## Studies on Adsorptive Interaction Between Natural Fiber and Coupling Agents

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ABSTRACT: Efficacy evaluation of various coupling-agent-treated sisal fibers was made by contact angle measurements and Fourier transform infrared spectroscopy. It was found that high contact angle and reduced hydroxyl groups on titanate-treated fibers favor its better hydrophobicity over the other treatments. The presence of adsorbed layer of coupling agent on the fiber surface was ascertained by appearance, shifting, and decreased intensity of absorption bands. The lowest polar component of surfacefree energy for N-substituted methacrylamide-treated fiber indicates the formation of ordered layers of its organofunctionality at the surface. The reason for enhanced interaction between sisal fiber and N-substituted methacrylamide is suggested by the formation of hydrogen bond, besides extracting a surface-active proton from the fiber surface by alkoxy group to form a covalent bond. An optimum treating condition of fiber for effective adsorptive interaction has been reported. The deposition of compound in the form of an aggregate on the fiber surface was also observed under scanning electron microscopy. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 1847–1858, 1998

Key words: sisal fiber; coupling agent; contact angle; treating condition; adsorption

## **INTRODUCTION**

The importance of interface interactions has been widely recognized in improving the hygrothermal stability and the mechanical properties of natural fiber-reinforced polymer composites.<sup>1–3</sup> Several methods<sup>4–9</sup> have been reported to modify the surface of natural fibers, not only to reduce the moisture absorption, but also to offer an opportunity to improve interfacial adhesion between the fiber and the matrix. The potential advantage of using the coupling agent method is their natural attraction with both the natural fibers and the resin matrix during curing.<sup>8,9</sup> The effectiveness of these coupling agents in composites is known to be influenced by its physical state on the surface of fibers and fiber surface characteristics itself.<sup>10</sup>

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Undercured coupling agent layers may diffuse into the matrix resin to form an entangled network with resin, which lost a potential crosslinking site at the interface.<sup>11</sup> Under wet conditions, the most acute problem is the moderate stability of coupling agent–fiber adhesion to hydrolysis.<sup>12</sup> This situation justifies the necessity of developing hygrothermally stable bond by properly adjusting treating conditions and selection of specificity of a particular coupling agent.

In earlier studies, performance of coupling agents in natural fiber composites has been reported in terms of physicomechanical properties and fractographic events.<sup>9,13–15</sup> In these, it was shown that proper treatment applied to the fibers could result in a compatible surface with the resin matrix that improves the quality of the polymer composites. Mercerization prior to silane treatment was another effective means of advocating better property retention of composites exposed to moisture.<sup>8</sup> In the previous work, the adsorptive

interaction of coupling agent to the natural fibers and its detection has not received much attention; therefore, an understanding on this aspect will be essentially required to obtain a controlled interphase morphology in the development of this rapidly emerging material. Most of the earlier work described in the literature has been concerned mainly with the interaction of coupling agent on cellulose<sup>12,16,17</sup> and not with the natural fibers.

In the present study, we report experiments in which sisal fibers were modified with various coupling agents. The difference in the physicochemical nature of untreated and surface-modified fibers was studied by a dynamic contact analyzer and Fourier transform infrared (FTIR) and IR microanalysis. The effectiveness results obtained in these experiments and, also, in our earlier findings on sisal-polyester composites,<sup>13</sup> N-substituted methacrylamide was chosen, for further study. Advantages of being water-soluble and nontoxic, and having a low cost over other coupling agents (solvent-based) were the additional reasons in favor of their selection. In a subsequent experiment, adsorption of N-substituted methacrylamide on the fiber surface was performed under various treating conditions to obtain further information regarding fiber-coupling agent interactions.

### **EXPERIMENTAL**

#### Materials

Sisal fibers free from extractives were used in this study. Coupling agents, such as gamma-methacryloxy propyl trimethoxy silane–A-174 (Union Carbide, USA), neopentyl (diallyl) oxy, tri(dioctyl) pyro-phosphato titanate–LICA-38, neopentyl (diallyl) oxy triacryl zirconate–NZ-39, and N-substituted methacrylamide–QB-012 (Kenrich Petrochemicals Inc., USA) were used without further purification.

