Mechanical Properties and Water Sorption Behavior of Phenol–Formaldehyde Hybrid Composites Reinforced with Banana Fiber and Glass Fiber

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ABSTRACT: Banana fiber, which is rich in cellulose, relatively inexpensive, and abundantly available, has potential for polymer reinforcement. This study explores the merits of combining high-modulus glass fibers with banana fiber in phenolic resoles to develop high-performance, cost-effective, lightweight hybrid composites. Of particular interest is the effect of varying layering patterns of banana fiber and glass fiber on the tensile, flexural, and impact properties of hybrid composites. The highest tensile strength value has been obtained for an intimate mixture of both fibers, and the maximum flexural and impact strength has been obtained for composite samples prepared from interleaving

layers of banana fiber and glass fiber. Tensile, flexural, and impact properties of the composites increase with an increasing volume fraction of glass fiber. The water uptake of these composites decreases with the incorporation of glass fiber into banana fiber, and the composites with glass fiber at the periphery and banana fiber at the core have the maximum resistance to water absorption. Scanning electron micrographs show the fracture mechanism and fiber/matrix adhesion in these composites © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 1439–1446, 2008

Key words: composites; fibers; mechanical properties

INTRODUCTION

The application of lignocellulosic material as a reinforcement component in polymer composites has received much attention because of its low price and specific weight. Lignocellulosic reinforcements offer several advantages with respect to inorganic ones, such as lower density, greater deformity, less abrasiveness to molds and mixing equipment, and low cost. Besides, lignocellulose-based reinforcements are derived from renewable resources and are biodegradable. Lignocellulosic natural fibers such as jute, coir, sisal sun hemp, sugarcane, and bagasse have received considerable attention as potential reinforcements in polymer composites.¹⁻⁶ Banana fiber, presently a waste product of banana cultivation, can be used as a reinforcement in phenolic resoles. Phenolic resins are well known for their excellent hightemperature resistance and fire tolerance combined with low smoke emission and reasonable price. A comparative study of the mechanical properties of banana-fiber/resole and glass-fiber/resole composites has already been done. The presence of methelol and phenolic OH groups in resoles makes them hydrophilic, and they can easily form hydrogen bonds with lignin and cellulose in natural fibers. Therefore, high compatibility is achieved between banana fiber and resole, which is absent in a glassfiber system.⁷ The major disadvantages of phenolics, such as mediocre mechanical properties and, in particular, brittleness, can be overcome by reinforcements. However, the mechanical properties and especially the impact strength are higher in glassfiber composites. Another problem to be met for natural-fiber composites is swelling of fibers due to moisture absorption, which continues until the cell wall is saturated with water. Beyond the saturation point, moisture exists as free water in the void structure, leading to composite defects such as delamination and void formation.

Investigations of lignocellulosic fibers have shown that the properties of fibers can be better used in hybrid composites.^{8–11} Clark and Ansell⁸ reported the improvement of various jute/glass hybrid laminates with different arrangements of jute and glass in laminates. Studies on sisal glass in polyester have shown a linear increase in the work of fracture with the variation of the volume fraction of glass at the core.⁹ Hybridization using glass fiber is an effective method for improving the mechanical properties of oil-palm-fiber-reinforced phenolics¹⁰ and bananafiber-reinforced polyester.¹¹ The degree of mechanical reinforcement that could be obtained by the

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introduction of glass fibers in biofiber (pineapple leaf fiber and sisal fiber)-reinforced polyester composites was assessed by Mishra et al.¹² The addition of a relatively small amount of glass fiber to the pineapple-leaf-fiber- and sisal-fiber-reinforced polyester matrix enhanced the mechanical properties of the resulting hybrid composites.

