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Dielectric characteristics of sisal–oil palm hybrid biofibre reinforced natural rubber biocomposites

Maya Jacob · K. T. Varughese · Sabu Thomas

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Abstract Natural rubber was reinforced with sisal and oil palm fibers. Biocomposites were prepared by varying the weight fraction of the fibers. The dielectric properties such as dielectric constant, volume resisitivity and dielectric loss factor of the biocomposites were evaluated as a function of fiber loading, frequency and chemical modification of fibers. The dielectric constant values were found to be higher for fiber reinforced system than the gum due to polarization exerted by the incorporation of lignocellulosic fibers. Chemical modification of fibers resulted in decrease of dielectric constant values and volume resisitivity values. The volume resisitivity of the composites was found to decrease with fiber loading due to increase of hydrophilicity imparted by the lignocellulosic fibers. The dissipation factor was found to increase with fiber content.

Introduction

Over the last few years, natural fibers have emerged as the main reinforcement for high performance composite materials. The uses of lignocellulosic fibers have ranged from the construction industry to the automotive industry. The main attraction of bio-fiber reinforced composites lie in their low density and high strength. The most interesting aspect about natural fibers is their positive environmental impact. As they come from a natural resource they are completely biodegradable and nonabrasive material and can be easily eliminated after the degradation of the polymer. Also the hollow cellular structure provides excellent insulation against heat and noise. The high sound attenuation of lignocellulosic composites is another added advantage. Vegetable fibers also provide a relativity reactive surface, which can be used for grafting specific groups and inducing new functional entities. In addition, the recycling by combustion of polysaccharide filled composites is easier in comparison with inorganic fillers [1].

Alongside the emergence of natural fibers there has also been a growing interest in electrically conductive polymer composites. A great deal of work has been performed to make polymers conductive by incorporating conductive fillers. For a given polymer type the electrical properties is determined by the amount and type of conductive additives. The conductivity of such composites is due to the formation of a continuous network of filler particles throughout the polymer matrix. Such electrically conductive composite materials are widely being used in the areas of electrostatic discharge dissipation, electro-magnetic interference shielding and various other electronic applications.

Hong and Wool [2] developed a new low dielectric constant material suited to electronic materials applications using hollow keratin fibres and chemically modified soybean oil. The unusual low value of dielectric constant obtained was due to the air present in the hollow microcrystalline keratin fibres and the triglyceride molecules. The authors are of the opinion that the low cost composite made from avian sources and plant oil has the potential to replace the dielectrics in microchips and circuit

M. Jacob · S. Thomas (✉)
School of Chemical Sciences, Mahatma Gandhi University,
Priyadarshini Hills P.O., Kottayam, Kerala 686 560, India
e-mail: sabut@sancharnet.in

K. T. Varughese
Central Power Research Institute, Polymer Laboratory,
Bangalore 560 080, India

boards in the ever growing electronics materials field. In an extension of the above study the authors have also observed that the coefficient of thermal expansion (CTE) of the composite was low enough for electronic applications and similar to the value of silicon materials or polyimides used in printed circuit boards [3].

Recently the electrical properties of vapor grown carbon fiber (VGCF) reinforced vinyl ester composites was investigated by Xu et al. [4] The electrical properties exhibited a percolation behavior with a sharp drop in resistivity occurring between 2 and 3 wt% VGCF loading. Composites made from nitric acid-oxidized VGCF were found to act as insulators even at 15 wt% VGCF loading. The dielectric properties of short jute fiber-reinforced polypropylene composites were investigated by Cabral et al. [5]. A change in the dependence of dielectric properties with fiber loading at critical fiber content was observed.

In an interesting study, the electrical properties of sisal and coir fiber reinforced LDPE composites were studied by Paul and Thomas [6]. The authors found that dielectric constant values increased with fiber loading this was more predominant at low frequencies. Volume resistivity values were found to decrease with fiber content. In an extension of the above studies, Paul and Thomas [7] looked into the effect of chemical modification on the electrical properties of sisal fiber reinforced LDPE composites. They found that the dielectric values decreased with chemical modification because of the decrease in hydrophilicity of fibers. The volume resistivity values of the composites containing chemically treated fibers were found to be higher than untreated ones.

The influence of sisal fiber orientation on the electrical properties of sisal fiber reinforced epoxy composites was studied by Chand and Jain [8]. The authors found that dielectric constant and tan δ of the epoxy, 0° and 90° oriented sisal fibre epoxy composites decreased with increasing frequency and a.c. conductivity increased with increasing frequency. Near the transition temperature of the epoxy matrix, the observed properties showed anomalous behavior. Clear relaxation peaks for tan δ around 169 °C were observed in epoxy resin, shifting to the lower temperature side with increasing frequency.

