Effect of Hybridization and Chemical Modification on the Water-Absorption Behavior of Banana Fiber–Reinforced Polyester Composites

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ABSTRACT: The water sorption characteristics of banana fiber–reinforced polyester composites were studied by immersion in distilled water at 28, 50, 70, and 90°C. The effect of hybridization with glass fiber and the chemical modification of the fiber on the water absorption properties of the prepared composites were also evaluated. In the case of hybrid composites, water uptake decreased with increase of glass fiber content. In the case of chemically modified fiber composites, water uptake was found to be dependent on the chemical treatment done on the fiber surface. Weight change profiles of the composites at higher temperature indicated that the diffusion is close to Fickian. The water absorption

showed a multistage mechanism in all cases at lower temperatures. Chemical modification was found to affect the water uptake of the composite. Among the treated composites the lowest water uptake was observed for composites treated with silane A1100. Finally, parameters like diffusion, sorption, and permeability coefficients were determined. It was observed that equilibrium water uptake is dependent on the nature of the composite and temperature. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 91: 3856–3865, 2004

Key words: banana fiber; diffusion; chemical modification; composites; glass fibers

INTRODUCTION

The plant cell wall is a highly versatile composite material that is the basis of products from wood and paper to paint thickeners and explosives. The meshwork of insoluble cellulose fibrils embedded in a matrix of gelatinous sugar polymer is responsible for the high tensile strength of the material. These lignocellulosic materials have slowly been introduced as reinforcement materials in polymeric matrices. These composites have appreciable mechanical properties, which make them good candidates in low-load-bearing applications.

However, for many applications, knowledge of the water absorption properties is important. Since the 1970s, tremendous efforts have been made to understand the mechanism of moisture absorption in polymers and composites and to improve their moisture resistance. Besides moisture, most applications also expose the material to a wide range of temperatures. As a result, the temperature effect of moisture absorption is also an important field of investigation. Natural fibers are potentially hydrophilic because of the presence of –OH groups. This hydrophilicity in turn affects the long-term mechanical properties of the composite,

when natural fibers are used as reinforcement in various polymeric matrices. Moisture diffuses into polymers to different degrees depending on a number of molecular and microstructural aspects.¹ The main factors that affect the diffusion process are as follows:

- 1. Polarity of the molecular structure, that is, the presence of chemical groups capable of forming hydrogen bonds with water.
- 2. Degree of crosslinking.
- 3. Presence of residual monomers or other waterattacking groups.
- 4. Crystallinity.

The water permeability of an overall composite, however, is decided mainly by the nature of the fibers.² Many matrix resins also absorb moisture reversibly by Fickian diffusion. Complexities involved in resin chemical structure and microstructure attributed to crosslinking density, polarity, and impurities cause non-Fickian processes to occur.³ The absorbed water causes plasticization of the matrix⁴ and also cracking through swelling.⁵ In addition, the diffused moisture may reduce the bond strength by breaking the bonds in the matrix. Incorporation of vegetable reinforcement in polymeric matrices aggravates the complexity of the situation. The polymeric matrix and the reinforcement differ significantly in their independent responses to moisture. Expressions relating the compos-

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 TABLE I

 Properties of Isophthalic Polyester

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 A clear pale liquid

Appearance	A clear pale liquid
Visocity at 25°C (cps)	
(Brookfield viscometer)	650
Specific gravity at 25°C	1.11
Typical properties of cured unreinfor	rced resin (specimens
cured for 24 h at room temperat	ture followed by
postcuring for 4 h at 80°C)	-
Tensile strength (psi)	9000
Flexural strength (psi)	16,000
Water absorption at 25°C (%),	
28 days	0.65
-	

