], low dielectric constant [3], and low coefficient of thermal expansion [4]. Besides, they have other desirable properties include light weight, low cost, less machine wear, less equipment abrasion, renewable character, availability in variety of forms throughout the world, high sound attenuation, desirable fiber aspect ratio, less skin, and respiratory irritation [5–11].

The incorporation of natural fiber into thermoplastics afford scientists an opportunity to synthesize NFRP composites that potentially rival the most advanced materials in nature which have good mechanical properties, and at the same time can be used for dielectric applications such as industrial and house hold plugs, switches, connectors, printed circuit boards (PCBs), solder mask, cable pillar, and cables, etc. [12]. In addition, they have some potential applications in the making of packaging trays, solder mask, seat back, paper making sheets, and arm rest [13, 14]. Plastics act as an effective insulator but their use is limited to non-load bearing dielectric applications. Therefore, studies on the dielectric properties of NFRP composites assume greater importance [15–18]. Dielectric applications require low dielectric constant and coefficient of thermal expansion (CTE) for successful implementation. For, e.g., $\varepsilon'$ for FR-4 PCB base material is nearly 4.6 at 1 MHz frequency and CTE is as low as 58 ppm/K [19].

The main drawbacks of natural fibers are their hydrophilicity, low weathering resistance that adversely affect the fiber/matrix interface and lead to degradation, loss of strength, and delamination of composites [20, 21]. Being hydrophilic, natural fibers need to be treated first to make them more compatible with hydrophobic thermosets and thermoplastics [21–23]. In addition, bio-composites present a more advantageous end-of-life scenario, as assessed from

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Life Cycle Analysis (LCA) studies, especially if little chemical treatment is required to have sufficient material properties for the envisaged applications. Several researchers have reported improvement in dielectric properties of cellulose fibers when alkalized at different NaOH concentrations [24, 25]. Geethamma and co-workers [26] used 5% NaOH to remove surface impurities on oil palm fibers and short coir fibers.

Based on its anatomical properties, ultra structure and plant fracture mechanism bamboo establishes itself as a tough engineering material and superior natural fiber as compared to other known natural fibers like industrial hemp, flax, kenaf, coir, sisal, and henequen, etc. [27, 28]. Bamboo and many other natural plant fibers have also been used in the European automotive industry. In composites it is usually desirable to orient the fibers for enhanced mechanical and dielectric properties. It is normally very difficult, if not impossible, to achieve perfect alignment of fibers. The orientation distribution of fibers in a composite is dominated by the processing conditions. The effect of frequency, temperature, and fiber orientation on dielectric properties of sisal fiber-reinforced plastic composite has been investigated [12, 29] but there is no such study on bamboo fiber-reinforced epoxy (BFRE) composite. The objective of our study is to see the effect of fiber alignment and alkali treatment on the dielectric properties of bamboo fiber composites using epoxy resin as a matrix and to examine the performance of a standard laminating resin. In this paper the volume resistivity, dielectric constant, and tan $\delta$ of 0° and 90° oriented BFRE composites at different temperatures and frequencies are determined; values have been explained on the basis of structural changes.

## Experimental methods

### Materials

Orthogonal bamboo strip mats in 60 × 60 $cm^2$ size were procured from Tripura complex New Delhi. The areal weight of the mat is 424 $g/m^2$. The epoxy resin (CY-230) and hardener (HY-951) were purchased from CIBA-GEIGY. Sodium hydroxide pellets of 98% strength were supplied as general laboratory reagents. Material specifications are described by the authors in a previous article [30].

### Alkali treatment

Bamboo mats were soaked in 5% NaOH [31] solution in a water bath at room temperature for 1 h. The treated mats were thoroughly rinsed in water and then neutralized in 2% HCl solution. The mats were then left to dry at room temperature before being put in an oven for 24 h at 110 °C. A litmus paper test was carried out to check the neutrality [30]. Untreated mats were also dried in an oven.