The electrical conductivity of nitrile rubber composites filled with different blends of short carbon fiber and conductive carbon black was studied by Pramanik et al. [9]. It was seen that the conductivity of the composites increased with increasing carbon fiber loading. The effect of coupling agent on the electrical properties of carbon fiber reinforced phenolic resin composites has been studied by Choi et al. [10]. The authors observed that surface treatment of carbon fibers affected the dielectric behavior of composites with short carbon fibers while they did not affect those of the composites with fabric type carbon fibers. Composites containing carbon fibers treated with coupling agent also exhibited higher electrical conductivity.

The mechanical and electrical anisotropy of pineapple fibers was investigated by Dutta et al. [11]. They observed a sharp increase of dielectric constant and fall of loss factor along the fiber direction compared with transverse direction. The authors established that crystallinity was greater along the fiber direction. Modification of dielectric and mechanical properties of rubber ferrite composites containing manganese zinc ferrite have been studied by Anantharaman and co-workers [12]. These modified composites were found to be ideally suited for many modern applications where ceramic materials have some drawbacks.

In an innovative study, the biodegradability of protein filled polymer composites using dielectric measurements was investigated by Tchmutina et al. [13] The composites comprised of a metallocene-based linear low-density polyethylene and a biopolymer, hydrolysed protein (HP), which was isolated from the chrome-tanned solid leather waste of the tanning industry by enzymatic hydrolysis. Biodegradation experiment was conducted in an aqueous phase of malt extract medium by an enzyme, Aspergillus oryzae for 3 weeks. The authors observed that the biodegradation coefficients determined using dielectric measurements were in good agreement with the weight loss of the composites. An added advantage of the above method was that it can be used for investigation of uniformity of biodegradation at different points of the sample area and layer-by-layer distribution of biodegradation coefficient in the sample.

The effects of particle compositions, particle shapes, and oxidation temperatures on the dielectrical characteristics of conductive adhesives were investigated by Lin and Chiu [14]. The authors observed that the electrical properties of the conductive adhesives were strongly affected by the particle compositions and oxidation temperatures. Particle shape was not found to have much effect.

Studies involving dielectric properties of waste paper [15] and newsprint fibre [16] reinforced natural rubber composites have also been reported. The reinforcement of biofiber in rubber composites and their applications has been well documented [17]. We have also reported on the mechanical [18] aspects of sisal and oil palm hybrid biofiber reinforced natural rubber biocomposites. However there has not been a systematic study of the electrical properties of hybrid bio-fiber reinforced natural rubber biocomposites till date. This manuscript attempts to investigate the influence of fiber loading, frequency and chemical modification on the dielectric properties of the composites. The influence of chemical modification on the resistivity is also investigated.

Theoretical background

Electrical conduction in composites can be explained by three main theories.

(a) Conduction path theory [19] – The conducting filler forms few continuous chains in the matrix. Electrons move through this continuous network from one place to another causing electrical conduction.
(b) Tunnel effect theory [20] – Electrical conduction is assumed to take place not only by interparticle contact but also by electrons being able to jump across gaps between conducting elements in the polymer matrix. There is a threshold value of these gaps which is equivalent to interparticle contact.
(c) Electric field radiation theory [21] – It is assumed that an emission current is caused to flow by the high electric field being generated between conducting elements separated by a gap of a few nm.

The dielectric constant or static permittivity E' of a material is defined as the ratio of the capacitance of a condenser containing the material to that of the same condenser under vacuum. The capacitance of a condenser measures the extent to which it is able to store charges.

Dielectric constant (ε') can be calculated from the capacitance using the equation:

$$\varepsilon' = \frac{Ct}{E_o A} \tag{1}$$

where C – capacitance of the material

t – thickness of the sample.
E_o – permittivity or capacitivity of free space (8.85×10^{-12} Fm^{-1})
A – area of sample under electrode.

The insulation resistance of a material depends on its volume resistance, thus the volume resistivity (ρ) can be calculated by using the equation

$$\rho = \frac{RA}{t} \tag{2}$$

where R is the volume resistance (Ω), A is the area of cross-section and t is the thickness of the samples.

The electrical conductivity (σ) is calculated according to the following equation

$$\sigma = {}^{1}\!/_{\rho} \tag{3}$$

The ratio of the imaginary to the real dielectric loss angle constants $\varepsilon''/\varepsilon'$ or tangent of the dielectric loss angle is commonly employed as a direct measure of the dielectric loss. It is also known as the dissipation factor and is a measure of the power dissipated.

Dissipation factor tan δ can be calculated from the equation

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \tag{4}$$

where ε'' is the loss factor and ε' is the dielectric constant.

Experimental

Materials

Sisal fiber was obtained from Sheeba Fibers, Poovancode, Tamil Nadu. Oil palm fiber was obtained from Oil Palm India Limited. Natural rubber used for the study was procured from Rubber Research Institute of India, Kottayam. All other ingredients used were of commercial grade. The physical properties of sisal and oil palm fibers are given in Tables 1 and 2.