ite diffusion coefficient to the fiber volume fraction and its orientation have been given by investigators like Shen and Springer.⁶ Rao et al.⁷ presented a comprehensive moisture absorption analysis in a jute– epoxy composite. They showed that a Fickian diffusion model is valid for this type of composite. In the case of Fickian diffusion, after a long period of time the *M* (moisture content) versus root *t* curves asymptotically approach the maximum moisture contents M_m . The initial slope of the curve is proportional to the moisture diffusivity, represented by

$$D = \pi \left(\frac{h}{4M_m}\right)^2 \left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}}\right)^2$$
(1)

where M_1 and M_2 represent the moisture content at temperatures t_1 and t_2 , respectively. In this study, composites made of jute and glass were found to have a reduction in moisture uptake from 25 to 2%. Hybridization of other natural fibers like oil palm empty fruit bunch fiber also showed a reduction in water uptake with the incorporation of glass fiber.⁸ The effectiveness of other natural fibers as reinforcement in various matrices has been proved from the extensive research work carried out by Thomas and coworkers in the field of natural fiber composites. Our earlier investigations proved banana fiber to be an excellent reinforcement in polymeric matrix.^{9–13} However, the water uptake of the composites incremented with the amount of fiber incorporated in the matrix. The present study focused on investigating the water uptake of composites prepared from chemically modified banana fibers as well as composites prepared from banana/glass hybrid fibers. The effect of the various chemical agents in controlling the water absorption properties of the composites was also investigated.

EXPERIMENTAL

Materials

Banana fiber obtained from Sheeba Fiber and Handicrafts (Poovancode, Tamil Nadu, India) was used in this study. Unsaturated polyester HSR 8131 (specific gravity: 1.12; viscosity: 65 cps; gel time: 25 min) obtained from M/s Bakelite Hylam (Hyderabad, India) was used as matrix. Ceat Ltd. (Hyderabad, India) supplied the multidirectional glass strand mat used for the study. Methyl ethyl ketone peroxide and cobalt naphthenate were of commercial grade supplied by Sharon Enterprises (Cochin, India). Table I gives the physical and chemical characteristics of the polyester resin used. Tables II and III give the characteristics of the banana fiber and glass fiber used. Figure 1 shows the layering pattern used in preparation of the composites.

Preparation of composites

Randomly oriented glass mats and neatly separated banana fiber cut at a uniform length of 30 mm were evenly arranged in a mold measuring $150 \times 150 \times 3$ mm in the required layering pattern for preparing the samples. Composite sheets were prepared by impregnating the fiber with the polyester resin to which 0.9 vol % cobalt naphthenate and 1% methyl ethyl ketone peroxide were added. The resin was degassed before pouring and the air bubbles were removed carefully with a roller. The closed mold was kept under pressure for 12 h; samples were postcured and test specimens of the required size were cut out from the sheets. Different volume fractions of glass were used for the preparation of samples as detailed in Table IV. In all these samples, glass was used as the core material. The

TABLE II Mechanical Properties of Banana Fiber

Sample	Diameter of fiber (µm)	Initial Young's modulus (GPa)	SD of initial Young's modulus (GPa)	Breaking strength (MPa)	Percentage strain
1	50	32	8.190	779	2
2	100	30	4	711	2
3	150	29	8	773	3
4	200	27	7	789	3
5	250	29	4	766	3

TABLE III Physical and Mechanical Properties of Glass Fiber

3	
Density (g/cm ³)	2.540
Tensile strength (MPa)	1.7–3.5
Elongation at break (%)	3
Young's modulus (Gpa)	65-72
Diameter (µm)	5–25
Microfibrillar angle	11°

density of banana fibers being known, the volume was calculated by dividing the weight of the fiber by the density. The ratio of the fibers to the matrix was calculated in each case by weighing the composite and subtracting the weight of fiber from that. The various silanes, silane A174, A151, A1100, and Si-69, were obtained from Sigma–Aldrich (Mumbai, India). Silane F8261 was obtained from ABCR GmbH and Co. (Germany). All the other chemicals were of commercial grade.

