Electrical Properties of Banana Fiber-Reinforced Phenol Formaldehyde Composites

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ABSTRACT: The electrical properties of banana fiber-reinforced phenol formaldehyde composites have been studied with special reference to fiber loading, fiber treatments, and hybridization with glass fibers. The dielectric constant decreased with frequency and fiber loading. Treatments with silanes, NaOH and acetylation, latex treatment, heat treatment, and cyanoethylation decreased the dielectric constant value. The dielectric constant is higher for banana/PF composites than that of glass/PF composites. In hybrid composites, the dielectric constant decreased with increase in glass fiber concentration. The volume resistivity of banana fiber-reinforced phenol formaldehyde composites decreased with frequency and fiber loading. Chemical modifications in the fiber increased the volume resistivity of the composites. Volume resistivity of banana/glass hybrid composites increased with increase in glass fiber content. Layered composites with glass fiber at periphery were found to have higher volume resistivity than other layering patterns. The loss factor decreased by the chemical treatments and in hybrid composites it decreased with increase in volume fraction of glass fibers. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 256–263, 2008

Key words: natural fiber composites; hybrid composites; electrical properties

INTRODUCTION

The most desirable combination of characteristics such as ease of fabrication, low cost, light weight, and excellent insulation properties have made plastics one of the most desirable materials for electrical applications. The use of plastics in electrical applications was limited to nonload bearing general-purpose applications. Fiber-reinforced plastic materials not only act as effective insulators, but also provides mechanical support for field carrying conductors. Phenol formaldehyde resin is well known for its electrical insulator characteristics. But it is very brittle and has poor mechanical properties. Incorporation of fibrous reinforcements in polymer matrices leads to high performance composite materials having very good mechanical properties and at the same time suitable for electrical applications. They can be used as terminals, connectors, industrial and house hold plugs, switches, and printed circuit boards.

In recent years, natural fiber-reinforced polymer composites have attracted more and more research interests owing to their potential as an alternative for synthetic fiber composites such as glass or carbon fiber composites.[1–5] Compared with synthetic fiber composites the natural fiber composites are having the advantages such as easy availability, renewability of raw materials, low cost, light weight and high specific strength, and stiffness. The drawbacks of natural fibers like moisture absorption, low aging and weathering resistance and their hydrophilicity that weakens the fiber/matrix interface in hydrophobic polymer matrices could be overcome by different surface modifications. Several researchers have used mercerization, treatments with silanes, isocyanates, acetylation, and grafting for the development of high performance natural fiber composites.[6–14] Development of hybrid composites using natural fibers and synthetics such as glass fiber is also reported to be an effective way for improving the performance of natural fiber composites.[15–18]

The applications of the composite materials in electrical applications make the study of the electrical properties of composites such as dielectric constant, volume resistivity, and loss factor important. Electrical properties of polybithiophene-polystyrene composites were analyzed by Morsli et al.[19] Kortschot and Woodhams[20] reported that for maximum efficiency of conductive composites very high aspect ratio flakes are recommended and these particles will be most effective when they have random orientation in the composite.

Wu and Tung[21] did dielectric studies of mineral filled epoxy composites containing different dielectric fillers. The results indicated that the dielectric con-

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stants increase with the addition of the dielectric filler and with an increase of dielectric constant of the filler.

Dutta et al.[22] studied on the mechanical and electrical anisotropy of pineapple fibers. They got a sharp increase of dielectric constant and fall of loss factor along fiber direction compared with that of the transverse direction because of increase in crystallinity along the fiber direction. Electrical properties of pineapple fiber-reinforced polyethylene composites were done by Jayamol et al.[23] They got an increase in the dielectric constant of composite with fiber loading due to increased orientation and interfacial polarization. Effect of sisal fiber orientation on electrical properties of sisal fiber reinforced epoxy composites has been studied by Chand and Jain. They found that the composites show electrical anisotropic behavior.[24]

The electrical properties of natural fibers were studied by Kulkarni et al.[25] Paul and Thomas[26] compared the electrical properties of sisal fiber-reinforced low-density polyethylene composites with that of carbon black and glass fiber filled low-density polyethylene (LDPE) composites. At low frequencies, the dielectric constant varied in the order: neat LDPE < glass-LDPE < sisal-LDPE < coir-LDPE < carbon black-LDPE. Paul et al.[27] also analyzed the effect of surface treatments on the electrical properties of low-density polyethylene composites reinforced with short sisal fibers. They found that treatments such as alkali, CTIDIC (urethane derivative of cardanol), stearic acid, peroxide permanganate, and acetylation decreased the dielectric strength of the composites as these treatments decreased the hydrophilicity of the composites. Fraga et al.[28] analyzed the relationship between water absorption and dielectric behavior on polyester matrix composites of glass and jute fibers. They observed that dielectric constant of jute fiber composite is higher than that of glass fibers because of higher water uptake of jute fibers than glass fibers.