### Composite manufacturing method

Woven bamboo strip mats were cut to size 300 × 200 $mm^2$. The mats were cleaned with detergent powder and water solution and washed thoroughly to remove dust and other deposits from the surface. They were left in the open to dry out for 4 h. 30 mm thick clear Perspex sheets of size 300 × 250 $mm^2$ were used as mold to prepare the composite. Epoxy resin mixed with the hardener (10% by weight) was poured onto each layer of fiber in a zigzag configuration to ensure even delivery of the resin and the procedure repeated for six layers of treated mats, piled one over the other. These were then placed in between two Perspex sheets and pressed in the hydraulic press under a load of 180 kN for 24 h. The composites were post-cured at 80 °C overnight in an oven. The volume fraction of bamboo fiber mats was 68 ± 1% in the BFRE composites. Thickness of sample was 5 mm.

### Electrical testing

Test samples of size 12 × 10 $mm^2$ of two fiber orientations, parallel (0° oriented) and perpendicular (90° oriented) to the electric field were prepared to carry out the dielectric measurements. The surfaces of the samples were coated with silver paste on two faces and cured at 50 °C. Copper wires were fixed as electrodes on each face of the specimen. The capacitance, tan $\delta$, and resistance of the samples were measured as a function of frequency (95 Hz–2 MHz) using an impedance analyzer (HP4192A) for temperatures in the range from 30 to 180 °C.

## Results and discussion

### Untreated bamboo epoxy composite

#### *Dielectric constant and dissipation factor*

Dielectric properties of NFRP composites at different frequencies and temperatures depend very strongly on the contribution of interfacial, ionic, and orientation polarization [32–34]. Figures 1a, c and 2a show the variation of $\varepsilon'$ with temperature of 0° and 90° oriented BFRE composites. Figures 1b, d and 2b show the variation of tan $\delta$ with temperature of 0° and 90° oriented BFRE composites at different frequencies. At lower and intermediate frequencies $\varepsilon'$ and tan $\delta$ values in BFRE composites are due to orientation, space charge and interfacial polarization contributions. Contribution of orientation polarization

decreases at high frequency because molecules do not have time for orientation which is evidenced by the decrease in $\varepsilon'$ and tan $\delta$ of BFRE composites with frequency. Room temperature $\varepsilon'$ of 90° oriented BFRE composites ranges from 5.53 to 4.4 (95 Hz–2 MHz) and goes as high as 10.18 at 80 °C (95 Hz). Increase of temperature up to ~80 °C increases $\varepsilon'$ and tan $\delta$ for both 0° and 90° oriented BFRE composites. Rise in temperature from 80 to 140 °C decreases $\varepsilon'$ as shown in Fig. 1a and c. Such behavior implies the removal of moisture from the surface of bamboo fiber and occurrence of glass-rubbery transition process at ~80 °C [35]. The dielectric behavior of NFRP composites is affected to the relaxation processes [36].

The values of $\varepsilon'$ remain almost constant in the temperature range 140–160 °C. This is in agreement with the results obtained earlier for some polymers [37]. Beyond this temperature, degradation of the fibrous molecules starts. Due to voids in the composite, intrinsic defects occur. These defects contribute to the interfacial polarization between the samples and gases evolved out of the structure during decomposition stage. As a result, increase in $\varepsilon'$ is attributed in the composites during decomposition stage. The decrease in the values of $\varepsilon'$ with increasing frequency at higher temperature occurs due to influence of interfacial polarization. 90° oriented BFRE composites have low dielectric constant as compared to 0° oriented composites because series combination of dielectric constant has lower value than that of parallel combination [35].

*Volume resistivity*

The volume resistivity of natural fiber composites is the function of cell (shape, size, and number), microfibril angle of cellulose, crystalline, and amorphous components present, chemical composition, impurity content, moisture, and the voltage applied. Plots of volume resistivity as a function of temperature of 0° and 90° oriented BFRE composites are given in Fig. 3a and b, respectively. It has been found that volume resistivity decreases with increase in temperature up to 80 °C and after this remains almost constant in the temperature range from 80–140 °C. Beyond this temperature, thermal degradation of composite starts and volume resistivity shows anomalous behavior. As the frequency increases, the resistivity shows a decrease due to the interfacial polarization arising because of the heterogeneity of the system. The volume resistivity of 0° oriented BFRE composites is smaller than that of 90° oriented composites. The volume resistivity of 90° oriented BFRE composites is higher than that of 0° oriented composites. It is due to series and parallel combination of bamboo fibers resistance with epoxy matrix resistance in 90° and 0° oriented BFRE composites, respectively. Jain et al. [27, 28] reported that bamboo

