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Comparative Study of Dielectric Properties of Hybrid Natural Fiber Composites

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Abstract

The dielectric properties, such as dielectric constant, dissipation factor and dielectric loss factor of jute/bamboo natural fibers reinforced with polypropylene and unsaturated polyester hybrid composites were studied with different fiber loadings, fiber ratios, frequencies and chemical modifications of natural fibers. The dielectric constant, dissipation factor and loss factor increased with an increase in the fiber content for the entire range of frequencies than the pure polypropylene and pure unsaturated polyester materials. This increase was high at low frequencies, low at medium frequencies, and very low at high frequencies. Dielectric constant values were observed to decrease in frequency due to the decreased interfacial and orientation polarization at higher frequencies. Whereas, dielectric constant increased with an increase in the fiber loading, due to the increase in the number of polar groups, after the addition of hydrophilic lignocellulose fibers. Sodium hydroxide treatment was carried out on jute and bamboo fibers to improve the adhesion between fibers and matrix. The dielectric constant was lower for composites consisting of fibers subjected to alkaline treatment due to the increased hydrophobicity of fibers. When the weight percentage of jute fiber was increased in the total fiber content of the hybrid composites, the dielectric constant, dissipation factor and loss factor were found to increase. It is evident that types of polymer have little influence on the dielectric properties of the hybrid composites.

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1. Introduction

Polymers are commonly used in electrical and electronic industries as housings or assemblies. The most desirable combination of characteristics such as simplicity of fabrication, light weight, low cost, and excellent insulation properties have made plastics one of the most suitable materials for electrical and electronics applications. The function of plastics in electrical and electronic applications was limited to no-load bearing general-purpose applications. Fiber reinforced plastic materials not only act as effective insulators, but also provides mechanical support for field carrying conductors [1]. Nonetheless, many applications such as wire and cable sheathing and shielding against electromagnetic interference require the polymers to be made conductive in order to disperse electrostatic charges. This is done by the incorporation of conductive reinforcements into them. Fillers in the form of fibers and flakes have been found to be most effective for this purpose due to the lower volume fractions needed to achieve a targeted conductivity [2]. 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 and electronics applications. They can be used as connectors, terminals, industrial and household plugs, switches, and printed circuit boards.

Natural cellulose fibers have been frequently used as the reinforcement component in polymers to add the specific properties in the final product. With increasing awareness towards sustainable product design and the rising trends in its demand, natural based fiber materials are once again gaining popularity to replace synthetic fiber in the fabrication of composites [3]. Natural cellulose fibers from different bio-renewable resources have attracted the considerable attraction of research community all around the globe owing to their unique intrinsic properties such as biodegradability, environmental friendliness, easy availability, flexibility, easy processing and impressive physico-mechanical properties. Bio-composites are competitive with synthetic composites such as glass–polypropylene and glass–epoxies, and gaining attention over the last decade [4]. Natural plant fibers are renewable, non-abrasive and reduce health and safety concerns while handling. In addition, they show excellent specific mechanical properties, low cost, low density and enhanced energy recovery compared to traditional glass and carbon fibers. This favorable price-performance ratio, in combination with a marked environmentally friendly character, is crucial for their acceptance in large volume engineering markets [5]. However, natural fibers also have disadvantages, like inhomogeneous quality and supply cycles, poor water resistance and dimensional stability (swelling), as well as susceptibility to rot. These represent a major restriction in the successful use of natural fibers in durable composites not necessarily limited to interior and non-structural applications [6].

Natural fiber is highly hydrophilic due to its polarity owing to the free hydroxyl groups from cellulose and lignin. These hydroxyl groups can hold water molecules by hydrogen bonding. Cellulose is a hydrophilic glucan polymer consisting of a linear chain of 1, 4- β anhydro-glucose units. The hydroxyl groups in crystalline regions can form hydrogen bonds between parallel chains thereby reducing the water absorption. Natural fibers have poor water resistance, low durability, and poor fiber-matrix interactions. It is now well established that composite materials are sensitive to humidity through absorption of water leading to differential swelling between the fibers and the matrix [7]. The poor interfacial bonding between non-polar organophilic polymer matrix and highly polar natural fibers may result in undesirable mechanical and physical properties of the composites, which may discourage industrial usage of such composites. To complement this deficiency in compatibility, different strategies have been applied, including the use of surface modification techniques and/or coupling agents [8]. In the last two decades, many researchers focused on improving the interfacial adhesion by modifying the fiber surfaces via physical and chemical durability (moisture and temperature) and wear resistance of the composite [9]. Numerous surface treatments have been published in the literature such as electric discharge, alkalization, acetylation, peroxide, permanganate, benzoylation, silane and cyano-ethylation.