Recently in our laboratory we have developed banana fiber-reinforced phenol formaldehyde composite. The properties of these composites were comparable with that of glass fiber composites.[1] Fiber was given different surface treatments and the mechanical and dynamic mechanical properties of these treated fiber composites were analyzed.[29,30] Hybrid composites were prepared using banana fiber and glass fiber and they had very good mechanical and dynamic mechanical properties indicating a positive hybrid effect.[31] In the present work, the electrical properties of banana fiber-reinforced phenol formaldehyde composites and glass fiber reinforced composites are analyzed. The effect of chemical treatments with alkali, silanes, acetylation, latex treatment, heat treatment, and cyanoethylation on the dielectric constants, volume resistively, and loss factor are studied. The electrical properties of banana/glass hybrid fiber-reinforced composites with varying hybrid ratios and layering patterns are also analyzed.

EXPERIMENTAL

Materials

Phenol formaldehyde resole type resin obtained from M/S West Coast Polymers Pvt. Ltd., Kannur, Kerala, India was used as matrix. Banana fiber obtained from Sheeba Fibers and Handicrafts, Poovancode, Thamilnadu, India was used in this study. The chemical constituents of lignocellulosic banana fiber are cellulose (63–64%), hemicellulose (19%), and lignin (5%) with moisture content of (10–11%).[32] The glass fiber used was E-glass roving obtained from Hitech Fiber Corporation, Bangalore.

Fiber modifications

NaOH treatment

Fibers were immersed in 4% NaOH solution at 27°C for 2 h, washed many times with distilled water and finally washed with water containing little acid and dried.

Silane treatments

Banana fiber pretreated with NaOH solution for 2 h were dipped in alcohol water mixture (60:40) containing 0.6% silane as coupling agents. The pH of the solution was maintained between 3.5 and 4. The fibers were washed in distilled water and dried. The silane coupling agents used was vinyl triethoxy silane and 3-aminopropyl triethoxy silane.

Heat treatment

Banana fiber was heated at 150°C in an air-circulating oven for 4 h. The fiber was then cooled to room temperature in a dessicator.

Cyanoethylation

Banana fiber was immersed in a 4% solution of NaOH saturated with sodium thiocyanate for 30 min. Subsequently, the fiber was pressed to drain of the excess water and removed quickly in to round bottomed flask allowing a reaction with acrylonitril (banana fiber to AN ratio 1 : 3) for 1 h at 40–45°C. After the reaction, a solution of acetic acid was used to neutralize the alkali catalyst. The product was well washed with water and alcohol, respectively.

Acetylation

The NaOH pretreated fiber was neutralized, washed with water, and air-dried. The fiber was soaked in a

50% solution of acetic acid for 5 min, washed, and air-dried.

Latex treatment

The NaOH pretreated fibers were given a latex coating by dipping it in to natural rubber latex having 10% dry rubber content.

Preparation of composites

Prepreg route was followed for the preparation of composites. Hand lay-up method followed by compression molding was adopted for composite fabrication. Randomly oriented fiber mats of uniform thickness were prepared from chopped banana fibers (30 mm) and glass fibers (40 mm). The fiber mats were dried and impregnated in PF resin and the prepreg was kept at room temperature up to semicured stage. It was then pressed at 100°C in a mould measuring $150 \times 150 \times 3$ mm^3 to get a three dimensionally crosslinked network. Composites were prepared varying the banana fiber loading (volume %) from 0 to 48% (0, 18, 28, 32, and 48%). Composites of 30% fiber loading were prepared using the treated fibers. Hybrid composites were prepared varying the relative volume fractions of fibers such as 0 : 1, 0.09 : 0.91, 0.12 : 0.88, 0.24 : 0.76, 0.32 : 0.68, 0.50 : 0.50, 0.30 : 0.70, and 1 : 0 (glass : banana). Samples with different layering patterns were also made as given in Figure 1.

Electrical property evaluation

The capacitance, resistance, and dissipation factor were measured directly using Hoiki 3532 LCR Hitester version 1.03, by varying frequencies 10 Hz to 5 MHz at room temperature. Square samples of thickness 3 mm, length 10 mm, and breadth 10 mm were used for the study. The test samples were fixed between two electrodes.