Plain-woven hybrid ramie-cotton fabrics were used as reinforcements in polyester matrix composites by Júnior et al.¹³ The tensile strength of the composites was shown to follow a common rule of mixtures, disregarding the contribution of the cotton fibers. Imielinska and Guillaumat¹⁴ subjected two different woven glass/aramid-fiber/epoxy laminates to water immersion aging followed by instrumented low-velocity impact testing. No important effect was found in the case of an aramid/glass-fiber configuration (woven hybrid fabric of glass and aramid fibers or aramid-fiber fabric interlaminated with layers of glass-fiber fabric) on moisture absorption and impact test characteristics. Recently, the properties of sisal/ oil-palm hybrid fiber reinforced natural rubber and sisal/banana hybrid fiber reinforced polyester composites were analyzed in our laboratory.15-17 Composites can be used for structural applications by the enhancement of their performance through hybridization.18,19

In this study, a detailed investigation was carried out to study the enhancement of the mechanical properties of banana-fiber-reinforced phenol-formaldehyde (PF) composites by the incorporation of glass fibers with different layering patterns and volume fractions. The objective of this work was to produce high-performance composite materials with banana fiber by the incorporation of glass fiber in small amounts (up to a volume fraction of 0.3%) while maintaining the cost effectiveness and improving the moisture resistance and mechanical properties of the banana-fiber composites.

EXPERIMENTAL

Banana fiber, obtained from Sheeba Fibers and Handicrafts (Poovancode, India), was used in this study. A PF resole-type resin, obtained from M/S West Coast Polymers Pvt., Ltd. (Kannur, India), was used as the matrix. Important characteristics of PF resin are given in Table I. The glass fiber used was E-glass rowing obtained from Hitech Fibers (Bangalore,

TABLE I Characteristics of PF Resole

Appearance	Deep brown color	
Viscosity (cps)	18–22	
Water tolerance	1:18	
Solid content (%)	50	

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TABLE II				
Tensile Properties of the Banana Fiber and Glass Fiber				

Fiber	Tensile strength (GPa)	Tensile modulus (GPa)	Elongation at break (%)
Banana fiber	0.5	12	7
Glass fiber	3	65	3

India). The physical and mechanical properties of the banana fiber and glass fiber are given in Table II.

Preparation of the composites

The prepreg route was followed for the preparation of the composites. The hand lay-up method followed by compression molding was adopted for composite fabrication. Mats of uniform thickness were prepared from chopped banana fibers (30 mm) and glass fibers (40 mm). The mats were impregnated in PF resin, and the prepreg was kept at room temperature up to the semicured stage. It was then pressed at 1000 C in a mold measuring $150 \times 150 \times 3 \text{ mm}^3$ to get a three-dimensionally crosslinked network. Different volume fractions of glass fiber were used for the preparation of the composites, the total volume fraction of fibers being kept constant (0.4). Samples with different layering patterns were also made, as shown in Figure 1.

Mechanical tests

Test specimens were cut from composite sheets. Tensile testing was carried out in an FIE TNE-500 universal tensile testing machine (Fuel Engineers, Mumbai, India) according to ASTM D 638-76. Five samples were tested in each set, and the average value is reported. The three-point flexural properties were determined with the same machine according to ASTM D 790. The load–displacement curves were obtained, and the flexural strength and modulus were calculated. The Izod impact test was performed on a notched specimen with an impact speed of 3.46 m/s and incident energy of 2.75 J. A minimum of four samples were tested in each case, and the average value is reported.

Water absorption studies

Samples of approximately $15 \times 3 \times 15 \text{ mm}^3$ (length \times width \times thickness) were cut from composite sheets for the measurement of water absorption. The specimens were immersed in distilled water at 25°C. The specimens were periodically taken out of water, surface-dried with absorbent paper, and reweighed. The water absorption was calculated with the following equation:

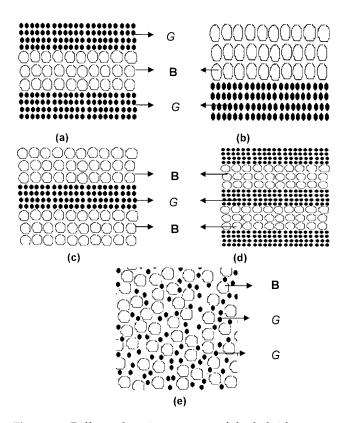


Figure 1 Different layering patterns of the hybrid composites: (a) GBG (trilayer), (b) BG (bilayer), (c) BGB (trilayer), (d) GBGBG, and (e) an intimate G/B mixture (G = glass; B = banana).