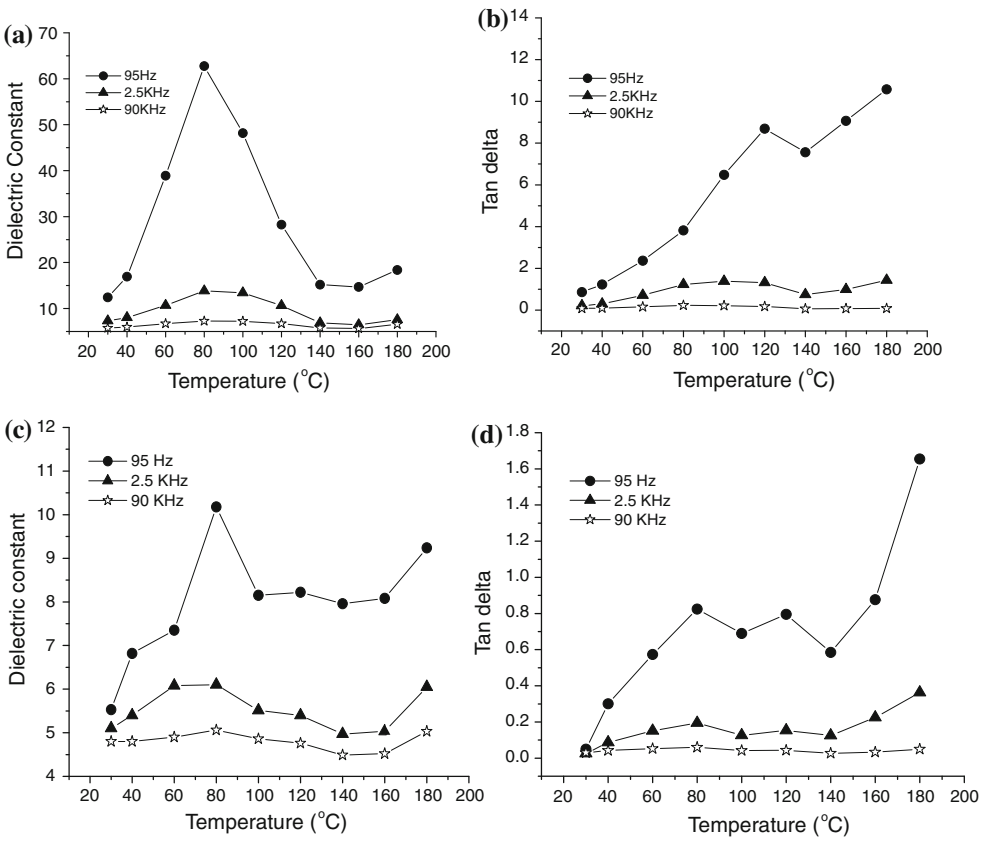


**Fig. 1** Temperature dependence of real part of dielectric constant and tan $\delta$ at different frequencies **a**, **b** 0° and **c**, **d** 90° oriented bamboo epoxy composites