The volume resistivity (ρ) can be calculated from the resistance using the equation,

$$\rho = RA/t \qquad (1)$$

where ρ is volume resistivity, R is resistance, A is the area of cross section of the sample, and t is the thickness of the sample.

The dielectric constant ε' was calculated from the capacitance using the equation

$$\varepsilon' = Ct/\varepsilon_0 A \qquad (2)$$

where ε' is the dielectric constant of the material, ε_0 is the permittivity of air (8.85×10^{-12} F m^{-1}), C is the capacitance, A is the area of cross section of the sample, and t is the thickness of the sample.
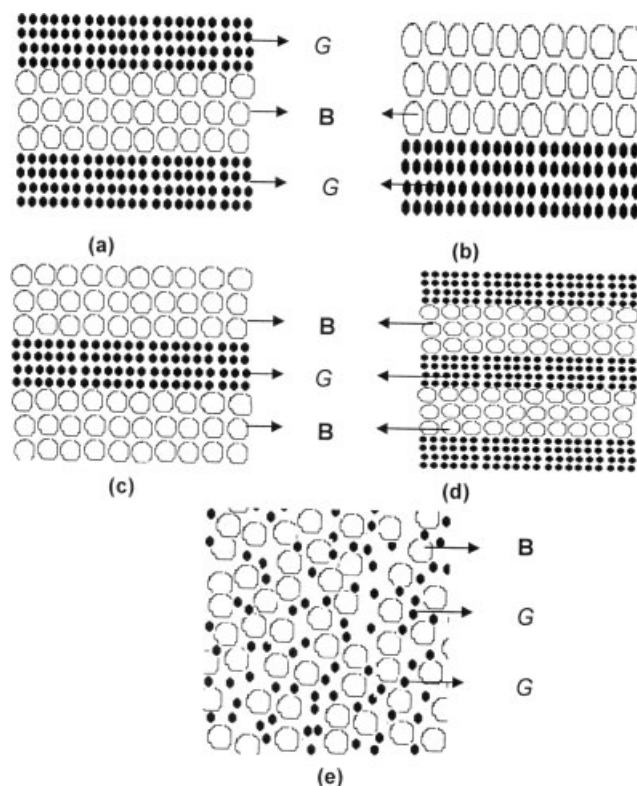
Figure 1 Schematic representation of various layering patterns in hybrid composites (a) GBG (trilayer), (b) BG (bilayer), (c) BGB (trilayer, (d) GBGBG, (e) Intimate mixture of glass and banana fibers.

RESULTS AND DISCUSSIONS

Dielectric constant (ε)

Banana fiber-reinforced PF composites

A plot of dielectric constant as a function of log frequency for neat PF and banana fiber-reinforced PF composites at different fiber loadings are given in Figure 2. The dielectric constant decreases with increase in banana fiber loading for the entire range of frequencies.

This decrease is higher for low frequencies and lower for very high frequencies. The highest dielectric constant value is obtained for neat PF. The dielectric constant decreases with increase in frequency for neat PF as well as for all the composites. The change of dielectric constant with frequency is the highest at very low frequencies. The change of ε with frequency can be explained in terms of interfacial polarization.

The dielectric constant of a material depends upon the polarizability of the material. If there is greater polarizability of the molecule, ε will be high. The dielectric constant of a polymeric material depends on interfacial, dipole, electronic, and atomic polarization.[23] The atomic and electronic polarizations are instantaneous polarization components, the effect of
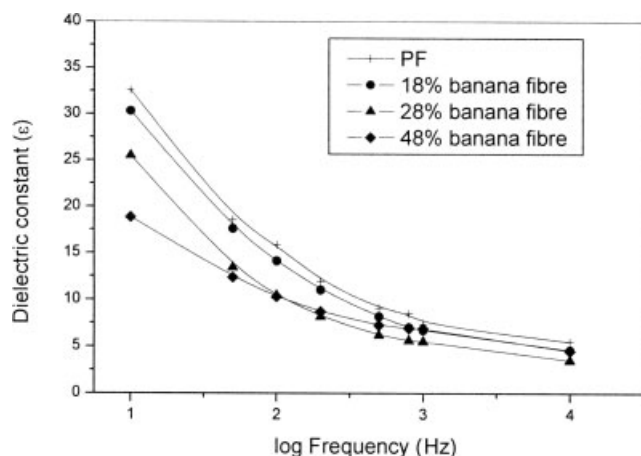
Figure 2 Plot of dielectric constant as a function of frequency for neat PF and banana fiber reinforced composites at different fiber loadings (volume %).