Alkali treatment

Sisal and oil palm fibers of lengths 10 mm and 6 mm were treated for 1 h with NaOH solutions of concentrations 0.5, 1, 2 and 4%, respectively. The fibers were further washed

Table 1 Properties of sisal fiber

Chemical constituents (%)	
Cellulose	78
Hemicellulose	10
Lignin	8
Wax	2
Ash	1
Physical properties of sisal fiber	
Diameter (mm)	0.1212
Tensile strength (MPa)	530–630
Young's modulus (GPa)	17–22
Microfibrillar angle	20–25
Elongation at break (%)	3–7

Table 2 Properties of oil palm fiber

Chemical constituents (%)	
Cellulose	65
Hemicellulose	–
Lignin	19
Ash content	2
Physical properties of oil palm fiber	
Diameter (μm)	150–500
Tensile strength (MPa)	248
Young's modulus (MPa)	6700
Elongation at break %	14
Microfibrillar angle (°)	46

with water containing acetic acid. Finally the fibers were washed again with fresh water and dried in an oven.

Silane treatment

The silanes used were fluorosilane (F8261), 3-aminopropyl tri-ethoxy silane (A1100) and vinyl triethoxysilane (A151). 0.4% of the respective silanes was prepared by mixing with an ethanol/water mixture in the ratio 6/4 and was allowed to stand for 1 h. The pH of the solution was maintained at 4 with the addition of acetic acid. Sisal fabric was dipped in this solution and was allowed to stand for 1.5 h. The ethanol/water mixture was drained out and the fabric was dried in an air and then in an oven at 70 °C until it was completely dry.

Preparation of composite

Formulation of mixes is shown in Tables 3 and 4. NR was masticated on the mill for 2 min followed by addition of the ingredients. The composite materials were prepared in a laboratory two-roll mill (150 × 300 mm). The nip-gap, mill roll, speed ratio, and the number of passes were kept the same in all the mixes. The samples were milled for sufficient time to disperse the fibers in the matrix at a mill opening of 1.25 mm. The bonding system consisting of resorcinol and hexamethylene tetramine was incorporated for mixes containing treated fibers. The fibers were added at the end of the mixing process, taking care to maintain the direction of compound flow, so that the majority of fibers followed the direction of the flow.

Measurements

Rectangular specimens of 1.9 mm thickness were used. Samples were prepared by cutting from the composite specimens using a die. The test samples were coated with conductive graphite paint on either side and copper wires are fixed on both sides of the samples as electrodes. The capacitance, resistance and dielectric loss factor were measured directly at room temperature using a Impedance Analyzer by varying the frequencies (500 Hz–6 MHz)

Scanning electron microscopic studies were conducted using JEOL, JSM 5800 to analyze the fracture behavior of

Table 3 Formulation of mixes A to E (Fiber Loading)

Ingredients	Mixes [phr]			
	G	B	A	E
NR	100	100	100	100
ZnO	5	5	5	5
Stearic acid	1.5	1.5	1.5	1.5
TDQ[a]	1	1	1	1
CBS[b]	0.6	0.6	0.6	0.6
Sulphur	2.5	2.5	2.5	2.5
Sisal fiber Untreated Fiber length, (10 mm)		5	15	25
Oil palm fiber Untreated Fiber length (6 mm)		5	15	25

[a] 2,2,4 trimethyl-1,2-dihydro quinoline

[b] *N*-cyclohexylbenzothiazyl sulphenamide

Table 4 Formulation of mixes (I to R) {phr}

Ingredients	I	J	K	L	P	Q	R
NR	100	100	100	100	100	100	100
ZnO	5	5	5	5	5	5	5
Stearic acid	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Resorcinol	7.5	7.5	7.5	7.5	7.5	7.5	7.5
Hexa[a]	4.8	4.8	4.8	4.8	4.8	4.8	4.8
TDQ	1	1	1	1	1	1	1
CBS[b]	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Sulphur	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Sisal fiber	15	15	15	15	15	15	15
Treated Fiber length, (10 mm)	0.5% NaOH 1 hr	1% NaOH 1 hr	2% NaOH 1 hr.	4% NaOH 1 hr	0.4% Fluoro-silane	0.4% vinyl-triethoxy silane	0.4% 3-amino propyl tri-ethoxy silane
Oil palm fiber	15	15	15	15	15	15	15
Treated Fiber length (6 mm)	0.5% NaOH 1 hr	1% NaOH 1 hr	2% NaOH 1 hr	4% NaOH 1 hr	0.4% Fluoro-silane	0.4% vinyl triethoxy silane	0.4% 3-amino propyl tri-ethoxy silane

[a] Hexamethylene tetramine

[b] *N*-cyclohexylbenzothiazyl sulphenamide

the composites. The fracture ends of the tensile specimens were mounted on aluminium stubs and gold coated to avoid electrical charging during examination.