#### Surface Treatment Method

Sisal fibers were washed with distilled water to eliminate maximum water solubles in order to avoid its possible interference during fiber-coupling agent interaction. Prior to surface modification, the fibers were dried in an oven to remove moisture. In a typical experiment, a given amount of coupling agents was hydrolyzed for about 60 min and adjusted to a desired pH with acetic acid or sodium hydroxide. An appropriate amount of fibers was added to hydrolyzed coupling agents solution (6.6 g/100 mL). The fiber suspension was slowly stirred with a glass stirrer for 30 min. After treatment, the fibers were taken out allowing the excess of solution to be dipped off. These treated fibers were subsequently washed 2–3 times with the solvent and/or distilled water to remove the excess of loosely bonded physisorbed compounds attached to the fiber surface. The samples were kept at room temperature to a complete dryness and then subsequently placed in an air-circulating oven at 80°C for 4 h to complete the polymerization reaction.

The adsorption of N-substituted methacrylamide onto sisal fibers was carried out by varying fiber suspension pH in the range of 2-11. The treatment time was varied from 15-240 min. The concentration of solution were used between 0.2-3% (by weight of fibers), which covers the range usually practiced in the application of fiber reinforced plastics technology. The amount of coupling agent deposited on the fiber surface was measured with respect to the change in concentration of treating solution. For this purpose, a standard calibration curve between the known concentration of N-substituted methacrylamide, and pH was plotted. The reduced pH of fiber suspended solution was measured at regular periods. The adsorbed coupling agent onto sisal fibers was also determined by substracting the weight of dry fibers from the weight of treated fibers. The drying of treated fibers was done in an oven at different temperatures for various periods.

#### **Evaluation of Surface-Modified Fibers**

Single fiber tensiometer (Krusses K12 C GMBH) has been used to measure dynamic advancing contact angles of untreated and surface-treated fibers with the Wilhelmy technique using water and diiodomethane as probe liquids. The fiber is attached by one end to the hook of the electrobalance with an adhesive tape. Subsequently, it was slowly immersed and then emerged in the probe liquids (2 mm length). An increase in the weight of fiber is recorded to yield a dynamic advancing contact angle using the following Wilhelmy equation:

$$\cos \theta = \frac{F}{l. \sigma}$$

where  $\theta$  is the contact angle, F is the measured force, l is the wetted length, and  $\sigma$  is the surface

Surface Treatment	Fiber Diameter (mm)	Water a	s Probe Liquid	Diiodomethane as Probe Liquid	
		Contact Angle (°) (Advancing)	Work of Adhesion (J/m <sup>2</sup> )	Contact Angle (°) (Advancing)	Work of Adhesion (J/m <sup>2</sup> )
Untreated	0.595	54.80	1.14	68.20	0.69
Silane-treated	0.497	54.80	1.14	66.10	0.79
Zirconate-treated	0.599	72.20	0.95	62.50	0.74
Titanate-treated	0.532	76.80	0.89	71.30	0.66
N-substituted methacrylamide- treated	0.480	76.00	0.90	51.80	0.82

 Table I
 Contact Angle Data of Untreated and Surface-Treated Fibers

Surface tension of water, 72.8 mN/m; surface tension of diiodomethane, 50.80 mN/m.

tension of the liquid. The measurement was made at the elevator speed of 194  $\mu$ m/s, reducing the risk of fiber swelling. The dispersive and nondispersive components of surface-free energy of fibers were obtained from the advancing contact angle data. The average of five measurements was reported with standard deviations in contact angle values of ±3. A major advantage of the dynamic method is the reduction of the effect of local surface inhomogenities on the observed contact angles due to the averaging over a large part of the surface.<sup>18</sup>

FTIR (Perkin–Elmer 1760) purged with nitrogen gas was also employed to collect the spectra of fibers at a resolution of 4 cm<sup>-1</sup>. The untreated and treated fibers were cut into fine powder, ground, and ratioed against a KBr reference spectrum. IR microanalysis of untreated and treated fibers was also carried out using an IR microscope with a movable X–Y stage (Perkin–Elmer Spectrum 2000 Auto Image System). The size of tantalum aperture was kept at 100  $\mu$ m. Transmission electron microscope was used for this experiment. The spectra was recorded at 8  $cm^{-1}$  resolution with the accumulation of 1000 scans in ATR mode.

The surface topography of surface-modified fibers was performed on a scanning electron microscope (Phillips 501, Holland). The fiber surface was coated with a thin film of Au/Pd to render them conductive.