Silane treatment for banana fibers

A 0.6% sample of the respective silane was mixed with an ethanol/water mixture in the ratio 6:4, mixed well, and was allowed to stand for 1 h. The pH of the solution was maintained at 4 by adding acetic acid. Neatly separated and cut banana fiber was dipped in the above solution and allowed to remain there for 1.5 h. The ethanol/water mixture was drained out and the fiber was dried in air for 0.5 h followed by drying in an oven at 70°C until the fiber was fully dry.

The structures of the various chemical agents used are given in **Scheme 1**.

Treatment with NaOH

Cleaned and well-separated banana fibers were dipped in 0.5 and 1% solution of NaOH for 0.5 h and



 $SH = (CH_2)_2 = S = OCH_3 OCH_3 OCH_3$ $OCH_3 OCH_3$





then washed in very dilute acid to remove any particles of alkali. Washing was continued until the fibers were alkali free. The washed fibers were then dried in an oven at 70° C for 3 h.

Treatment with acetic anhydride

Acetylation of the fibers was done as follows. The fibers were dipped in glacial acetic acid for 30 min, the acid was drained, and the fibers were dipped in acetic anhydride containing a few drops of concentrated sulfuric acid for 5 min, washed in distilled water, and then dried.

Preparation of composites

Banana fibers were neatly separated by hand; adhering pith, if any, was removed and the fibers were cut to uniform size of 30 mm length. The fibers were then evenly arranged in a mold measuring $150 \times 150 \times 3$ mm³ and pressed into a mat. Composite sheets were prepared by impregnating the fiber with the polyester resin to which 0.9 vol % cobalt naphthanate and 1% methyl ethyl ketone peroxide were added and the air bubbles were removed carefully with a roller. The closed mold was kept under pressure for 12 h, samples were postcured, and test specimens of the required size were cut.

Testing techniques

Disc specimens (diameter: 20 mm; thickness: 2.5 mm) were used for studying the kinetics of water absorption. The specimens were immersed in distilled water and the percentage weight change was determined until the equilibrium values were reached. After immersion in water, samples were removed at different times, wiped with filter paper to remove surface wa-

TABLE IV Description of Composite Samples with Different Glass Volume Fractions (3 Layer)

Sample marking	Volume fraction of glass
В	0.07
С	0.11
D	0.15
Е	0.16
F	0.17

Scheme 1





ter, and weighed with an analytical balance with 0.1 mg resolution. The molar sorption Q_t of water by the composite at time *t* was calculated from

$$Q_t \,(\mathrm{mol\%}) = \frac{W_2 - W_1}{18W_1} \times 100$$
 (2)

where W_1 is the weight of the dry specimen and W_2 is the weight of the wet specimen. The molar sorption at equilibrium (infinite time) is represented by Q_{∞} . The weight gains of maximum moisture contents and diffusivities of such materials during immersion in distilled water were also estimated.

RESULTS AND DISCUSSION

Table IV shows the description of the various hybrid samples and Figure 2 shows the moisture absorption curves for various fiber volume fractions of bananaglass composites at room temperature. The water absorption curves show a multistage mechanism. In all the samples, glass was kept as the core material and banana at the periphery. The initial portion of the moisture absorption curve is linear, after which the mechanism changes. Water diffusion in polymers was found to lead to typical phenomena of composite swelling and physical relaxation. The absorption of water by polymer depends mainly on two factors: (1) the availability of free nanosized holes in the polymer and (2) the polar sites present in the polymer. There are two states of water molecules present in the polymer: (1) the unbonded molecules in the nanoholes of the polymer and (2) the water molecules that form hydrogen bonds with the polar groups. The dynamics of local stage motion of the sub- T_{q} relaxation process of the polymer appears to be the rate-determining factor for moisture transport. The rate of formation

and breakage of internal hydrogen bonds dictates the rate of moisture transport. Further, the water in the nanopores may form hydrogen bonds with the polymer, blocking the nanopores and reducing water uptake. Besides affecting the diffusivity and equilibrium uptake, temperature also changes the fundamental diffusion mechanism in a material. Although the rates of diffusion and relaxation are both sensitive to temperature, their temperature dependencies may be different.