Results and discussion

Dielectric constant

Effect of fiber loading and chemical modification

The dielectric constant of a material depends upon the polarizability of the molecules. The polarizability of non-polar molecules arises from electronic polarization (in which the application of applied electric filed causes a displacement of the electrons relative to the nucleus) and atomic polarization (in which the application of applied electric filed causes a displacement of the atomic nuclei relative to one another). In the case of polar molecules a third factor also comes into play which is orientation polarization (in which the application of applied electric filed causes an orientation of dipoles)

Figure 1 presents the effect of fiber loading on the dielectric constant values of sisal/oil palm hybrid fiber reinforced natural rubber composites. It can be seen that dielectric constant increases with fiber loading at all frequencies. The dielectric constant values decrease with increase in frequency for all the composites. The minimum dielectric constant is exhibited by the gum compound and maximum by composite containing 50 phr of fiber. This is because of the fact that natural rubber is a non-polar material and has only instantaneous atomic and electronic polarization to account. For a non-polar material, ε' equals the square of the refractive index (n) as given by the equation

$$\varepsilon' = n^2 \tag{5}$$

The refractive index of natural rubber is 1.591 and hence theoretical dielectric constant is 2.5381while experimental dielectric constant is 3.84. The higher experimental value may be due to the occurrence of interfacial polarization, which arises due to the presence of impurities. The presence of two lignocellulosic fibers (sisal and oil palm) in natural rubber leads to an overwhelming presence of polar groups giving rise to dipole or orientation polarizability. The over all polarizability of a composite is therefore the sum of electronic, atomic and orientation polarization giving rise to higher dielectric constant. Hence the dielectric constant increases with increase in fiber loading at all frequencies.

The decrease in dielectric constant with frequency is due to decease in orientation polarization at high frequencies. At low frequencies, complete orientation of the molecule is possible while at medium frequencies there is only little time for orientation. Orientation of the molecules is not possible at all at very high frequencies.

Figure 2 shows the influence of chemical modifications on sisal and oil palm fibers on dielectric constant values of sisal/oil palm fiber reinforced composites. Here also we can see that dielectric constant decreases with increase in frequency due to decrease in orientation polarization. Another observation is that chemical modification of fibers results in lowering of dielectric constant. This is due to the decrease of orientation polarization of composites containing treated fibers. Chemical treatment results in reduction of moisture absorption capacity of fibers due to the reduction in interaction between polar –OH groups of lignocellulosic fibers and water molecules. The resultant decrease of hydrophilicity of the fibres leads to lowering of orientation polarization and subsequently dielectric constant.

On analyzing the graph further we can see that dielectric constant of composites containing alkali treated fibers decreases with concentration of alkali. The composite containing fibers treated with 4% alkali exhibits the lowest dielectric constant among the other alkali treated composites. Besides the removal of hemicellulose and waxes, the treatment with NaOH solution promotes the activation of hydroxyl groups of cellulose unit by breaking the hydrogen bond. The authors in a previous study observed that mercerization of sisal and oil palm fibers in natural rubber composites resulted in enhanced tensile properties [22].

Alkali treatment results in unlocking of the hydrogen bonds making them more reactive. In the untreated state the cellulosic –OH groups are relatively unreactive as they form strong hydrogen bonds. In addition to this, alkali

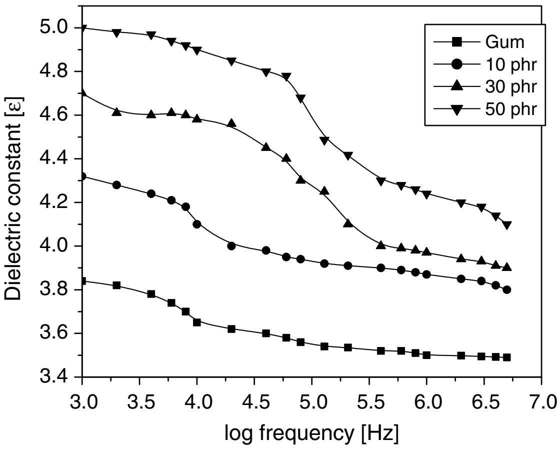


Fig. 1 Variation of dielectric constant with frequency as a function of fiber loading

treatment can lead to fibrillation i.e. breaking down of fibers into smaller ones. All these factors provide a large surface area and give a better mechanical interlocking between the fiber and matrix and thus reduce water absorption. This results in lowering the overall polarity and hydrophilicity of the system. This results in reduction of orientation polarization and consequently dielectric constant of the composites.

Another interesting observation from Fig. 2 is that out of the three types of silanes, fluorosilane, vinyl silane and amino silane the minimum dielectric constant is exhibited by fluoro silane, while the highest ε is given by vinyl silane treated samples. Mercerization treatment prior to silane treatment is an effective means of attaining better property retention to composites exposed to moisture. The potential advantage of using silane coupling agents are their inherent natural attraction with both the natural fiber and resin matrix.