#### **RESULTS AND DISCUSSION**

#### Wettability Studies

The effectiveness of coupling agent treatments applied to the fiber surfaces are given in Tables I and II. It is evident that the advancing contact angle of water on the treated fibers exhibited higher values (except organosilane) over the control, whereas when using diiodomethane as a probe liquid, a decrease in contact angles was noticed with respect to untreated fibers (except organotitanate). These results demonstrate the

 Table II
 Surface Energetic Properties of Untreated and Surface-Treated Fibers

Treatment	$\begin{array}{c} Surface-Free \ Energy \\ (mJ \ m^{-2}) \end{array}$	$\begin{array}{c} Polar \ Component \\ (mJ \ m^{-2}) \end{array}$	Dispersive Component $(mJ \ m^{-2})$	
Untreated	48.30	25.90	22.40	
Silane-treated	48.60	25.30	23.30	
Zirconate-treated	39.50	14.30	25.20	
Titanate-treated	35.10	13.80	21.30	
N-substituted methacrylamide- treated	40.90	10.50	30.40	

variability in the physicochemical nature of fiber surfaces. The difference in wetting force is attributed mainly due to involvement of intermolecular interactions (secondary bonding) in constraining chain movements of adsorbed layers. In spite of the presence of polar components on the fiber surface, the long-chain organofunctionality of titanium and zirconium-containing compounds resulted in higher values for the contact angle, indicating even distribution of polar elements.<sup>19</sup> This would be an indicative of poor wetting and enhanced hydrophobicity of fibers. It is mentioned that the surface solubility of adsorbed layers leads to an incorporation of wetting liquid into the fiber surface and thus causes an increased surface wettability, resulting in higher values for  $\cos \theta$ .<sup>20</sup> The high contact angles of water and diiodomethane onto titanate-treated fibers are considered because of the existence of attached hydroxyl group to phosphorus atom in its organofunctionality contributing interactions with wetting liquids. It is also observed that the work of adhesion in water for untreated and treated fibers was higher than that in diiodomethane, probably due to their affinity towards water. However, it is not fair to draw a conclusion on the effectiveness of coupling agents at this time. It is reported that the same coupling agent may yield different values, depending on the orientation of the organofunctional groups. A typical example is that an amino-functional silane yields contact angle data similar to the hydrocarbon rather than the amine<sup>21</sup> due to a bridge-like structure.<sup>22</sup>

Determining the surface energetics, the confirmation of efficacy of treatments was further made (Table II). The surface-free energy of untreated and silane-treated fibers was almost similar. On the other hand, a lowering in the surface-free energy was observed when fiber surface was modified with titanate, zirconate, and N-substituted methacrylamide coupling agents. These modifications have changed the surface property of the fiber from the initial inorganic to organic in nature. The polar component of surface-free energy reduces significantly for all treatments except organosilane, whereas the dispersive component was increased marginally, except organotitanate. Similar results were also reported in the case of cellulose fibers.<sup>16</sup> Chain movements of adsorbed modifiers that could bury the polar groups away from the surface into bulk structure<sup>23</sup> favoured such behavior. It is also assumed that interchain hydrogen bonding could not prevent the effectively local as well as the long-range chain motion

necessary to move a modified layer of finite thickness. The reduced polar component of surface-free energy signifies the compatibility of fiber surfaces with liquid resin. The presence of polar groups on the surface tries to pull the whole molecules to the interface and orient themselves to the next phase, affecting surface wettability. It can be concluded that N-substituted methacrylamide-treated fiber was more favorable as compared to other treatments in terms of the spreading of the resin on the fiber surface (lower nondispersive surface-free energy). This data was consistent with earlier findings in assessing the efficacy in sisal-polyester systems.<sup>13</sup>

#### **FTIR Studies**

FTIR-attentuated total reflectance (ATR) microanalysis of untreated and treated sisal fibers are given in Figures 1 and 2. The degree of hydrophobicity of fiber was characterized by the availability of surface hydroxyl groups. Evidence for an intense absorption band at 3413 cm<sup>-1</sup> is an indicative of these groups. Introducing surface modifiers onto fiber, the intensity of hydroxyl groups was reduced due to its reactivity with alkoxy functional groups of surface modifiers. The magnitude of this reduction depends on the ability of coupling agents to either mask surface hydroxyl groups of fibers or extract proton from the surface by forming fiber—O—M linkage (M = Ti, Zr, and)Si). The reduced hydrophilicity of organotitanate over other treatments was mainly due to appearance of additional -OH stretching band (polymeric association,  $3085 \text{ cm}^{-1}$ ) at the expense of free hydroxyl groups. Further, a strong band of absorbed water in the region of  $1630-1645 \text{ cm}^{-1}$ was observed for both untreated and surfacetreated fibers (Fig. 2). Expectedly, the intensity of peak was reduced for all treatments, except organotitanate, which shows an intense band because of the attached hydroxyl group in the pyrophosphato organofunctionality. The confirmation of these free hydroxyl groups and/or absorbed water was further made by heat treatment at 110°C. A drastic reduction in the peak intensities near 3413 and 1630  $\text{cm}^{-1}$  was noticed, leaving other absorption bands unaffected (Fig. 3).