which is seen only at high frequencies. The dipole or orientation polarization occurs due to the presence of polar groups in the material. The interfacial polarization arises due to heterogeneity, which is highest at lower frequency. Because of the presence of polar groups in PF resin, ε value is found to be high. Since composite is heterogeneous, the interfacial polarization also exists. Interfacial polarization influences the low frequency ε values. The frequency dependence of dielectric constant is due to the fact that at low frequencies complete orientation of the molecule is possible. But at medium frequencies there is only little time for orientation. Orientation of molecule is not possible at all at very high frequencies. The frequency dependence of dielectric constant due to interfacial polarization is similar to those corresponding to dipole polarization. The ε value of lignocellulosic fiber is lower than that of phenol formaldehyde resin.[33] So the incorporation of fibers will decrease the dielectric constant.

Effect of chemical treatment

Figure 3 shows the variation of dielectric constant with frequency for chemically treated banana fiber-reinforced phenol formaldehyde composites. The influence of chemical treatments on the moisture content of banana fiber and banana/PF composites have been reported earlier.[34] Untreated banana fiber/ PF composite was found to have 4% moisture content. Previous studies in our laboratory[29] showed that sodium hydroxide treatment removes wax, cuticle, and alkali soluble constituents of hemicellulose and increases the crystallinity of the fiber thus decreasing the water sorption capacity of the fiber. So moisture content is decreased by the alkali treatment of fiber to 3.8%. Hence, the polarization due to the presence

of water molecule is decreased. So dielectric constant of alkali treated composite is lower than that of untreated fiber. The dielectric constant value of acetylated banana/PF composite is lower than untreated composite. This is because the hydrophilic nature of the fiber is decreased by acetylation there by reducing moisture content to 2.4%. The sodium hydroxide pretreatment before acetylation results in partial removal of hydrogen bonding in cellulose chains and make the fiber surface more reactive for acetylation.

Similarly, silane treatment also reduces the moisture content to 3.7%, which decreases the dielectric constant. In the case of heat-treated composites the interface is not affected. But what happens is that there is an increase in crystallinity of the fiber,[35] which can improve the composite properties. Similarly, the decomposition of less stable components and volatile components occurs by the heating thus reducing the moisture content to 2.9%. So the dielectric constant is lower for heat treated composite also. The very low value of dielectric constant for cyanoethylated and latex-treated fiber composites is also due to the decrease in orientation polarization resulting from the increased hydrophobicity of the fiber. The moisture content decreases to 1.5%, due to the presence of hydrophobic latex coating in the latex-treated fiber. The cyanoethylation reduced the moisture content to 2%.

Effect of hybridization

Effect of hybridization using glass fibers on the dielectric constant of banana fiber-reinforced phenol formaldehyde composites is shown in Figure 4. The dielectric constant value is found to be higher for ba-
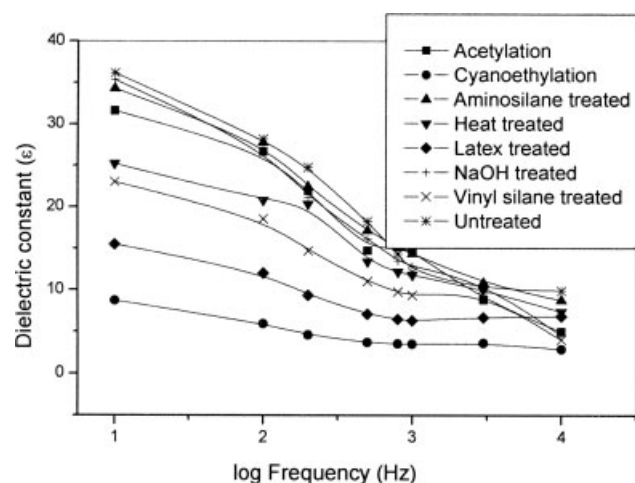

Figure 3 Effect of chemical treatments on dielectric constant versus log frequency for banana fiber reinforced PF composites.
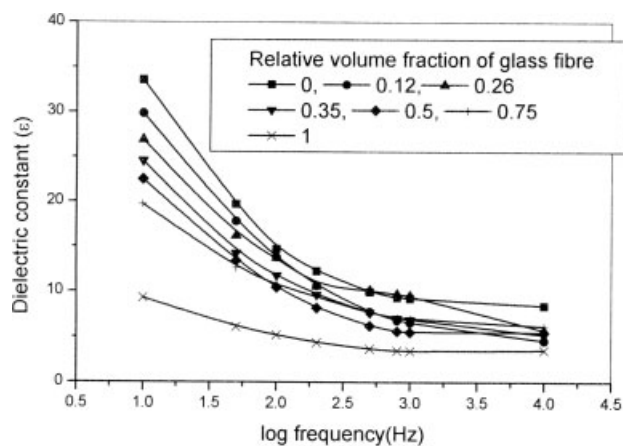
Figure 4 Effect of hybridization using glass fiber on the dielectric constant versus log frequency curve of banana fiber reinforced phenol formaldehyde composites.