The reaction mechanisms can be explained as follows. First, silane reacts with water to form silanol and an alcohol.

$$NH_2(CH_2)_3Si(OC_2H_5)_3 + 3H_2O \rightarrow NH_4(CH_2)Si(OH)_3 + 3C_2H_5{-}OH \qquad (6)$$

In the presence of moisture the silanol reacts with hydroxyl group attached to the cellulose molecules of the fiber through an ether linkage with removal of water [23].

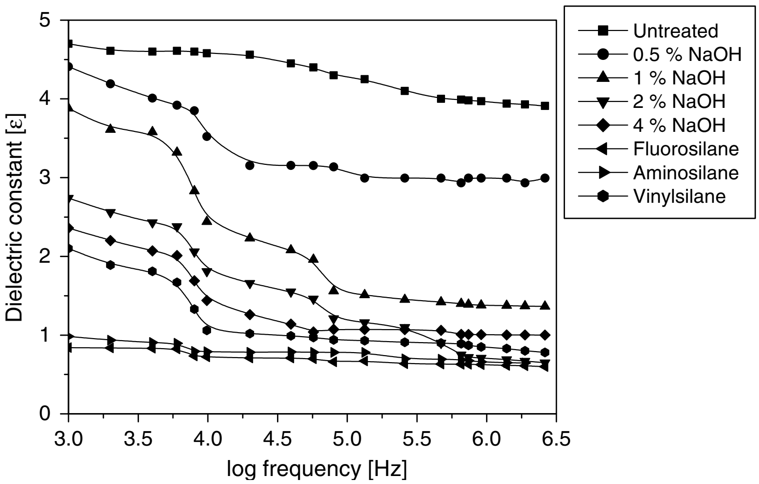
$$\begin{aligned} &NH_2\,(CH_2)_3\,Si\,(OH)_3 + H_2O + \text{Fiber - OH}[-H_2O] \\ &NH_2\,(CH_2)_3Si(OH)_2 - \text{O - Fiber} \end{aligned} \qquad (7)$$

Similar reactions takes place for fluoro silane and vinyl silane coupling agents. The rubber matrix gets attached to the organo functional group of silane coupling agent either through a covalent bond or hydrogen bond. Here the possibility of hydrogen bonding is greater due to the presence of nitrogen in amino group and fluorine in fluro silane group, while no such possibility exists in vinyl group. The schematic sketch of the interaction between rubber, amino and fluoro silane coupling agent and fiber is shown in Fig. 3.

This kind of bonding cannot be possible for the vinyl silane group. The hydrogen bond formed by the fluorine to the matrix will be stronger than hydrogen bond formed by nitrogen to the matrix due to the high electronegative character of fluorine atom . Thus in fluro-silane treated composites the interfacial adhesion will be stronger leading to decreased hydrophilicity and therefore lowering of dielectric constant when compared to amino-silane treated and vinyl-silane treated composites.

The improved interfacial adhesion in chemically modified composites can be further understood by looking into fracture topography of the system. Figure 4 depicts the scanning electron micrographs (SEM) of tensile fracture surfaces of untreated, alkali treated and silane treated composites. Figure 4(a) shows the tensile failure surface of composite containing untreated composite. The presence of cavities is clearly visible in both the figures. This indicates that the level of adhesion between the fibers and the matrix is poor and when stress is applied it causes the fibers to be pulled out from the rubber easily leaving behind gaping cavities. The SEM of composite containing fibers treated with 4% NaOH is presented in Fig. 4(b). This figure shows short broken fibers projecting out of the rubber matrix. This indicates that the extent of adhesion between the fibers and rubber matrix is greatly improved and when stress is applied the fibers break but do not wholly come out of the matrix. Figure 4(c) presents the tensile fracture of composite containing fluorosilane treated fiber. Here we can see the presence of rubber particles adhering to the fiber surface and there are no considerable cavities seen indicating good interfacial adhesions.

Fig. 2 Variation of dielectric constant with frequency as a function of chemical modification



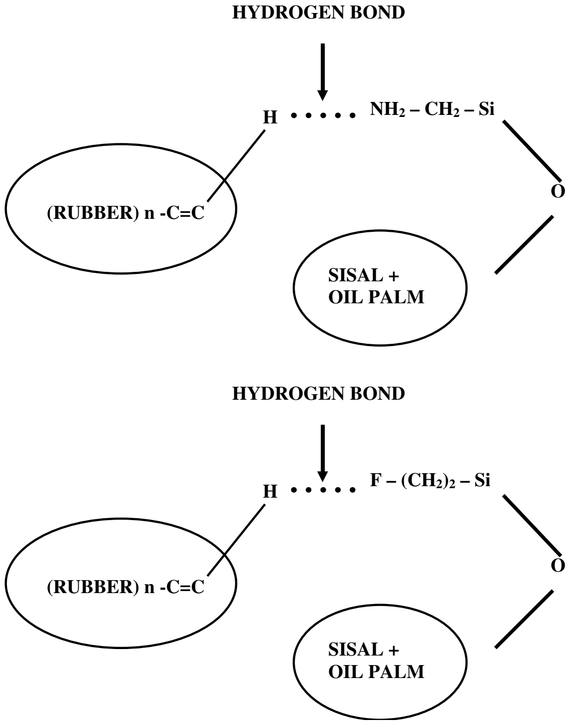


Fig. 3 Schematic sketch of the interaction between rubber, silane (amino and fluoro) coupling agents and fiber