Aziz and Ansell [10] studied the effect of alkalization on mechanical and thermal properties of long kenaf fiber – cashew nut shell liquid (CNSL) resin composite. They showed that the alkalized fiber composites have higher storage modulus and lower loss factor, corresponding to higher flexural modulus and lower work of fracture. Although natural fiber–plastic composites (NFPCs) have been commercialized, their potential for use in many industries including automotive, decking, and electrical and electronics has been limited for their high brittleness, low impact resistance and mainly high density compared to the neat plastic. Therefore, most studies conducted in

this area focus on enhancing the mechanical and physical properties. One way to improve the mechanical properties may be done by producing hybrid composites, which involves a combination of various types of reinforcers with polymers. Each reinforcer has a feature that compensates for defects of other reinforcers [11]. The incorporation of several different types of fibers into a single matrix led to the development of hybrid composites. The behavior of hybrid composites is a weighted sum of the individual components in which there is a favorable balance between the inherent advantages and disadvantages. The properties of a hybrid composite mainly depend upon the fiber content, fiber-orientation, and length of individual fibers, extends of intermingling of fibers, arrangements of the fibers, and fiber-matrix bonding [12]. They offer a range of properties that cannot be obtained with a single kind of reinforcement.

By careful selection of reinforcing fibers, the material costs can be substantially reduced [13]. Although several fillers can be incorporated into the hybrid system, a combination of only two types of the fiber would be beneficial. By carefully selecting reinforcing fibers, the mechanical properties of the composite may be improved significantly, while substantially reducing the cost of material. Hybrid composite materials provide a good balance between the cost of the composite and the performance properties that cannot be obtained with a single kind of reinforcement [14]. In the past few years, the natural fiber reinforced composite materials have received a significant amount of attention in the automotive, construction and packaging industries. However, their electric applications have become popular only recently. The natural fiber reinforced composite materials have been used as dielectric materials in microchips, parts of transformers, terminal, connectors, switches, circuit boards, etc. Therefore, studies of dielectric properties of natural fibers reinforced unsaturated polyester and jute/bamboo reinforced polypropylene hybrid composites. The surface modification of these fibers was carried out by using sodium hydroxide (NaOH). The dielectric properties of the composites have been analyzed with special reference to effects of fiber loading, fiber ratio and chemical treatment of fibers.

2. Experimental

2.1. Materials

Two natural fibers jute and bamboo were used in this study. The information on chemical composition and physico-mechanical properties reported in the literature for fibers is summarized in Table 1 and Table 2.

Table 1. Chemical composition of selected fibers	[8]].
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Fibers	Cellulose (wt.%)	Hemicellulose (wt.%)	Lignin (wt.%) Ash (wt.%)		Equilibrium Moisture content (wt.%)	
Bamboo	26 – 65	30	5 - 31	1.3 – 2.0	8.9	
Jute	59 - 71.5	13.6 - 20.4	11.8 – 13	2	12	

Table 2. Physico-mechanical properties of selected fibers [8].