According to Flory's two-stage theory the swelled polymer chains induce increased elasticity of chain structure and thus increase chemical potential. The increased chemical potential inhibits further absorption of water, which may be observed as the first equilibrium of water uptake. However, the swelled polymer chains start relaxing with time and subsequently reduce chemical potential. Consequently, the second equilibrium is attained by the decreased elasticity of the polymer chains. In the hybrid composites, in addition to the above phenomena, the change in mechanism can also be attributed to the delamination occurring in the composites. Penetration of water into the matrix causes absorption of water by the fibers as well. In the current case, because both glass and banana fibers are present, the rate of absorption of water is different for the two fibers. Glass fibers principally consist of silicates of various metals. When immersed in water, the nucleophilic attack of the OH⁻ at silicon takes place, with the formation of a transition metal complex followed by the breakage of the bonds, and formation of new bonds. In the case of banana fibers, hydrogen bonding through the -OH of the glucose is the principal water-absorption method. The absorption of water causes delamination of the two layers of the fiber attributed to the differential swelling. This causes further absorption of water into the free voids



Figure 3

created by delamination. The matrix resin also absorbs water. The chain-end –OH of the polyester, oxygen of the ester linkage, or residual cobalt ions are all sites for formation of the hydrogen bond.

In addition, based on the fiber–matrix interaction, the water diffusion occurs through the interface and from there to the bulk material. Transport of water also occurs through microcracks that occur on the surface of the composite and also through microchannels that occur inside the material because of defects.¹⁴

In the case of natural fibers, the diffusion of water has been reported to be anomalous.¹⁵ There are reports in the literature on the penetration of water through polyester by capillary effects through the matrix¹⁶ and wicking along the polymer/glass interface.¹⁷ Composites with higher glass content (0.16 and 0.17; i.e., where banana fiber content is low) were observed to give a two-step water absorption curve, whereas composites with glass volume fractions of 0.07, 0.11, and 0.15 were found to give a three-stage water absorption mechanism. The proportion of glass and banana in the two cases are different and possibly the cause for the difference in absorption behavior of the composites. The diffusion of water in a watercellulose system was also reported to be non-Fickian or anomalous by Newns¹⁸ and Stamm.¹⁹ Figure 3 shows the variation in equilibrium water content of the composites with increasing glass volume fraction, where the same geometry had been adopted. The response of the addition of impermeable fibers to permeable fiber composites is clear from the graph. The maximum water content decreases with increase in glass volume fraction.

The diffusion coefficient *D*, which is a predominantly matrix-dominated parameter, can be calculated from the following equation:

$$D = \pi \left(\frac{h\theta}{4Q_{\infty}}\right)^2 \tag{3}$$

where θ is the slope of the linear portion of the sorption curves and *h* is the initial sample thickness. The diffusion coefficient characterizes the ability of the water molecules to move among the polymer segments.

The diffusion coefficient value in the case of samples with different glass fiber volume fractions was considered. Table V shows the values of the diffusion coefficient of various hybrid samples at different temperatures. Because diffusion is a thermally activated process, an increase in temperature was found to increase