Volume resistivity

Effect of fiber loading and chemical modification

The study of the volume resistivity of an insulating material is important because the most desirable property of an insulator is its ability to resist the leakage of electric current. Figure 5 shows the variation of volume resistivity with fiber loading. We can see that volume resisitivity decreases with frequency. Another interesting observation is that volume resistivity decreases with fiber loading. This implies that the conductivity increases upon addition of lignocellulosic fibers. This is due to the presence of polar groups which facilitate the flow of current. In polymers it is well known that most of the current flows through the crystalline regions and the passage of current in the amorphous regions is due to the presence of moisture. The presence of two lignocellulosic fibers increases the moisture content and hence increases the conductivity of the system.

The variation of volume resistivity with frequency as a function of chemical modification is presented in Fig. 6. It can be seen that volume resisitivity increases upon chemical treatment of fibers. As described earlier chemical modification of fibers results in lowering of moisture content and increased interfacial adhesion leading to increased resisitivity values. The resistivity is also dependent on dielectric constant by the equation

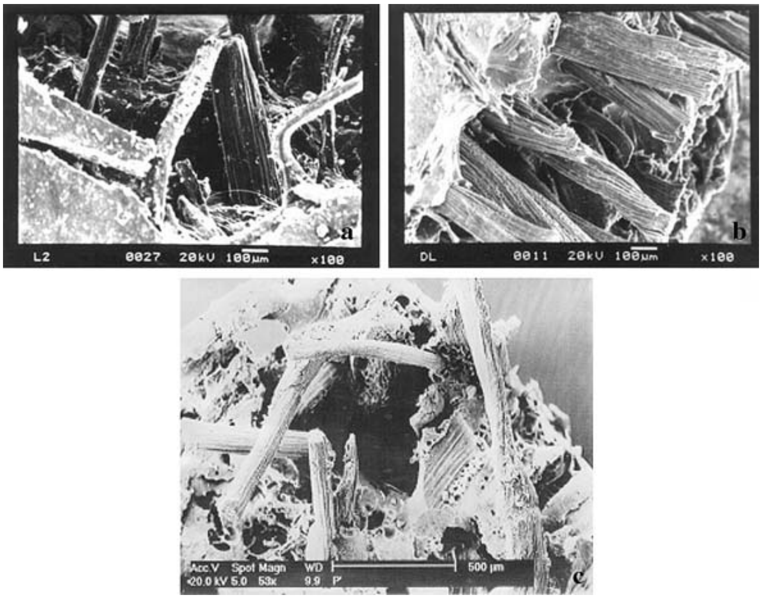
$$\log R_{10}(298^\circ\mathrm{K}^\circ) = 23 - 2\varepsilon'(298^\circ\mathrm{K}) \qquad (8)$$

where R is the resistivity and ε' is the dielectric constant. According to this equation the electric resistance of the composites decreases exponentially with increasing dielectric constants. As seen earlier chemical modification had resulted in lowering of dielectric constant which would automatically lead to an increase of resistivity values.

Electrical conductivity

The variation of electrical conductivity with fiber loading at two varying frequencies (4 and 6 Hz) is presented in Fig. 7. It can be seen that as fiber loading increases

Fig. 4 Scanning electron micrographs of (**a**) composite containing untreated fiber (**b**) composite containing fibers treated with 4% alkali (**c**) composite containing fibers treated with fluorosilane couping agent



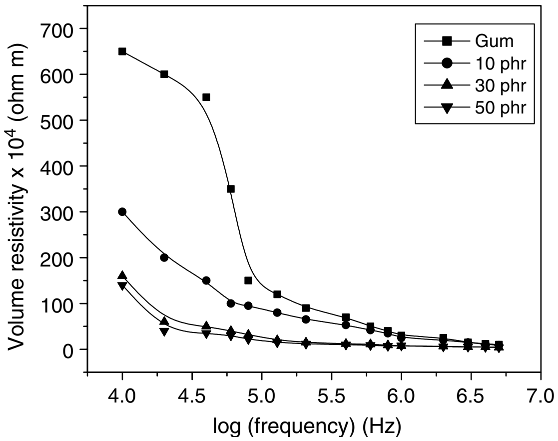


Fig. 5 Variation of volume resistivity with frequency as a function of fiber loading