Water absorption (%) =
$$\frac{(M_2 - M_1)}{M_1} \times 100$$
 (1)

where M_1 is the mass of the sample before water absorption and M_2 is the mass of the water-absorbed composite.

RESULTS AND DISCUSSION

Mechanical properties

Effects of hybridization

Tensile properties. Figure 2 presents the tensile stressstrain behavior of banana/PF (with a volume fraction of 0.3) and banana/glass hybrid composites [with the total fiber volume fraction kept constant (0.3) and the glass-fiber volume fractions varied]. The general nature of the stress-strain curve is altered by an increase in the glass-fiber volume fraction. The stress-strain curves of the hybrid composites with different glass-fiber volume fractions show an inflection point, as observed by Summerscales.²⁰ As the content of the low-modulus fiber in the composite is greater than the critical content, an inflection occurs in the stress-strain curve, corresponding to the high-modulus glass fiber. The stress-strain curves show that the hybrid composites are more brittle than the banana-fiber composites.

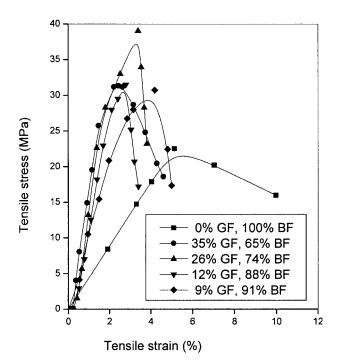


Figure 2 Tensile stress–strain behavior of banana-fiber (BF)/glass-fiber (GF) hybrid PF composites with the addition of different percentages of GF.

The variation of the tensile modulus and tensile strength of banana/PF composites with various glass-fiber volume fractions when the total volume fraction of the two fibers is kept constant is shown in Figure 3. The modulus values increase as the glass-fiber volume fraction is raised. Glass fiber has a greater tensile modulus than banana fiber, and the incorporation of high-modulus glass fiber increases the tensile modulus of the composite. The incorporation of even 0.09% glass fiber produces more than a 100% increase in the tensile modulus in comparison with a banana/PF composite of the same volume

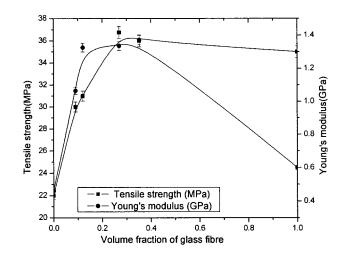


Figure 3 Variation of the tensile properties of the banana/glass hybrid PF composites with the addition of different percentages of glass fibers.

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 15kU
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 (1)
 (b)

Figure 4 Scanning electron micrographs of the tensile fracture surfaces of (a) glass/PF and (b) banana/PF composites.

fraction. The tensile strength of the samples increases linearly with the increase in the glass-fiber volume fraction. The occurrence of a hybrid effect, negative or positive, will depend on the relative volume fraction of the two fibers. An analysis of the fracture surface of samples subjected to tension reveals that the fracture of a sample is proceeded by the failure of glass fiber, the low-elongation component in the hybrid.²¹ As a result, the tensile strength of the hybrid composite uniformly increases with the glassfiber content. However, at a higher glass-fiber content, failure by delamination occurs, and the tensile strength shows a slight decrease. When the volume fraction of glass fiber is increased from 0.26 to 0.35, the tensile strength value shows a slight decrease. This is because at a higher volume fraction of glass fiber, fracture occurs in the composite, mainly by interlayer delamination.