 TABLE V

 Diffusion Coefficient of the Various Hybrid Samples

Sample	Temperature (°C)	Diffusion coefficient $D (cm^2/s^{-1})$	Sorption coefficient S (g/g)	Permeability coefficient P (cm ² /min ⁻¹)
A	30	2.62400E-10	0.16	0.004
	50	1.95762E-09	0.28	0.040
	70	8.85359E-10	0.04	0.315
	90	1.09788E-10	0.35	0.004
В	30	5.11119E-10	0.24	0.012
	50	4.74454E-10	0.27	0.012
	70	1.95383E-09	0.31	0.06
	90	3.09162E-09	0.28	0.09
С	30	6.90319E-10	0.21	0.01
	50	5.12547E-10	0.21	0.01
	70	9.29134E-10	0.19	0.02
	90	5.94593E-09	0.28	0.17
D	30	6.26464E-10	0.22	0.01
	50	1.00002E-09	0.25	0.03
	70	1.65228E-09	0.26	0.04
	90	4.13954E-09	0.13	0.05
Е	30	6.87973E-12	0.14	0.00009
	50	1.00810E-09	0.15	0.02
	70	1.80815E-10	0.16	0.003
	90	6.80403E-09	0.26	0.17
F	30	8.48487E-12	0.13	0.0001
	50	4.91521E-09	0.14	0.07
	70	1.42897E-09	0.15	0.02
	90	1.12282E-08	0.14	0.15



the diffusion coefficient of the various samples. The diffusion coefficient value was found to be the lowest in the case of samples with high glass fiber content (0.16 and 0.17) at room temperature.

The percentage weight gains of the various samples at different temperatures are compared and shown in Figure 4. It is observed that the water uptake decreases with increasing temperature between 50 and 70°C. Interestingly, it increases again at 90°C. The reversed trend at 90°C suggests that additional diffusion mechanisms come into play at this temperature. Although moisture absorption below 70°C is controlled by the thermodynamics of simple diffusion, a critical temperature seems to exist between 70 and 90°C, beyond which additional mechanisms become active during the diffusion process. A similar observation in the case of other resins was reported by other researchers. The reason for the higher water uptake at 90°C can be attributed to the combined effect of the increased water uptake in the voids created by the degradation of the resin, as well as to resin cracking and fiber debonding. At 70°C, leaching of the material leads to chemical degradation, which results in a lowering of the observed net composite weight. Except in the case of samples with high banana fiber content, in all the other cases, the highest water uptake was at 90°C. The relatively lower value of water uptake in the case of composites with higher banana fiber content can be attributed to the dissolution of soluble materials like short-chain hemicellulose, pectin, and so forth from the banana fiber at higher temperatures. Because the relative banana fiber fraction is higher, the weight loss becomes prominent only in the aforementioned situation.

The optical photograph of the hybrid composite with a glass volume fraction of 0.13 is shown in Figure 5. The photograph clearly reveals the arrangement of the fibers. The decrease in moisture content for samples with high glass fiber content can be explained as being attributed to the role of the glass fibers as a barrier for preventing water diffusion.

The cellulose-water interaction depends strongly on the supramolecular structure of the specimen in question, and on factors (such as temperature) that influence the water structure. For example, from this study it is clear that the maximum water uptake of sample A at lower temperatures (i.e., 30, 50, and 70°C) is only 0.01. The water uptake values of the various composites at temperatures 30, 50, 70, and 90°C are given in Table VI. At 90°C, the maximum water uptake was increased to 0.09 because of the activation of the diffusion by temperature and also because of the delamination at high temperature, which promotes the diffusion of water. Delamination at higher temperature can be attributed to the different water-absorption capabilities of the two fibers. The increased absorption at high temperature also points to the fact that the curing reaction was over in the resin. Absorbed water into a rigid thermoset polymer-like polyester might induce a complex physical change like surface cracking or surface mass loss, leading to abnormal water absorption behaviors. In addition, typical phenomena of composite swelling and physical relaxation are known to occur by microscopic interactions between polymer chains and water molecules. All these lead to an abnormal equilibrium water uptake.

Figure 6 presents the schematic representation of the diffusion paths in permeable and impermeable fiber polymer composites. Figure 6(A) shows the diffusion path attributed to a permeable fiber composite and Figure 6(B) shows that attributed to an impermeable fiber composite. The permeability results are attributed to the natural fiber and the impermeability results are attributed to the glass fiber.