Figure 5 Effect of hybrid layering patterns on the dielectric constant versus log frequency curve of banana/glass hybrid fiber reinforced phenol formaldehyde composites.

nana fiber/PF composites than that of glass fiber/PF composites. In hybrid composites, the dielectric constant decreases with the increase in glass fiber content. This is due to the hydrophobic nature of glass fiber while banana fiber is hydrophilic.

The part of the orientation polarization, which is due to the presence of polar water molecules, is very low in glass fiber composites when compared with banana fiber composites. So low ε value is obtained for glass/PF composites. But as the banana fiber content is increased, the polarization increases and the ε value increases.

Effect of hybrid layering pattern

The effect of hybrid layering pattern on the dielectric constant value of banana/glass/ hybrid fiber-reinforced composites are given in Figure 5. The dielectric constant values of hybrid composites with layering patterns GB, BGB, and intimate mix of B and G are found to be higher than that of GBG and GBGBG layering patterns. This is because of the presence of banana fiber in the outer layers of composites, which will increase the dielectric constant of hybrid composites with layering patterns GB, BGB, and intimate mix of B and G. But in GBG and GBGBG arrangements the banana fiber is in the core and glass fiber mat at the periphery, resulting in low interfacial polarization due to polar water molecules.

Volume resistivity

Banana fiber-reinforced PF composites

The resistivity of lignocellulosic fibers depends on the moisture content, crystalline, and amorphous component present, presence of impurities, chemical composition, cellular structure, microfibrillar angle etc. The shape of reinforcement determines the interparticle
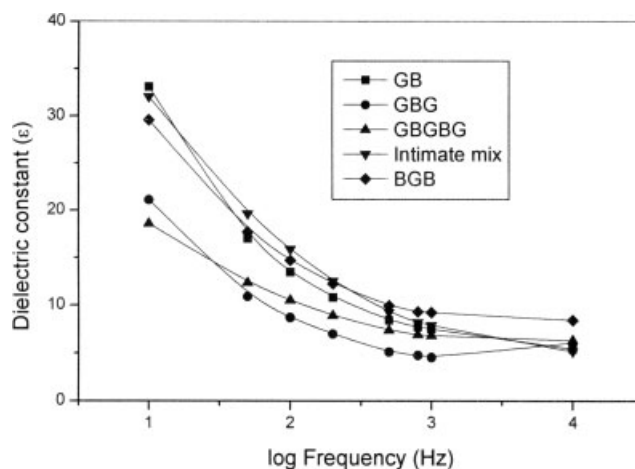
contact, which affects the conductivity of the system. Fibers and flakes having elongated shapes enhance electrical conductivity effectively.[36] The moisture content in fibers increases conductivity and the composite defects during processing will increase the resistivity.[25] A plot of volume resistivity as a function of log frequency for banana fiber-reinforced phenol formaldehyde composite is given in Figure 6. In all the composites the volume resistivity decreases with increase in frequency.

As the frequency increases the resistivity shows a decrease due to the interfacial polarization arising because of the heterogeneity of the system. The maximum volume resistivity is obtained for neat PF. The incorporation of banana fiber decreases the volume resistivity of the composite. In other words, the electrical conduction increases with increase in banana fiber loading.
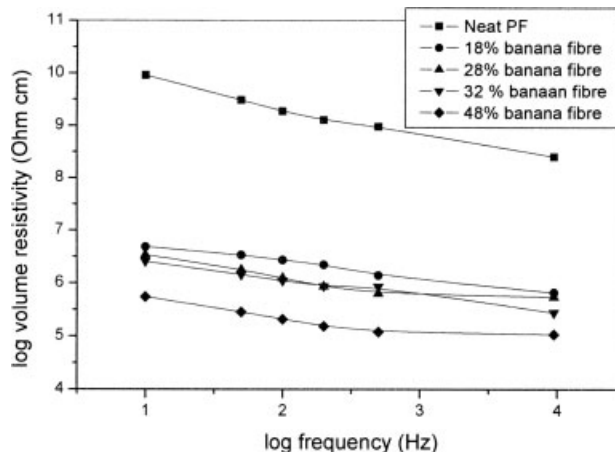