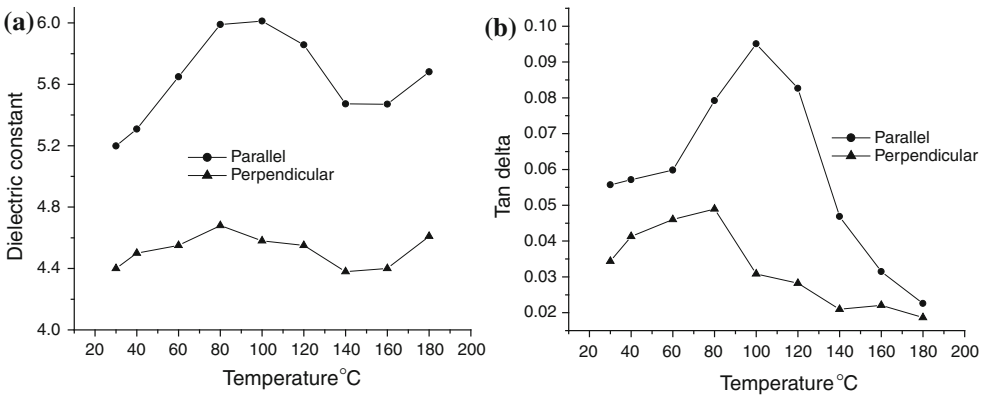


**Fig. 2** Orientation dependence of **a** real part of dielectric constant and **b** tan $\delta$ at different temperature for 2 MHz frequency

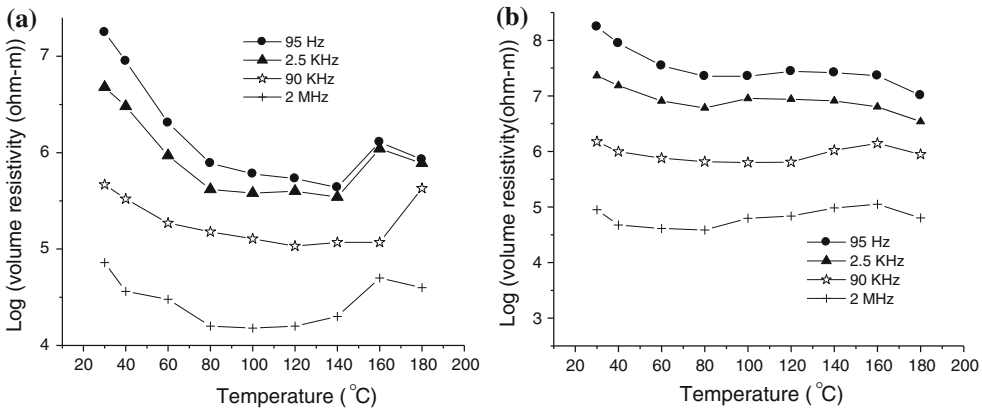


**Fig. 3** Variation of volume resistivity with temperature at different frequencies **a** 0° oriented, **b** 90° oriented Bamboo epoxy composites

has higher percentage of lignin with considerably small microfibrillar angle. These facts about bamboo support its high tensile strength as compared to other natural fibers sisal, jute, coir, banana, etc. Cellulose content in bamboo fiber is small as compared to other natural fibers [38]. According to relation cellulose content (%) = 73.78–3.09 $\rho$ [39], where $\rho$ is the volume resistivity, bamboo fiber should have higher volume resistivity among all other natural fibers.

*Conductivity*

The study deals with the ac and dc conductivity of BFRE composites. Temperature dependence of ac conductivity for 0° and 90° oriented BFRE composites is shown in Fig. 4a and b, respectively, at different frequencies. It was found that ac conductivity increases with frequency at all the temperatures. Orientation of fibers in composite changes the structure of the total composite, due to which ac conductivity changes with the orientation angle. The ac conductivity values at room temperature for 0° and 90° oriented BFRE composites are found to be of the order of $10^{-7}$ and $10^{-8}$ (mho/m), respectively. At low temperature region, the ac conductivity depends significantly on the frequency. However, with the increase in temperature dielectric relaxation takes place and the dependency of the conductivity on frequency gets reduced. Figure 5a shows the variation of conductivity ($\sigma(\omega)$) with frequency for 90° oriented BFRE composites. The frequency behavior of conductivity at given temperature in BFRE composites may be divided into two parts, i.e., dc and ac conductivity and expressed as [40]

$$\sigma(\omega) = \sigma_{dc} + A\omega^n$$

where $\sigma_{dc}$ and $A\omega^n$ represents the frequency independent and dependent conductivity. At low frequencies, samples show a very distinct plateau which represents the dc conductivity of the composites at different temperatures. At higher frequencies ($\geq 10^3$ Hz), the spectra exhibit a strong dispersion due to the restricted motion of the OH ions and other impurities in the disordered structures represents the ac conductivity. An Arrhenius plot of the logarithm of conductivity versus the reciprocal of the absolute temperature is shown in Fig. 5b. The dc and ac conductivity activation energy values calculated using Arrhenius plots are 0.33 and 0.30–0.21 eV for 90° oriented composites. The activation energy values of dc conductivity are more when compared with ac conductivity. This is due to the fact that at low frequencies, the conductivity is due to the mobility/transportation of charge carriers over long distances. However, at high frequencies they take relaxation/orientation mechanism,