Fibers	Diameter (µm)	Length (mm)	Tensile strength (MPa)	Tensile Modulus (GPa)	Elongation at break (%)	Density (g/cm ³)
Bamboo	25 - 40	1.5 - 4	140 - 800	11 - 32	2.5 -3.7	0.6 - 1.1
Jute	20 - 200	1.5 - 120	320 - 800	8 - 78	1-1.8	1.3 - 1.49

Jute is the common name given to the fiber extracted from the stems of plants belonging to the genus *corchorus*, family *tiliaceae*. About 40 species of *corchorus* are known throughout the world, but *c.capsularis* (white jute) and *c.olitorius* (tossa jute) are the ones that are cultivated for their fiber. Bamboo (*bambusa shrep*) is a perennial plant. In monsoon climates, bamboo can grow up to 40 m in height. In order to extract bamboo fibers from raw bamboo trees, steam explosion technique was used. The experimental results showed that the bamboo fibers had the necessary specific strength that was comparable to that of glass fibers [16]. All raw materials were washed with water to remove impurities and then dried in an oven at 75°C for 12 hours. Dried materials were then ground and screened. The samples with the particle size between 20 and 40 meshes were collected for specimen preparation. One of the polymers used in this research is commercially available unsaturated polyester resin with the trade name

of "Reversol P9509" supplied by Revertex Malaysia Sdn. Bhd. Company. This type of resin has high rigidity, low reactivity, and thixotropic general purpose othophthalic characteristics. The matrix needs to be mixed with a curing catalyst, namely methyl ethyl ketone peroxide (MEKP) with the concentration of 1wt% by weight ratio of the matrix.

Polypropylene pellets with a density 1.145 g/cc and a melt flow index of 2.9 dg/min at 190^oC was supplied by Polypropylene Malaysia Sdn. Bhd. The primary drawback of using natural fibers is the low process temperature permissible due to the possibility of lignocelluloses degradation. The processing temperatures are thus limited to about 190^oC although it is possible to use higher temperatures for short periods. Additives are used to improve the performance and production of composites. Coupling agents are essential ingredients when fabricating natural fiber reinforced composites, because of the incompatibility between the hydrophilic natural fibers and the hydrophobic thermoplastic matrix. In this research we used maleic anhydride grafted polypropylene (MAPP) as coupling agent to improve the bonding between jute/bamboo fiber and polypropylene hybrid composites.

2.2. Surface modification of natural fibers

Mercerization or alkaline treatment is one of the most commonly used chemical treatments of natural fibers when used to reinforce thermosets and thermoplastics. The disruption of hydrogen bonding in the network structure is the important modification caused by alkaline treatment. This treatment leads to higher surface roughness. Additionally, a certain amount of oils, lignin and wax covering the external surface of the fiber cell wall, were removed by this treatment, which depolymerizes cellulose and exposes the short length crystallites [17]. Jute and bamboo fibers were treated using sodium hydroxide with 5wt% concentration and were soaked in the solution for 48 hours at 25°C. They were then washed 5 times with distilled water and oven dried at 80°C for 24 hours.

2.3. Preparation of composites specimens

In this research, hot press method was used to fabricate polypropylene reinforced with jute/bamboo hybrid fiber composites and cold press was used to fabricate unsaturated polyester reinforced with jute/bamboo hybrid fiber composites. The composites were fabricated by varying the weight fraction of fibers, ranging from 10wt% to 30wt%. The polypropylene, jute/bamboo and coupling agent (maleic anhydride grafted polypropylene) were weighted to the required proportion. The polypropylene, jute/bamboo fiber and coupling agents are mixed and loaded into the disc shaped mold with a diameter of 50 mm and a thickness of 5 mm. The hot press was pre-set at 190°C and at a pressure 6.8 MPa.

The mold was placed between the lower and the upper jaws of the hot press and compressed for about 30 minutes. After 30 minutes the mold was cooled down using water as the coolant. Cold press was used to fabricate jute/bamboo hybrid fibers reinforced unsaturated polyester composites. The known weight of unsaturated polyester resin was mixed with 1wt% of MEKP catalyst to initiate polymerization of polyester resin and stirred thoroughly. The known weights of jute/bamboo fibers were mixed with a solution of catalyst and polyester resin. The mixture was then placed in the mold, making sure that the mold cavity was properly filled and sufficient pressure was applied. The composite was then left to cure for about 24 hours at room temperature. The mold was later opened to remove the composites.