Figure 6 Log volume resistivity versus log frequency curves of banana fiber reinforced PF composites at different fiber loadings.
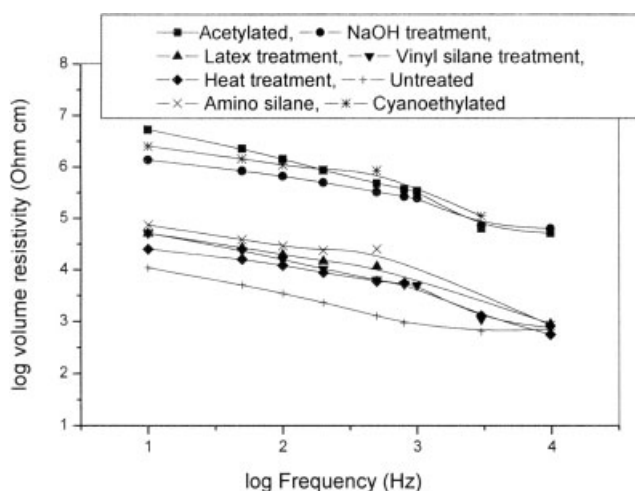
Figure 7 Effect of chemical treatments on log volume resistivity versus log frequency curves of banana fiber reinforced PF composites at different fiber loadings.

The hydrophilicity of the cellulose fiber is responsible for the greater conductivity of the composite. In the case of polymeric materials, most of the current flow through the crystalline regions and noncrystalline region allows current to pass through it mainly when moisture is present.[26] The phenol formaldehyde is an amorphous polymer and it has high volume resistivity. But the chemical composition and structure of lignocellulosic banana fiber is very complex. The hydroxyl groups in the hydrophilic fiber can absorb moisture and hence the presence of the natural fiber increases the conductivity of the resin.

Effect of chemical treatment

The log volume resistivity versus log frequency curves of untreated, alkali treated, cyanoethylated, acetylated, latex treated, silane treated, and heat treated banana fiber-reinforced phenol formaldehyde composites are given in Figure 7.

Volume resistivity values of all the treated composite samples remained higher than that of the untreated composites. Maximum values of volume resistivity are obtained for cyanoethylated and acetylated composites. In fact, conductivity of the cellulose fiber is mainly due to the presence of water molecules absorbed by fiber surface. Cyanoethylation results in reduction of hydroxyl groups and introduction of cyanoethyl groups by the following reaction.

$$Fibre-OH+CH_2=CHCN\longrightarrow Fibre-O-CH_2-CH_2CN$$

Silanes can be represented by YR—Si (OR$_2$)$_3$, where R is an aliphatic linkage that serves to attach the functional organic groups to silicon. OR$_2$ is a

hydrolysable alkoxy group. Traces of moisture in the fiber hydrolyse the silanes to form silanols.

$$YR_1Si(OR_2)_3 \xrightarrow{H_2O} YR_1Si(OH)_3$$
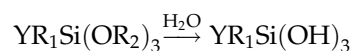
The hydrolyzed silanol forms strong covalent bonds or H— bonds with OH group of cellulose. The individual coupling agent molecules attached to cellulose forms a continuous link. The long hydrophobic polymer chain of polymerized silane can adhere to the matrix mainly because of Van der waals type attractive force. As a result, silane-coupling agents form a bridge at the interface. In aminosilane the Y unit is —NH$_2$ group, which can react with —OH groups of other molecules of silanes or cellulose or OH groups of PF matrix. In vinyl (2-ethoxy methoxy) silane the Y group is the vinyl group. The long hydrophobic polymer chain of polymerized silane reduces the moisture absorption of the composite. So both amino silane and vinyl silane treated composites containing lower moisture content is found to have higher volume resistivity value. Introduction of hydrophobic latex coating in the fiber increases the volume resistivity values of latex-treated composites. Heat treated fiber/PF composites are found to have lower moisture content than untreated fiber/PF composites. So heat treatment also increases the resistivity of the composite.