As a result of surface modification reactions, adsorbed coupling agent onto the fiber surface was evidenced by the change in peak intensity and the emergence of new peak assignments (Figs. 1 and 2). The treated fibers show the appearance of a shoulder near 2900 cm<sup>-1</sup> (interac-



**Figure 1** FTIR-ATR spectra of untreated and coupling-agent-treated sisal fibers (100  $\mu$ m aperture) showing displacement of the —OH group at 3413 cm<sup>-1</sup>.

tion of C-H bond with C=O) except silane; absorption bands at 1720 cm<sup>-1</sup> (C=O), except titanate; and absorption bands at  $1500 \text{ cm}^{-1}$ (>C=C<), except zirconate, and 1130 cm<sup>-1</sup> (M—O—C). The intense absorption bands at 1300 and 760 cm<sup>-1</sup> due to phosphorous-oxygen linkage and a weak doublet between 1550-1530 cm<sup>-1</sup> due to protonated amine (N–H bending vibration) were observed in the case of organotitanate and N-substituted methacrylamide, while zirconate and silane indicate an absorption band at 1070  $cm^{-1}$  (C—O stretching vibration) with respect to untreated fibers. The reduced peak intensities at 2922, 2853, 1742, 1400, 1340, 1225, and 1040 cm<sup>-1</sup> (only titanate) were attributed mainly to the deposition of coupling agents on the surface of the fibers. The shifting of the absorption band from  $1599 \text{ cm}^{-1}$  (for untreated) to  $1604-1613 \text{ cm}^{-1}$  (for treated fiber) is also noticed, showing the attachment of polymerized organofunctionality on the fiber surface (except silane).

The C=O stretching region for all treated fibers showed a reduced peak intensity at 1742

 $\mathrm{cm}^{-1}$  and the appearance of a shoulder near 1720  $cm^{-1}$  (Fig. 2). In the case of N-substituted methacrylamide, the absorption band at 1667  $\rm cm^{-1}$  has shifted to 1708  $\rm cm^{-1}$  upon dilution, showing the nonhydrogen-bonded C=O stretching mode of amide. On the other hand, the peak assignment near 1720  $\text{cm}^{-1}$  for zirconate and silane-coupling<sup>22</sup> agents is an indication of these compounds due to hydrogen-bonded carbonyl group. The slight shifting of the shoulder band towards a lower frequency  $(1700 \text{ cm}^{-1})$  for organosilane compared to the other treatments seems to indicate the possibility of conjugation with unsaturated groups along with a lesser degree of methacryl polymerization on the fiber surface. The precipitated oligomer is believed to be randomly oriented and is expected to give an absorption band near  $1720 \text{ cm}^{-1}$  due to the reaction of C=O groups and unreacted alkoxy groups of coupling agents. However, the lack of a distinguished peak and/or shoulder in this region could be explained by the interaction between >C==0group  $(1742 \text{ cm}^{-1})$  of fiber and >C=O stretching



**Figure 2** FTIR-ATR spectra of untreated and coupling-agent-treated sisal fiber (100  $\mu$ m aperture).

 $(1720 \text{ cm}^{-1})$  of organofunctionality of coupling agents. Since the mobility of the adsorbed coupling agent molecules is less than the molecules in the aqueous solution, irregular deposition would result in a significant amount of unreacted alkoxy groups.<sup>24</sup> These unreacted groups from the neighboring layers interact with one another, restricting the segmental motions of adsorbed layers. This phenomenon supports the variability of the surface-free energy of various treated fibers.

# N-Substituted Methacrylamide Adsorption onto Sisal Fibers

N-substituted methacrylamide is a water-soluble, quat-type-titanate prepared via quaternization with amines through the proton provided by the hydroxyl group of the pyrophosphato function. The interaction of N-substituted methacrylamide and the fiber surface can be seen in the FTIR spectra (Fig. 4). The spectra of pure liquid was also recorded for comparison purposes. The shifting of the amide band on higher frequency (carbonyl stretching) from 1667 to 1708  $\text{cm}^{-1}$  as also observed in FTIR-ATR microscopic analysis, and N–H stretching from 3316 to 3413  $\text{cm}^{-1}$ , may be considered only due to dilution of the compound. However, confirming the presence of traces of oligomer formation, the spectra of treated fibers sprinkled on KBr was further recorded in the diffuse reflectance mode (DRIFT). It was found that carbonyl stretching vibration of amide has shifted slightly from 1667 to 1659  $\text{cm}^{-1}$ , showing the presence of a hydrogen-bonded carbonyl group. The shifting of an unsaturated band from 1618 to 1597  $\text{cm}^{-1}$  also indicates its participation in the conjugation with the N-H band. The presence of adsorbed compound can be clearly evidenced in scanning electron microscopy (SEM) (Fig. 5). It is assumed that interaction of N-substituted methacrylamide with the fiber surface is expected to follow the extraction of the proton from the fiber by alkoxy groups of the compound