The diffusion coefficient values of the various hybrid composites are given in Table V. The diffusion



Figure 5 Optical photograph of the fractured surface of the hybrid composite.

		· ······		
Sample	Temperature (°C)	п	$k (g g^{-1} min^{-2})$	
A	30	0.143404	0.06478	
В	30	0.1787	0.08839	
С	30	0.188695	0.9808	
D	30	0.1905	0.09209	
E	30	0.17371	0.0566	
F	30	0.165577	0.0669	
А	50	0.185113	0.13887	
В	50	0.220219	0.17173	
С	50	0.223171	0.17272	
D	50	0.234426	0.1544	
Е	50	0.219825	0.13972	
F	50	0.212722	0.16736	
А	70	0.176111	0.10094	
В	70	0.298816	0.23278	
С	70	0.203483	0.12189	
D	70	0.257415	0.13765	
Е	70	0.223628	0.11531	
F	70	0.214415	0.12007	
А	90	0.384286	0.14527	
В	90	0.334402	0.30857	
С	90	0.260032	0.23662	
D	90	0.282213	0.2364	
Е	90	0.337602	0.27519	
F	90	0.283668	0.24383	

TABLE VI Values of n and k for the Various Hybrid Composities

coefficient *S* was found to be lowest for the neat polyester.

$$S = \frac{M_{\infty}}{M_p} \tag{4}$$

where M_{∞} and M_{p} represent the maximum moisture content and the initial composite weight, respectively. The value shows a steady increase with banana fiber content and the value was found to be the highest for samples with the maximum banana fiber content. Other investigators also reported a similar observation and the values are consistent with those of our previous studies on the water absorption of composites with different fiber loadings.²⁰ The sorption of water by the fiber determines the permeability of water molecules through the composite sample. The sorption coefficient of the composite was also calculated using eq. (4). The diffusion coefficient is related to the equilibrium sorption of the penetrant. The permeability P of the composite samples to water molecules can be expressed by²¹

$$P = DS \tag{5}$$

Permeability therefore treats the net effect of sorption and diffusion. Table VI gives the values of the permeability coefficient and sorption coefficient of the various hybrid composites. Development of microcracks on the surface and the bulk of the material as a result of the effect of high temperature and moisture environment can be given as the reason for the increase in water uptake. Peeling and surface dissolution of the composite result because of these crack developments.²²

$$\log\left(\frac{Q_t}{Q_{\infty}}\right) = \log k + n \log t \tag{6}$$

To understand the mechanism of sorption, the moisture uptake data of banana/polyester composites were fitted to eq. (6). In this equation, *n* and *k* give an idea about the mechanism of diffusion that takes place inside the composite. When the value of n = 0.5, the diffusion is Fickian and when n > 1, the diffusion is said to be anomalous. When the value of *n* is between 0.5 and 1, the diffusion is non-Fickian. It is interesting to note that the values of *n* and *k* increase with increase of temperature. The maximum value of *n* and *k* are obtained at 90°C. The increase of k at high temperature clearly shows the high extent of interaction of water molecules at high temperature. The low value of nclearly shows that the mechanism of transport deviates from Fickian. Table VII gives the value of *n* and *k* for the different hybrid composites.

Figure 7 shows the Q_t versus root t curve of the various chemically treated composites at room temperature. The water absorption curves show a two-stage mechanism in the case of all the treated samples. The equilibrium water absorption was found to be maximal in the case of acetylated fiber composites and minimal in the case of the silane A1100–treated fiber composite. The treatment with silane A1100 renders the surface more basic, as reported in our previous investigations.²⁴ The mechanism of water uptake, however, seems to be different in the case of untreated fiber composites. The fiber has a porous internal structure. The cross sections of the fibers become the main access to the penetrating water. Unlike organic penetrants, the water molecule is small and strongly asso-