Effect of hybridization

The effect of hybridization using glass fibers on the volume resistivity of banana fiber-reinforced phenol formaldehyde composites is given in Figure 8. The volume resistivity of glass fiber-reinforced composites is higher than that of banana fiber composite. This is due to the high hydrophilicity of banana fiber
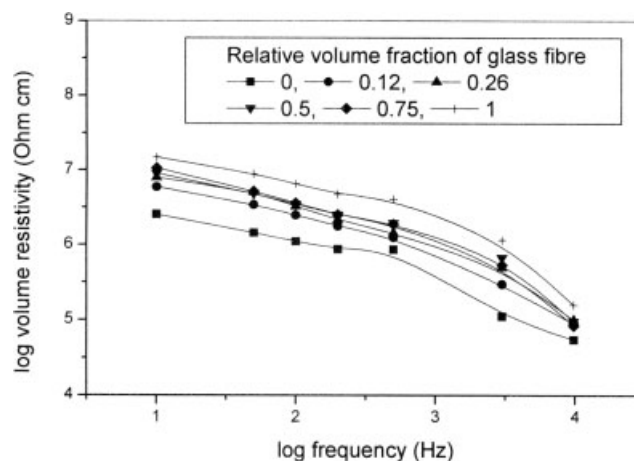

Figure 8 Volume resistivity versus log frequency curves of banana glass hybrid fiber reinforced PF composites at different relative volume fractions of glass.
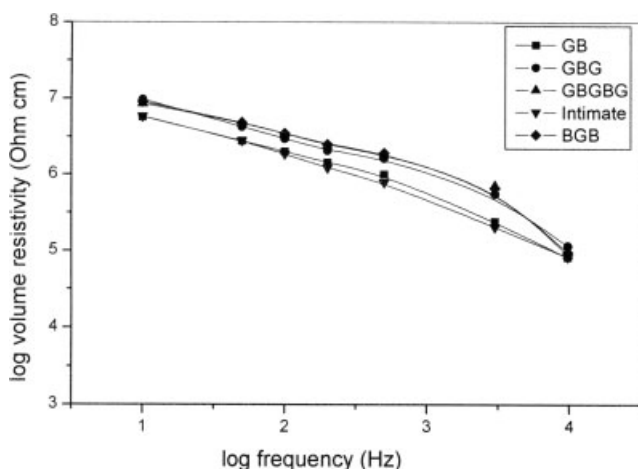
Figure 9 Volume resistivity versus log frequency curves of banana/glass hybrid fiber reinforced PF composites having different layering patterns.

compared with glass fiber. The volume resistivity values of hybrid composites do not show a marked difference. But there is a slight increase in volume resistivity with increase in glass fiber content.

Effect of hybrid layering pattern

Effect of fiber layering patterns on the volume resistivity of banana/glass hybrid composites containing banana : glass volume ratio 0.25 : 0.75 is given in Figure 9. The composite having intimate mixture of banana fiber and glass fiber is found to have minimum volume resistivity value. The layered composites are having higher volume resistivity than that of intimate mix. The hybrid composites having glass fiber at periphery (GBG and GBGBG) are found to have higher volume resistivity values than other arrangements (GB and BGB). This is due to high vol-
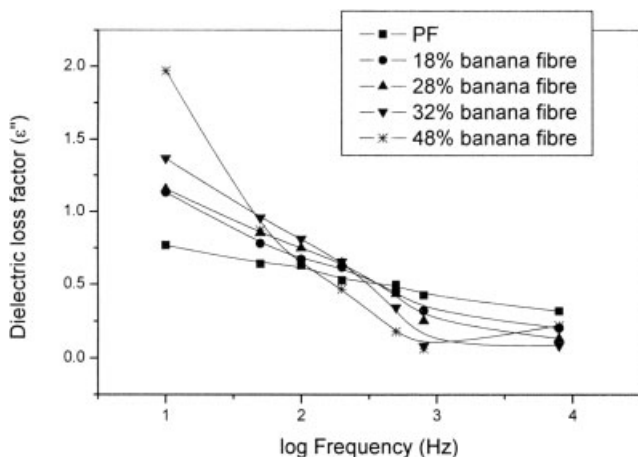

Figure 11 Effect of chemical treatments on loss factor versus log frequency curves of banana fiber-reinforced PF composites.

ume resistivity value of glass fiber compared with banana fiber. The resistivity of hybrid composites with layering patterns Intimate mix, GB and BGB arrangements are low due to the presence of banana fiber in the periphery in these composites.