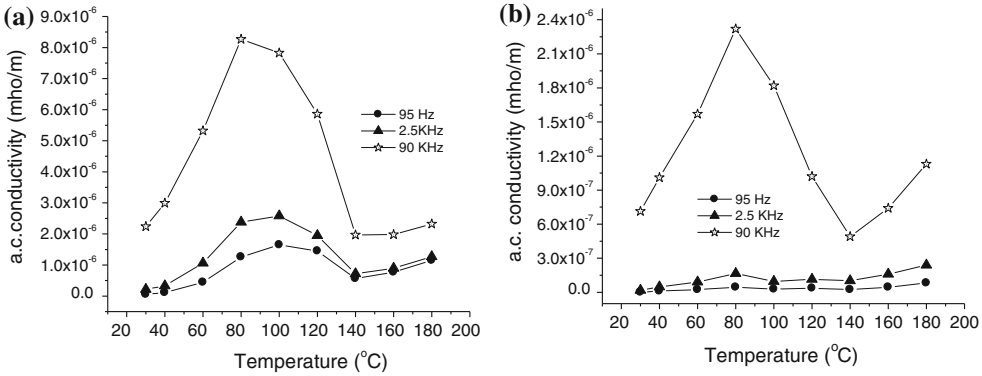


**Fig. 4** Temperature dependence of ac conductivity at different frequencies **a** for 0° and **b** 90° oriented bamboo epoxy composites

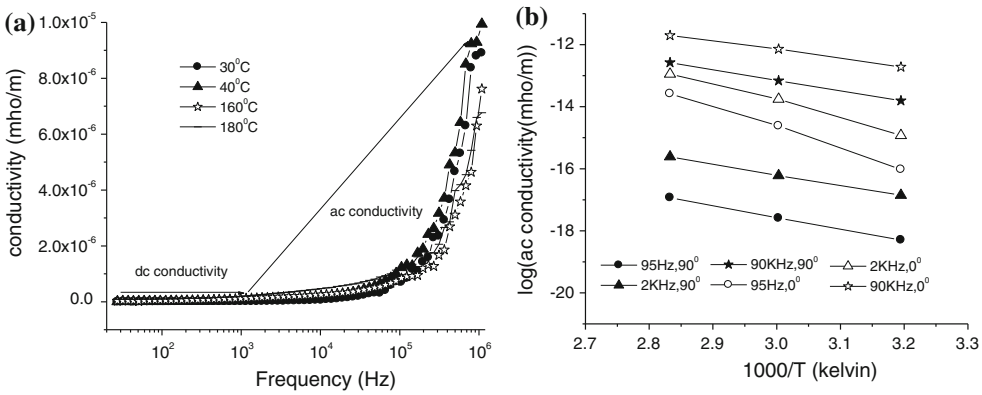


**Fig. 5** **a** ac and dc conductivity at different temperature for 90° oriented bamboo epoxy composites. **b** Reverse temperature dependence of ac conductivity for bamboo epoxy composites at chosen frequencies

in which charge mobility is restricted to only the nearest neighboring lattice sites. Conductivity activation energy values of BFRE composites are given in Table 1. The activation energy is seen to decrease with increase in frequency from 0.33 to 0.21 eV for 90° oriented BFRE composites in the frequency range "95–90 kHz". The energy required for the relaxation/orientation process is lower than the mobility of charge carriers over long distances, hence greater conduction activation energies are observed at lower frequencies than at higher frequencies.