Sample	Temperature (°C)	Diffusion coefficient $D (cm^2/s^{-1})$	Sorption coefficient S (g/g)	Permeability coefficient P (cm ² /min ⁻¹)
Untreated	30	0.123150	0.2261	0.0278
	50	0.6132	0.2431	0.1491
	70	0.052278	0.2388	0.01248
	90	0.19711	0.2982	0.0587
0.5% NaOH	30	0.1146	0.2117	0.0243
	50	0.1466	0.2321	0.0340
	70	0.055036	0.2250	0.01238
	90	0.2065	0.1811	0.0374
1% NaOH	30	0.1540	0.0726	0.0118
	50	0.175953	0.1948	0.0342
	70	0.20293	0.1637	0.0332
	90	0.43060	0.2322	0.0999
A174	30	0.03554	0.2155	0.00766
	50	0.0562	0.2404	0.0135
	70	0.2611	0.2382	0.6219
	90	0.1188	0.1879	0.0223
A151	30	0.09849	0.2066	0.0203
	50	0.1386	0.1944	0.0269
	70	0.0582	0.1679	0.0093
	90	0.65414	0.1846	0.1208
A1100	30	0.2438	0.1533	0.0374
	50	0.16573	0.1363	0.0226
	70	0.11208	0.1571	0.0176
	90	0.33426	0.3929	0.1313
F8261	30	0.1077	0.2282	0.0246
	50	0.6561	0.2429	0.1594
	70	0.8927	0.2419	0.2159
	90	0.24256	0.2154	0.0522
Acetylated	30	0.09274	0.2558	0.02372
-	50	0.3130	0.2115	0.0662
	70	0.10203	0.0172	0.00175
	90	0.13633	0.2392	0.0326

TABLE VII Diffusion Coefficent of the Various Treated Samples

ciated through hydrogen-bond formation. These water molecules form strong localized interactions with hydroxyl groups available on the different banana fiber constituents (i.e., cellulose, hemicellulose, and lignin). In the case of treated fibers, –OH groups are fewer in number because of the chemical interaction of the hydroxyl groups of banana fiber constituents with other moieties.



Figure 7





Alkali treatment has been found to reduce the polarity of other cellulose fibers.²³ The water absorption capacity was found to be lowered in the case of 1% alkali-treated fibers compared to 0.5% alkali-treated fibers. Increased alkali concentration induces greater crystallinity to the fibers, thus reducing the watersorption capacity of the fiber. The low water uptake of the alkali-treated fiber composite (Fig. 7) supports the improved mechanical property values of the composite, which results from the improved fiber/matrix adhesion. The effect of various silanes on the water absorption at 70°C is compared in Figure 8. Of the various silane-treated fiber composites, water absorption was found to be the highest for the silane F8261treated composite and the silane A174-treated composite. Silane F8261 is a fluorinated coupling agent and is water repellent; however, it leads to poor wetting between the fiber and the matrix, which invariably leads to water penetration stemming from the free voids present between the fiber and the matrix.

Our earlier studies showed that in the case of silane A174-treated fibers, the hydrogen bond donating acidity is the lowest compared to that of other silanetreated fibers.²⁴ Of the various chemically treated composites, the acetylated composites obtained the maximum water uptake. The zeta potential measurements of the alkali-treated fibers has shown that the fiber surface is relatively acidic. Interestingly, in all the cases, composites made of acidic fiber surfaces obtained the highest water uptake. Values of the diffusion coefficient calculated for the various composites were compared, of which the maximum value was for composites made from silane F8261-treated fibers at 50 and 70°C. However, at room temperature and at 90°C the value was much lower. The lowest value of the diffusion coefficient was for composites made out of silane A174-treated fiber, showing that after the initial absorption, other factors come into play in the

absorption phenomena. Except at 70°C the value was low in all the cases. The value of the diffusion coefficient is indicative of the diffusion barrier operating in the system. The diffusion coefficient was also relatively high in the case of untreated composites. The value of the maximum water uptake increased with increase of temperature in the case of treated-fiber composites: the highest value was in the case of silane F8261–treated-fiber composites.