2.4. Testing of composites specimens

The dielectric properties of the composite specimens were measured with an HP Impedance Analyzer E4980A. The dielectric constant measurement program assists in carrying out the measurements using the Agilent E4980A precision LCR meter and the 16451B dielectric test fixture. The LCR meter is capable of measuring up to a frequency of 2MHz. The disc shaped samples had a diameter of 50 mm and a thickness of about 5mm. The specimens were analyzed using a contacting electrode method, which uses a rigid metal electrode. The measurements were done at varying frequencies ranging from 1 kHz to 1 MHz in accordance with ASTM D-150-10 standard. All specimens were dried in a vacuum-chamber at 70°C for 24 hours before the tests. All specimens were tested 10 times at a given frequency and then the average value was recorded by the HP impedance analyzer.

2.5. Dielectric properties

The dielectric constant of a material is defined as a measure of the material's ability to become polarized and to store charge when an external electric field is applied to it through parallel plates acting as a capacitor [13]. The dielectric constant of a material depends upon the polarizability of the material. If there is high polarizability of the molecule, dielectric constant will be high. The dielectric constant of a polymeric material depends on dipole, atomic, polarization, electronic and interface. The electronic and atomic polarizations are instantaneous polarization components, the effect of which is seen only at high frequencies. Due to the presence of polar groups in the material, the dipole or orientation polarization takes place [1]. It is also called relative permittivity and is denoted by symbol ε . Dielectric constant of an insulation material is an essential parameter since it provides valuable information about its dielectric strength.

This parameter helps in selection of dielectric capacitor material for the purpose of designing and manufacturing appropriate value of the capacitor to be used in capacitor banks for electrical power factor improvement of an electric system installation as it facilitates the reduction of energy losses in transmission lines [15]. The electrical loss or the amount of energy dissipated by the insulating material when the voltage is applied to the circuit can be represented by means of a dissipation factor. Dissipation factor (tan δ) is the ratio of the electrical power dissipated in a material to the total power circulating in the circuit. The measurement of tan δ of an insulating material is 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 accelerates its deterioration. The average power factor over a given period of time, is expressed as ($\hat{\epsilon}$), the loss factor. The loss factor ($\hat{\epsilon}$), is used in the energy industry to express the losses in distribution and transmission [18].

3. Results and Discussion

3.1. Effect of fiber loading on dielectric constant

Fig. 1 represents the influence of fiber loading on the dielectric constant of jute/bamboo reinforced polypropylene hybrid composites as a function of the logarithm of the frequency. It was observed that the dielectric constant increased with an increase in the fiber content over the entire range of frequencies. The lowest dielectric constants were exhibited by the pure matrix material. This was due to the absence of permanent dipoles, since the matrix was composed only of carbon and hydrogen atoms. For the composites, dielectric constant depended on the contributions of interface, orientation, atomic and electronic polarizations in the material.



Fig.1. Effect of fiber content on the dielectric constant values of hybrid composites as a function of logarithm of frequency at room temperature.

The interfacial polarization occurred due to the differences in conductivities or polarizations of the matrix and fibers [2]. Both, the orientation and the interfacial polarization of a composite depended on the concentration of fibers. In hybrid composites the significant increment in dielectric constant with increase in fiber loading was attributed to the increment in interfacial and orientation polarizations resulting from the presence of polar groups of cellulose in natural fibers. Jute (59 – 71.5 cellulose %) and bamboo (26 – 65 cellulose %) contributed to higher orientation and interface polarizations that led to increases in dielectric constant. This may be due to the hydrophilic nature of cellulose fibers which absorb moisture content from the air and thus causes increased conductivity of the polymer materials [15].

However, in unsaturated polyester, the dielectric constant was low since it had only instantaneous atomic and electronic polarizations. It was also found that for a given fiber loading, the dielectric constant showed high values at lower frequencies due to the decrease in the orientation polarization with increase in frequency. The complete orientation of the molecules is possible only at lower frequencies and the orientation polarizations. Therefore, as frequency increased, the dielectric constant reduced due to the lag in the orientation of polarization.