Scanning electron micrographs of the tensile fracture surfaces of banana-fiber-reinforced PF composites and glass-fiber-reinforced composites are shown in Figure 4. As shown in Figure 4, there is very good adhesion in the banana/PF system, whereas the interaction between the glass fiber and PF resin is very poor. Therefore, the more prominent fracture mechanism in banana/PF composites is fiber fracture, whereas in the glass-fiber system, the fiber is pulled out of the matrix easily. Figure 5(a–c) shows the fracture surface of hybrid composites with 0.09, 0.12, or 3.5% glass fiber. In Figure 5(a), there is only a small amount of glass fiber, and the system seems to be like one having a single type of fiber. In Figure 5(b), which shows the composite with 0.12% glass fiber, the glass-fiber content is too low for interlayer delamination. However, as the glass-fiber content increases, the chance of delamination also increases. In Figure 5(c), in which the glass-fiber content is raised to 0.35%, layering out of the composite occurs, and the composite failure is mainly by interlayer delamination.

Flexural properties. Flexural strength is a combination of tensile and compressive strengths, which directly vary with the interlaminar shear strength. The flex-

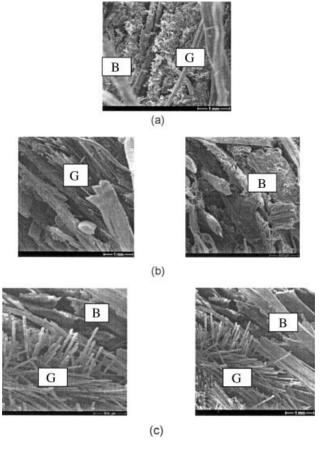


Figure 5 Scanning electron micrographs of the tensile fracture surfaces of banana (B)/glass (G) hybrid PF composites with the addition of glass fibers. The glass-fiber volume fractions were (a) 0.09, (b) 0.12, and (c) 0.35.

ural strength and modulus values of banana/PF composites with increasing glass-fiber concentration are shown in Figure 6. The flexural strength increases with an increasing amount of glass-fiber

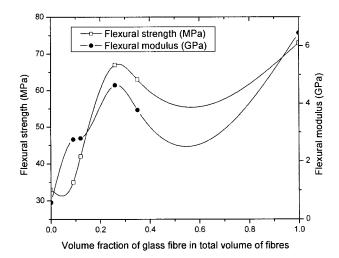


Figure 6 Influence of the glass-fiber volume fraction on the flexural properties of the banana/glass hybrid PF composites.

addition. Just like the tensile properties, the maximum values of the flexural strength and modulus are obtained with 0.26% glass fiber. A further increase in glass fiber to 0.35% reduces the flexural properties. This is due to higher interlayer delamination at a higher glass-fiber content, as is evident from scanning electron micrographs (Fig. 5).

Impact properties. The impact strength of fiber-reinforced composites depends on many factors, such as the nature of the constituents, fiber/matrix adhesion, construction and geometry of the composites, and test conditions. The energy-dissipation mechanism in fiber-reinforced composites is believed to be fiber pullout.²² The applied load, transferred by shear to the fibers, may exceed the fiber/matrix interfacial bond, and debonding may occur. The frictional force along the interface may transfer the stress to the debonded fiber. If the fiber stress level exceeds the fiber strength, fibers may fracture. The fractured fibers may be pulled out of the matrix, and this involves energy dissipation.²³ The variation of the impact strength of the banana-fiber composites with increasing glass-fiber volume fraction is shown in Figure 7. The impact strength of banana/glass hybrid composites increases with an increasing volume fraction of glass fiber. Misra et al.²⁴ observed a 34% increase in the impact strength by the addition of 8.5% glass fiber to sisal-fiber-reinforced polyester composites.²⁴ Because of the surface smoothness and regular cross section of glass fibers, they can be easily pulled out of the matrix, whereas in banana fibers, such a mechanism is not favored because of the mechanical interlocking between the fibers and matrix. The energy dissipated by fiber fracture is small in hybrid composites of banana fiber and glass fiber.