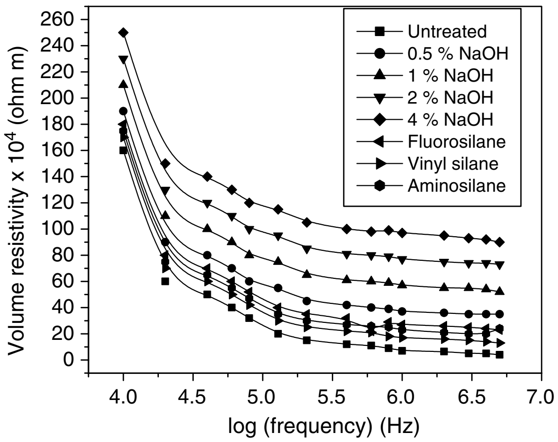


Fig. 6 Variation of volume resistivity with frequency as a function of chemical modification

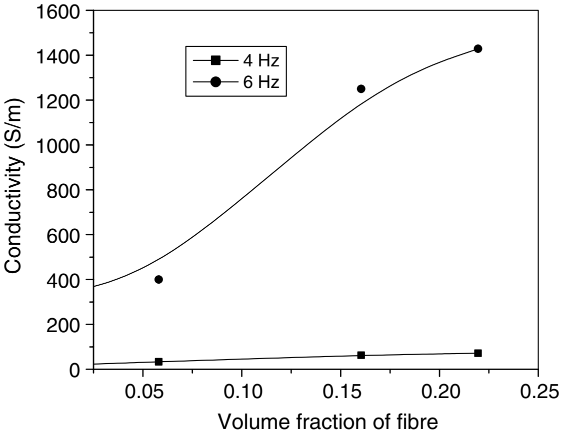


Fig. 7 Variation of conductivity with fiber volume fraction at varying frequencies

conductivity also increases. This is due to increment of polar groups present in the lignocellulosic fibers. Conductivity is also dependent on the dispersion of fibers; in order to get better conductivity the dispersed additive must either be arranged in a chain structure or be dispersed so that the particles are only separated by a few Angstrom from one another. At low volume fractions of fibers, there is a chaotic dispersion of fibers. The orientation of fibers is too random to promote the flow of current. At high volume fractions of fibers the population of fibers is just right to bring about uniform dispersion and facilitate the flow of current.

A plot of ε'' versus ε' has been presented in Fig. 8. This graph shows the Cole–Cole plots of various composites as a function of fiber loading. ε'' (Loss factor) and ε'' (Dielectric constant) are the real and imaginary parts of the complex impedance Z' respectively. The radius of semicircle gives an indication of electrical resistance of the composite. The Cole–Cole plot shows a peak for each case, which indicates a single relaxation mode and a resistive and capacitive network structure of the composite. The electrical conduction in sisal–oil palm hybrid fiber reinforced natural rubber composite would occur mainly through the polar lignocellulosic fibers as natural rubber is an insulator. This results in a single relaxation mode. It can also be seen that the addition of fibres has also led to the distortion of peak.

Dissipation factor

Dissipation factor or loss tangent is defined as the ratio of the electrical power dissipated in a material to the total power circulating in the circuit. The viscoelastic nature of the polymer creates responses in the material to both mechanical and electrical stimuli. The measurements of dissipation factor (tan δ) and loss factor (E'') of an insulating material are important since the loss tangent is a measure of the electrical energy which is converted to heat in an insulator. This heat raises the insulator temperature and accelerated its deterioration. The two prime factors

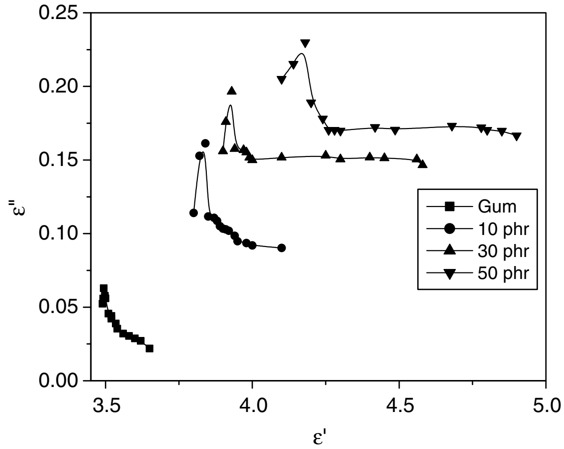


Fig. 8 Plot of loss factor against dielectric constant

contributing to loss factor (E'') are the dipole polarization and ionic conductance [8]. The numeric value of dissipation factor is determined by both polarity and carrier mobility. The polarity determines the nature of relaxation and the relaxation time determines the value of tan δ at a specific frequency for that relaxation.

The variation of dissipation factor with frequency as a function of fiber loading is presented in Fig. 9. It can be seen that dissipation factor increases with frequency and fiber loading. As fiber content increases, the polar groups that are present also increases which leads to an increment in orientation polarization. This causes dissipation factor to increase. Another reason is that natural rubber inherently possesses high strength due to strain induced crystallization. This crystallinity will result in lower dissipation of heat. The incorporation of fibers in natural rubber matrix disrupts the regular arrangement of rubber molecules and hence the ability for crystallization is lost. Again the addition of fibers enhances the flow of current through the amorphous region due to their ability to absorb moisture [24]. Therefore the addition of fibers resulted in higher loss or higher amount of dissipation that are associated with amorphous phase relaxations.