Sorption and permeation coefficients of treated-fiber composites

The values of the sorption coefficient and permeation coefficient were calculated using eqs. (4) and (5). The values are given in Table VII. To understand the mechanism of sorption, the moisture uptake data of banana/polyester composites were fitted into eq. (5) to obtain n and k values. Table VIII gives the values of n and k for the various treated composites. The value of n clearly shows that the diffusion process deviates from the Fickian mechanism.

Shen and Springer⁶ reported that the saturation times are inversely related to the respective diffusion coefficients. The value of n was maximum at higher temperatures for the composites. In other words, the mechanism approaches Fickian behavior at higher temperatures. The value of *k* is indicative of the interaction of the polymer with water. The value of ksteadily increases with increase of temperature. This clearly indicates the improved interaction of the polymer with water. The increased temperature promotes improved molecular motion. The increased water uptake at higher temperature can be attributed to the relaxation mechanism in the polymer whereby the polymer network is distorted. This inevitably leads to large-scale segmental motion, allowing further penetration of water.

Composites				
Sample	Temperature (°C)	п	$k (g g^{-1} min^{-2})$	
Untreated	30	0.152549	0.07317	
0.5%NaOH	30	0.214643	0.12399	
1%NaOH	30	0.350391	0.07681	
A174	30	0.170779	0.13159	
A151	30	0.181335	0.09883	
A1100	30	0.146322	0.09629	
F8261	30	0.154619	0.08641	
Acetylated	30	0.143445	0.11562	
Untreated	50	0.065742	0.03981	
0.5%NaOH	50	0.206125	0.13291	
1%NaOH	50	0.179053	0.11523	
A174	50	0.231593	0.24472	
A151	50	0.158155	0.13725	
A1100	50	0.133577	0.09798	
F8261	50	0.13816	0.10607	
Acetylated	50	0.041618	0.042396	
Untreated	70	0.10800	0.01716	
0.5%NaOH	70	0.19772	0.10306	
1%NaOH	70	0.195325	0.12508	
A174	70	0.1300	0.06975	
A151	70	0.211091	0.14088	
A1100	70	0.1846	0.14016	
F8261	70	0.18002	0.1182	
Acetylated	70	0.135783	0.0827	
Untreated	90	0.201041	0.053598	
0.5%NaOH	90	0.283248	0.10103	
1%NaOH	90	0.261547	0.06067	
A174	90	0.127068	0.008702	
A151	90	0.301029	0.06645	
A1100	90	0.255269	0.10873	
F8261	90	0.142911	0.05394	
Acetylated	90	0.148673	0.05407	

TABLE VIIIValues of n and k for the Various Chemically Treated

CONCLUSIONS

The water diffusion of banana/glass hybrid composites was found to be dependent on the cellulose fiber and the temperature. At room temperature, the diffusion coefficient as well as the water uptake were found to be the lowest for composites with high glass fiber content. Chemical modification was found to affect the water absorption behavior of the composite. Composites made from fibers treated with various chemical agents as well as banana/glass hybrid fibers were analyzed for their water absorption behavior. The equilibrium water uptake was found to be minimum for the silane A151– and silane A174–treated fiber composites. The diffusion coefficient, obtained from the slope of the initial slope of the water absorption curve, was found to be highest in the case of the untreated and silane F8261–treated composites. The value of the equilibrium water uptake was found to increase with increase of temperature in the case of treated fiber composites. Among the treated fiber composites, the value was found to be the highest in the case of silane F8261–treated fiber composites. Diffusion studies revealed that the interfacial adhesion plays a vital role in the water-transport process. Finally, it is important to add that water transport studies could be used as a probe to evaluate the strength of the interfacial adhesion in banana fiber–reinforced polymer composites.

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