Effect of banana/glass layering

Tensile properties. Hybrid composites of various layering patterns were prepared by the total volume

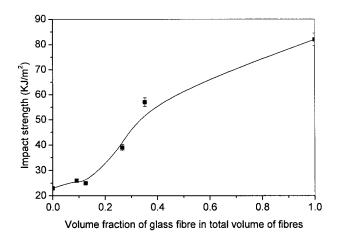


Figure 7 Variation of the impact strength with the glassfiber loading in the banana/glass hybrid PF composites.

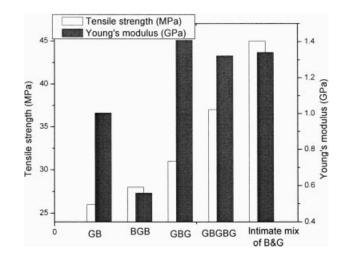


Figure 8 Tensile properties of the banana (B)/glass (G) hybrid composites with various layering patterns.

fraction of fibers in the composite being kept at 0.3 and the banana-fiber/glass-fiber ratio being kept at 0.7 : 0.3. Figure 8 shows the tensile strength values of the different layering patterns. Among these various layering patterns, the highest tensile strength is found in the intimate mixture of the two fibers. When banana fiber and glass fiber are intimately mixed, failure by delamination will occur with more difficulty because of the greater energy involved in creating a large amount of a new surface in an intimate mix versus that required to cause delamination of a layered hybrid. The tensile strength is 62.9% higher for the intimately mixed composite than for a bilayer composite of both fibers. Comparing the tensile strengths of the various layered hybrids, we find that the higher the number is of layers present, the higher the tensile strength is. The tensile modulus is maximum for the one having a higher number of layers present and minimum for a banana-glass-banana arrangement. This is based on the chances of delamination in hybrid composites.

The scanning electron micrographs of the tensile fracture surfaces of the hybrid composites with different layering patterns are shown in Figure 9. In the glass-banana and banana-glass-banana arrangement, there is interlayer delamination, as is clear from Figure 9(a,b). The chance for interlayer delamination decreases as the number of layers increases. However, the fracture mechanism in the intimately mixed composite is different. There is no chance for interlayer delamination here. In Figure 9(c), a crack initiates at the interface and propagates through the matrix to the fibers. Because of this stress concentration in the fiber/matrix interface, some of the fibers are pulled out of the matrix, and some others undergo fracture. These fractured fibers can also carry some load before the ultimate failure of the composite occurs. Therefore, the intimately mixed

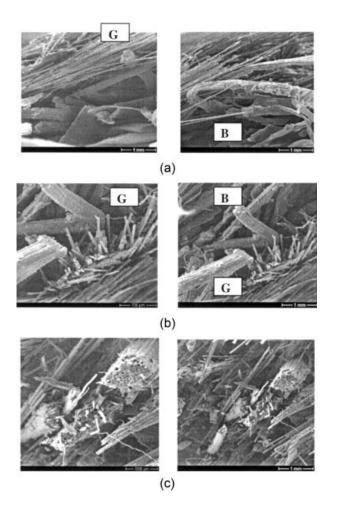


Figure 9 Scanning electron micrographs of the tensile fracture surfaces of the banana (B)/glass (G) hybrid PF composites (containing 0.26% glass fiber) with different layering patterns: (a) GB, (b) BGB, and (c) an intimate B/G mixture.

fibers can withstand stress to a greater extent than layered composites.

Flexural properties. The flexural strength and modulus of the composites with banana fiber and glass fiber in the ratio of 0.7 : 0.3 are given in Figure 10. The flexural strength is maximum in the composites in which glass and banana are arranged as interleaving layers. This is due to the small core thickness of each layer, which reduces the crack propagation. As the number of layers in the composites decreases, the flexural modulus shows a different trend because of the greater chance of interlayer delamination due to increased core thickness of individual layers in these composites. The modulus is maximum in the composites with glass fiber at the core and banana fiber at the periphery, and the minimum value is obtained for the one having only two layers.