9.3. Dielectric loss factor

Figure 10 shows the loss factor as a function of logarithm of frequency for banana fiber-reinforced PF composites at varying fiber loadings. In banana fiber composites, at low frequencies as the fiber loading increases, the loss factor also increases. But at high frequencies a reverse behavior is observed and the values come closer. This is due to polarization of the fibers at low frequencies, which is absent at higher frequencies. As the fiber loading increases, the heter-

Figure 10 The variation of loss factor as a function of logarithm of frequency for banana fiber reinforced composites with different fiber contents (volume).
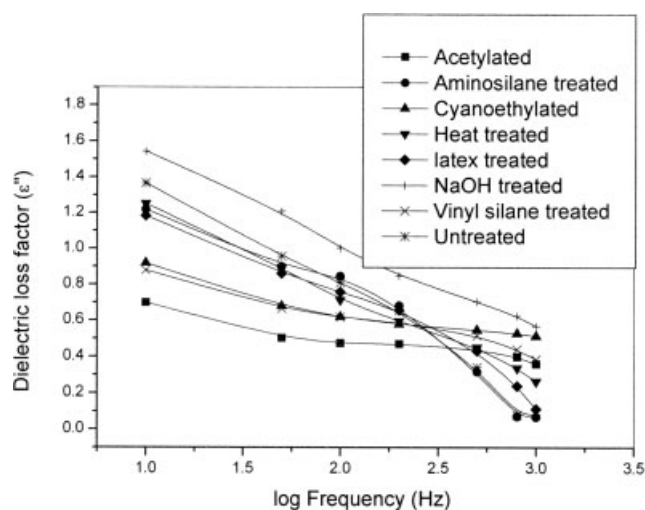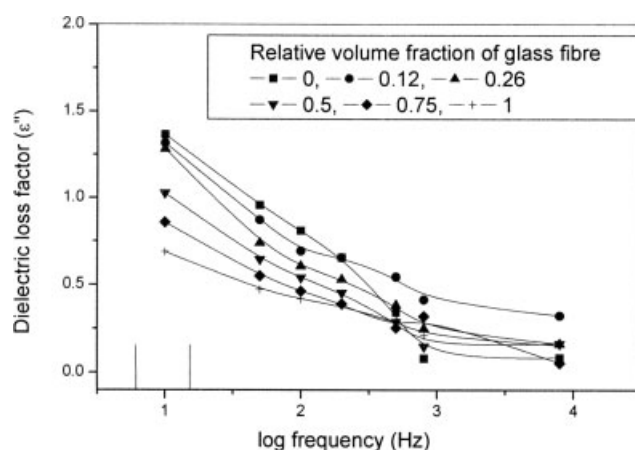

Figure 12 Effect of hybridization with glass fiber on loss factor versus log frequency curves of banana fiber reinforced PF composites.

ogeneity of the system increases and the polarization increases. Figure 11 shows the variation of loss factor with frequency for banana fiber-reinforced composites after chemical treatments. In this study, thus we see that all types of treatments except the NaOH treatment decrease the loss factor. This is due to reduced orientation polarization as a result of chemical treatment. The loss factor is found to be very low for acetylated and cyanoethylated composites. This is because these two treatments introduce acetyl and cyanoethyl groups in the fiber thus reducing the hydophilicity of the composite. Figure 12 is the loss factor versus log frequency plots of banana/glass/hybrid composites. Here, maximum loss factor is obtained for banana fiber composites. This is due to the higher degree of polarization of banana fiber, which is very low for glass fibers. As the glass fiber loading increases, the loss factor also increases.

CONCLUSIONS

The electrical properties of banana fiber-reinforced phenol formaldehyde composites have been evaluated. Dielectric constant (ε) values decreased with frequency for all the composites. As the ε value of banana fiber is lower than PF, the ε values of composites decrease with increase in banana fiber loading. Chemical treatments with silanes, acetylation, cyanoethylation, and latex treatment decreased the hydrophilicity of the fiber and the moisture content in the composite there by decreasing the ε value. Hybridization with glass fiber also decreased the ε value of banana/PF composites. Hybrid layering patterns with glass fiber at periphery and banana fiber at core were found to have lower dielectric constant value than other layering patterns having hydrophilic banana fiber at periphery. Acetylation, cyanoethylation, silane treatments, heat treatment, and latex treatment increased the volume resistivity of the composites due to reduced moisture content and increased fiber/matrix adhesion. Glass fiber composites were found to have higher volume resistivity than banana fiber composites and in hybrid composites, the volume resistivity increased with increase in glass fiber content. Layered composites with glass fiber at periphery got higher volume resistivity than other layering patterns. The loss factor was decreased by the chemical treatments and in hybrid composites; it increased with increasing volume fraction of glass fibers.

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