3.2. Effect of alkali treatment on dielectric constant

Fig. 2 represents the effect of alkaline treatment of natural fibers on the dielectric constant of the hybrid composites. It was observed that the treated fiber composites have a lower dielectric constant compared with the untreated ones for all frequencies. This was due to the decrease in orientation polarization as a result of the increased hydrophobicity of the treated fibers. Sodium hydroxide treatment of fibers also reduced the moisture absorption ability of the fibers and water molecules. In the untreated state, the cellulosic hydroxyl –OH groups of the fibers were relatively unreactive, since they formed strong hydrogen bonds. Sodium hydroxide treatment of fibers gets reduced by the treatment, which in turn decreases the orientation polarization [19]. Thus, the sodium hydroxide treated fibers.



Fig.2. Effect of chemical treatment on the dielectric constant values of hybrid composites as a function of logarithm of frequency.

3.3. Effect of hybridization on dielectric constant

Fig. 3 represents the dielectric constant as a function of the logarithm of frequency with different weight percentage of jute and bamboo fibers at a constant total fiber loading of 30wt%. It was observed that the composite system that contained only the bamboo fiber showed the lowest dielectric constant at all frequencies. It was also seen that the dielectric constant increased as the weight percentage of jute fiber increased in the total fiber content.



Fig.3. Effect of fiber ratio on dielectric constant values of hybrid composites as a function of logarithm frequency.

The composite which contained only jute fibers showed the highest dielectric value. Since both of these fibers, jute and bamboo are lignocellulosic in nature, they absorb moisture. However, the moisture absorption capacity of jute was 12wt%, which was higher than that of the bamboo fiber 8.9wt%. Therefore, the composite system, which contains high weight percentage of jute, showed higher dielectric constant values due to the presence of high moisture content in form of impurities at the interface.

3.4. Effect of fiber loading on dissipation factor

Fig.4 (a) and (b) represents the effect of different fiber loading on dissipation factor and loss factor as a function of the logarithm of the frequency. The dissipation factor was found to decrease with an increase in the frequency. When the fiber loading was increased, in the low frequency region, it showed a significant variation behavior of the result that was lowest for pure matrix material and highest for high fiber loading at a given frequency. However, in the higher frequency range, dissipation factor curves came closer, as seen in the Fig. 4. As the fiber content increased, the number of polar groups also increased, which led to an increment in orientation polarization [18]. This caused an increase in dissipation factor. Further, the addition of fibers enhanced the flow of current through the amorphous region due to their ability to absorb moisture.

In the high frequency region, orientation polarization of polar groups was difficult and hence the dissipation factor showed a significant decrease. It was seen that in the lower frequency region, the dielectric loss was high and was highest for the composites having a fiber content of 30wt% at a given frequency. However, the exact explanation for above behavior is somewhat difficult as dielectric loss or dissipation factor also depends upon fiber orientation [15]. Since fibers were inserted in the composite materials in a statistical random orientation manner, so

there may be significant variations in dielectric properties from region to region of the materials under investigation. Also water absorbance factor may also be responsible for the increase in conductivity as well as dielectric loss.



Fig.4. (a) Effect of fiber content on the dissipation factor of hybrid composites as a function of logarithm frequency.



Fig.4. (b) Effect of fiber content on the loss factor of hybrid composites as a function of logarithm frequency.

4. Conclusions

In the nutshell, the electrical properties of jute/bamboo reinforced polypropylene and unsaturated polyester hybrid composites were assessed. Dielectric constant values decreased with an increase in the frequency for all the hybrid composites. The maximum values of dielectric constant in the lower frequency region were attributed to the interfacial polarization. The dielectric constant, dissipation factor and loss factor increased with increasing fiber content because of an increase in the number of polar groups, which led to high orientation polarization. The increase in dielectric constant, dissipation factor and loss factor were more apparent with a fiber content of 30wt%.

As a result of chemical treatment of natural fibers, the interfacial adhesion between the polymer and natural fibers increased and led to a reduction in voids which in turn caused a reduction in moisture uptake. Chemical treatment also reduced the hydrophilic nature of jute and bamboo fibers. All these factors led to a reduction in dielectric constant values compared to untreated one. As the weight percentage of jute fiber increased in the composites, the dielectric constant also increased. Overall, the jute/bamboo reinforced unsaturated polyester hybrid composites exhibited higher values of dielectric properties compared with jute/bamboo reinforced polypropylene hybrid composites.

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