Figure 10 presents the variation of dissipation factor with frequency as a function of chemical modification. It can be seen that chemical treatment of fibers increases the dissipation factor. Also chemical modification does not affect the relaxation mechanism but increases the relaxation magnitude at each frequency. Plots of the reciprocal of dielectric constant ($1/\varepsilon'_c$) against volume fraction of fibre according to Fig. 10 are linear. Extrapolation of the plot to $V_1 = 0$ yields $1/\varepsilon'_c = 1/\varepsilon'_2$ where ε'_2 is the dielectric constant of rubber matrix at 4 Hz frequency. The graphically obtained value (3.73) is found to be in close agreement with the experimentally obtained value (3.65).

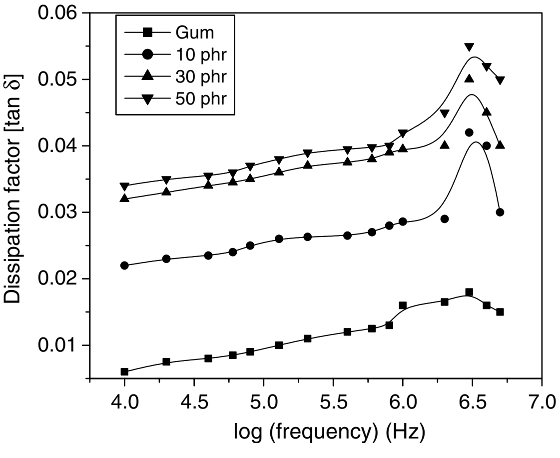


Fig. 9 Variation of dissipation factor with frequency as a function of fiber loading

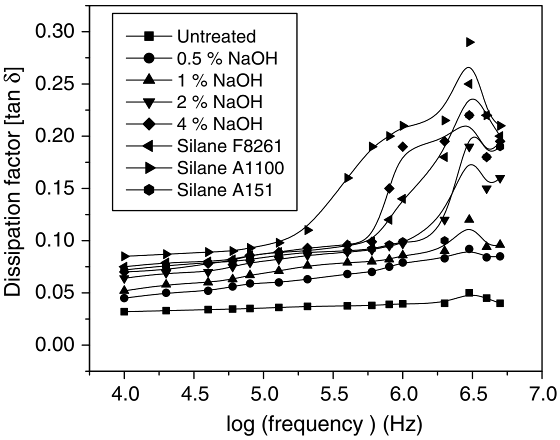


Fig. 10 Variation of dissipation factor with frequency as a function of chemical modifiication

Theoretical modeling of dielectric constant

Plots of the reciprocal of dielectric constant ($1/\varepsilon'_c$) against volume fraction of fibre according to Fig. 11 are linear. Extrapolation of the plot to $V_1 = 0$ yields $1/\varepsilon'_c = 1/\varepsilon'_2$ where ε'_2 is the dielectric constant of rubber matrix at 4 Hz frequency. The graphically obtained value (3.73) is found to be in close agreement with the experimentally obtained value (3.65).

Conclusions

The dielectric properties such as dielectric constant, volume resisitivity and dielectric loss factor of the composites were evaluated as a function of fiber loading, frequency and chemical modification of biofibers. The dielectric constant was seen to increase with fiber loading. This was

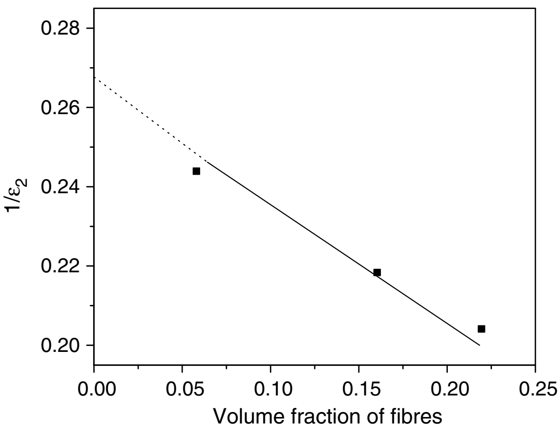


Fig. 11 Plot of reciprocal of dielectric constant as a function of volume fraction of fiber at frequency 4 Hz

attributed to the increase in orientation polarization of the polar groups present in lignocellulosic fibers. Chemical modification resulted in lowering of dielectric constant due to decrease of orientation polarization. The volume resistivities were seen to decrease with fiber loading and increase upon chemical modification. Ihis implies that incorporation of lignocellulosic fibers increased the conductivity of composites. The dissipation factor was seen to increase with loading which indicates that the electrical charges can be retained over a longer period of time. These cost effective biocomposites will therefore find its use in antistatic applications in order to dissipate static charges.

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