Impact properties. Figure 11 shows that the layering pattern considerably depends on the impact strength of the fibers. The highest value of the impact strength is obtained when the banana fiber and glass

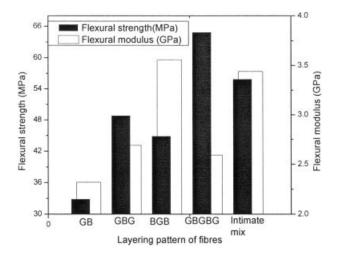


Figure 10 Flexural properties of the banana (B)/glass (G) hybrid composites with various layering patterns.

fiber are kept as interleaving layers because of the small core thickness. When a crack tip approaches a fiber, the crack crosses the fibers and cuts them as well as the matrix. Then, the crack changes its direction and moves through the matrix parallel to the fibers. The impact strength shows a decrease with a decreasing number of layers. Intimately mixed composites have the lowest impact strength. Harris and Bunssell²¹ also reported the inferior impact properties of intimately mixed hybrid composites due to a finer state of subdivisions.

Water absorption behavior. Banana fiber has 63–64% cellulose, 19% hemicellulose, and 5% lignin as its major constituents. Cellulose is hydrophilic because the large number of OH groups, which occur throughout the structure, can attract and hold water molecules by hydrogen bonding. The OH groups in the surface of the crystallites or in the amorphous

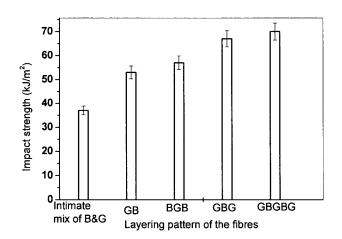


Figure 11 Influence of glass-fiber layering on the impact strength of the banana (B)/glass (G) hybrid PF composites (glass-fiber volume fraction = 0.26%) with different layering patterns.

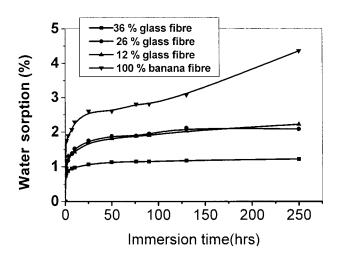


Figure 12 Water sorption behavior of the banana/glass hybrid composites with different volume fractions of glass fibers.

region may be available for bonding water, provided that they are not crosslinked with other OH groups.²⁵ Mainly, the water bonded by the amorphous region and the free water in the cellulose cavities are responsible for the changes in the mechanical properties. The dimensional stability and mechanical and electrical properties of composites reinforced by cellulose are affected by the moisture content of the cellulose.

Effects of hybridization

Figure 12 shows the effect of the incorporation of glass fiber on the water absorption of banana/PF composites. It is clear from Figure 12 that the maximum water uptake is shown by banana/PF composites (0% glass). The water enters through the interface and can diffuse through the porous structure of the fibers. The cross-sectional area of the composite becomes the main absorption face. The water penetration and diffusion are mainly through the fibermatrix interfacial region and cross-sectional portion of the fiber by a capillary mechanism. When glass fiber is incorporated, the water uptake is decreased, as the diffusion of water is not possible through glass fiber, as shown by banana/PF composites. The amount of water absorbed decreases with the glassfiber volume fraction increasing. All the hybrid composites exhibit a similar pattern of water absorption. Here an initial sharp uptake is followed by a gradual increase until the equilibrium water content is achieved. The water uptake percentage is found to be comparable in 0.26 and 0.12% glass-fiber-reinforced banana/PF composites. In the case of 0.36% glass-fiber-reinforced banana/PF composites, the moisture resistance is much higher. It is clear from Figure 12 that the incorporation of a small amount of glass fiber has increased the resistance of the banana/PF composites to water sorption very effectively.