## Electric modulus

The electric modulus analysis has been widely used to study dielectric behavior of materials. In general, the dielectric properties of composite materials arise due to fiber and matrix effects. The motion of charges could take place by charge displacement, dipole reorientation, space charge formation, etc. In order to understand the electric properties of a given sample fiber and matrix combinations must be separated out. In 0° oriented composites, fiber and matrix come in parallel and in 90° oriented composites, fiber and matrix come in series. The electric modulus is analog to the mechanical shear modulus as explained by Mccrum et al. [41]. The electric modulus is defined as [42]

$$M^*(\omega) = \frac{1}{\varepsilon^*(\omega)} = M' + JM''$$

where $\varepsilon^*$, $M'$ and $M''$ are dielectric constant, the real, and imaginary part, respectively, of the electric modulus. An advantage of using the electric modulus to interpret bulk relaxation properties is that variation in the large values of the real part of dielectric constant and loss factor at low frequencies are minimized. The electric modulus represents the real dielectric relaxation process [43]. The imaginary part of the electric modulus is plotted as a function of frequency. Broad electric modulus peaks occurring as a function of frequency of 0° and 90° oriented BFRE composites are shown in Fig. 6. The frequency region where the peaks occurs follow the condition $\omega\tau = 1$ where $\tau$ is the relaxation time and $\omega$ is the angular frequency at peaks. Relaxation times 39.80 μs and 258.5 μs for 0° and 90° oriented bamboo/epoxy composites were calculated respectively from the relaxation peaks observed in electric modulus spectra at 180 °C.

**Table 1** Conductivity activation energy of BFRE composites at different frequency

| Frequency (Hz) | Activation energy (eV) | |
|---|---|---|
| | 90° oriented BFRE | 0° oriented BFRE |
| 95 Hz | 0.33 | 0.58 |
| 2 kHz | 0.30 | 0.47 |
| 90 kHz | 0.21 | 0.25 |

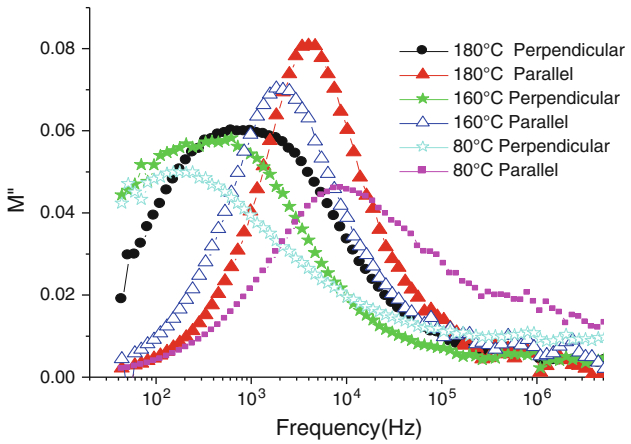


**Fig. 6** Dielectric relaxation peaks in bamboo epoxy composites for parallel and perpendicular orientation at different temperature