Effects of the layering pattern

Figure 13 shows the effect of the layering pattern on the water sorption of banana/PF composites. Although the total volume fraction of fibers and the volume fraction of glass fiber in the hybrid composites are the same, the water absorption of these composites is different. The one having the glass fiber at the periphery and banana fiber at the core of the composite has the highest resistance to water absorption. This is because here water sorption is possible only through the cut portions of the composites, whereas in the intimate mix and banana-glassbanana arrangement, there are banana fibers at the periphery also, through which the water can easily penetrate the interior of the composites. The high value of water absorption in the glass-banana-glassbanana-glass arrangement is due to the easy absorption of water through the interlayers. There is very good adhesion between the banana fiber and PF resin, which is absent with glass fiber. There are a large number of voids in the regions between the banana/PF layer and glass/PF layer.

Theoretical modeling

Several theories have been proposed to model the tensile properties of composite materials in terms of different parameters. For determining the properties of randomly oriented fibers in a rigid matrix, the series model and Hirsch's model are useful. According to these models, the tensile strength is calculated with the following equations:

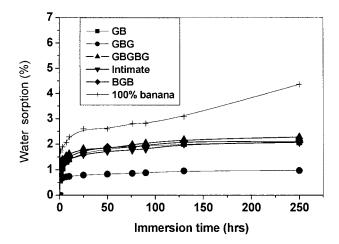


Figure 13 Water sorption behavior of the banana (B)/glass (G) hybrid composites with different fiber layering patterns.

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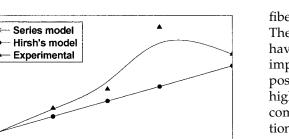
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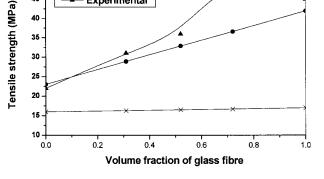


Figure 14 Experimental and theoretical tensile strength values of the banana/glass hybrid fiber reinforced PF composites.

Series model:

$$T_c = \frac{T_m T_f}{T_m V_f + T_f V_m} \tag{2}$$

Hirsch's model:

$$T_{c} = x(T_{m}V_{m} + T_{f}V_{f}) + (1 - x)\frac{T_{m}T_{f}}{T_{m}V_{f} + T_{f}V_{m}}$$
(3)

where T_{c_f} , T_{m_f} and T_f are the tensile strengths of the composite, matrix, and fiber, respectively. V_f and V_m are the volume fractions of the fiber and matrix, and x is a parameter between 0 and 1. It has been reported that parameter x in eq. (3) determines the stress transfer between the fiber and matrix.²⁵ For calculations, the value of x was varied to obtain the best fit values with experimental results.

The hybrid reinforcing effect of the two fibers was theoretically calculated. The law of the additive rule of mixtures was used to calculate the hybrid effect. The rule is given by

$$X_H = X_1 V_1 + X_2 V_2 (4)$$

where X_H is a characteristic property of the hybrid, X_1 and X_2 are characteristic properties of the individual composites, and V_1 and V_2 are the volume fractions of each reinforcement in the composite. Figure 14 shows the experimental and theoretical tensile strength values of the hybrid composites. The values obtained from the series model are lower than the experimental values. The experimentally obtained tensile strength values are found to fit Hirsh's model. A positive hybrid effect is observed in the hybrid composites.

CONCLUSIONS

The effect of hybridization with glass fibers on the tensile, flexural, and impact properties of bananafiber-reinforced PF composites have been studied. The tensile and flexural properties of the composites have been increased. The tensile, flexural, and impact properties of banana-fiber-reinforced PF composites have been improved by hybridization with high-modulus glass fibers. The properties of the composites increase with an increasing volume fraction of glass fiber. The water uptake of these composites decreases with the incorporation of glass fiber to banana fiber, and the composites with glass fiber at the periphery and banana fiber at the core have maximum resistance to water absorption. By the incorporation of glass fibers into banana-fiber/PF composites, we could develop composites with improved mechanical properties and decreased water absorption, thus removing the drawbacks of banana-fiber/PF composites, although it is difficult to recycle these hybrid composites.

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