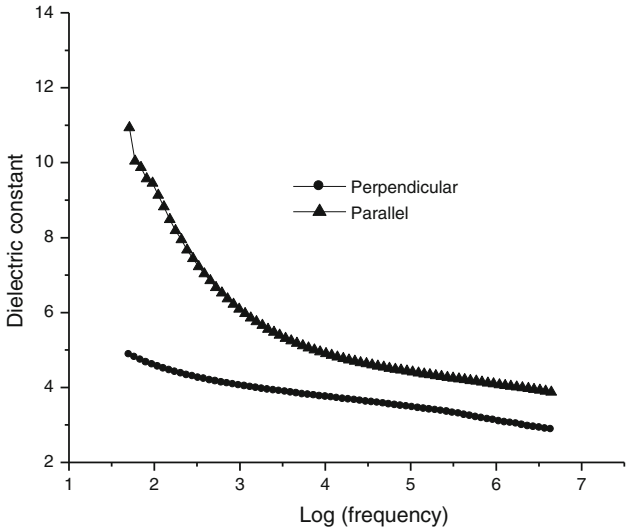


**Fig. 7** Orientation dependence of real part of dielectric constant for alkali treated bamboo epoxy composites

## Mercerization

The epoxy resin has been reinforced with alkali treated bamboo fiber mattings so as to develop a BFRE composite with a view to improve dielectric properties. The value of $\varepsilon'$ is 3.12 at 1 MHz for alkali treated 90° oriented BFRE composite as shown in Fig. 7. This property makes BFRE composites a promising material to use as a dielectric for PCBs [19]. Volume resistivity values of alkali treated composites remained higher than that of the untreated composites. Figure 8a depicts the dependence of volume resistivity with logarithm of frequency for untreated and alkali treated BFRE composites. The hydrophilicity of untreated cellulosic bamboo fiber is responsible for the greater dielectric constant and ac conductivity of the composite as compared to the treated one as shown in Fig. 8b. Earlier studies carried out by authors have shown that alkali treatment resulted in the reduction of water absorption from 41 to 26% [30]. In fact, conductivity of the cellulose fiber is mainly due to the presence of water molecules absorbed by the fiber surface [12]. The entire experimental procedure was done under the same external conditions as room temperature was recorded 29 ± 1 °C. With mercerization, the interfibrillar region loses hemicellulose and lignin, becoming less dense and less rigid, thereby making the fibrils more capable of rearranging themselves and undergoing reorientation and recrystallization. Removal of lignin and hemicellulose leads to fibrillation, i.e., breaking down of the composite fiber bundle into smaller fibers. This increases the effective surface area available for contact with the matrix. Volume resistivity of insulative material is greater than $1 \times 10^{11}$ (Ω m) and ranges from $1 \times 10^{7}$ to $1 \times 10^{11}$ (Ω m) for antistatic materials. Volume resistivity of untreated BFRE composite is of the order of $10^8$ (Ω m). After alkali treatment the volume resistivity enhanced by the factor 10 and is of the order of $10^9$(Ω m) as shown in Fig. 8a. Investigations are in progress to further improve the

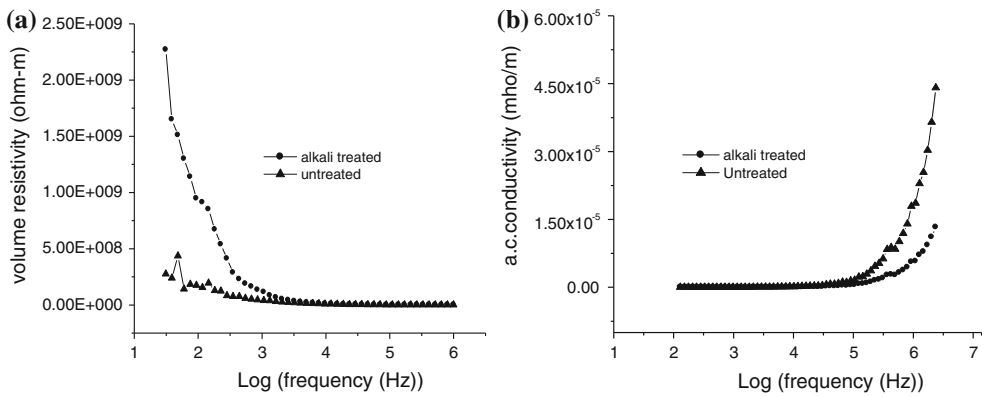


**Fig. 8** Effect of alkali treatment on **a** volume resistivity, **b** ac conductivity of bamboo/epoxy composites

volume resistivity of bamboo composites so that it can be use as an insulator.

## Conclusions

Impedance spectroscopy has been applied to rationalize the dielectric properties of BFRE composites. 90° oriented BFRE composites have better dielectric properties as compared to 0° oriented composites. The dielectric constant and dissipation factor of BFRE composites were seen to decrease with frequency. This was attributed to the decrease in orientation polarization of the polar groups present in lignocellulosic fibers. The dielectric properties were improved after mercerization due to decrease of orientation polarization. The volume resistivity was seen to decrease with frequency and increase with mercerization. Observed relaxation peaks in electric modulus spectra reveal the relaxation behavior of BFRE composites. As compared to other natural fibers bamboo fiber shows high volume resistivity, good mechanical properties because of its comparatively low cellulose content and small microfibrillar angle. An analysis of dielectric properties shows that these cost effective biocomposites could be use for dielectric applications.

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