**Figure 3** Effect of heat treatment on the spectra of N-substituted methacrylamidetreated sisal fiber: (a) fresh and (b) at 110°C.

to form fiber—O—Ti linkage. The probability of participation of the amide group in forming hydrogen bonds with the cellulose portion of fiber is also assumed, besides little hydrophobic interaction between the methacryl groups and the lignocellulosics. The presence of the methyl<sup>25</sup> group decreases the coupling agent–water interaction, thereby increasing the adsorption properties. A probable scheme is proposed for fiber-coupling agent interaction (Fig. 6).

#### **Optimization of Treating Conditions**

Figure 7 shows the effect of pH on the adsorption of N-substituted methacrylamide coupling agent onto fiber surfaces. The most efficient treatment occurred at pH 9. The reason for the varying deposition is the stability of compound, which, in turn, is affected by water-soluble anionic substances originating from the fibers.<sup>26</sup> At low pH, the fibers might already be positively charged, while at high pH (>10), the coupling agent has become negatively charged. Thus, a repulsive force created due to reversal of charge of either treating the solution or fibers prevents the deposition. It was also noticed that at low pH, colloidal particles are still interacting with fibers that resulted in deposition but, to a lesser extent, as observed on pulp fibers.<sup>26</sup>

The deposition of coupling agent onto fiber surface increases up to 30 min and then levels off with a further increasing treatment time (Fig. 8). The concentration and pH of treating solution used were 1% (by wt of fiber) and 9, respectively. The increased deposition at a certain level indicates the high reactivity of alkoxy group with the surface-active proton of the fiber surface. Only short treatment times are required in this case for the coupling agent adsorption. The longer treatment time leads to poor efficiency due to the interference of leached extractives from fibers with the treating compound, thereby preventing its further deposition.

The effect of the coupling agent concentration on the adsorption is shown in Figure 9. These experiments were carried out at pH 9 with a treatment time of 30 min. The amount of deposition onto fiber surface increases at low concentrations and then rises slowly at higher concentrations. The fibers treated with a 0.5% coupling agent shows deposition in the form of an aggre-



**Figure 4** FTIR difference spectra of N-substituted methacrylamide-treated fiber and liquid coupling agent: (a) liquid coupling agent; (b) KBr pellet; (c) sprinkled fiber on KBr.

gate, while a uniform distribution was observed at a 1% treating concentration [Fig. 5(a-d)]. However, the positioning of the N-substituted methacrylamide-treated fiber, upon IR microscopic examination through an aperture technique shows heterogeneity in the availability of chemical species over the whole surface (Figs. 1 and 2). The existence of a waxy surface resulted in irregular deposition of compound on the fiber. This indicates that the optimum concentration of coupling agent and removal of waxy materials from the surface of fiber are necessary for its uniform and continuous distribution. At a higher concentration, a large number of molecules layered on the fiber surface was not optimally efficient due to more loosely bound physisorbed types. These loose layers were removed during washing, affecting minimal weight gain. Attachment of precipitated oligomer to the fiber surface in the form of large aggregates, as observed in SEM [Fig. 5(d)] is

attributed mainly to the formation of intramolecular hydrogen bonding between the alkoxy and amide group. It is also reported that amide groups, as follows,

$$\begin{array}{c} \text{OCH}_{3} \\ \parallel \\ \mathbb{R} - O \_ \text{Ti} - (OR^{1})_{3} \cdot nR_{2}^{2}NR^{3}NCC = CH_{2} \\ \downarrow \\ \text{I} \\$$

form hydrogen bonds with each other as a result of proton donor and proton acceptor properties of this group.<sup>27</sup> This reduces the availability of functionality for direct adsorption to the fiber surface. It can also be observed from Table III that the moisture content and water absorption of fiber were reduced significantly due to deposition of layers as compared to untreated fiber. This be-



**Figure 5** SEM of untreated and N-substituted methacrylamide treated fibers (by wt of fiber): (a) untreated; (b) 0.5%; (c) 1%; (d) 2%.

havior was further supported by wetting force measurement. The higher contact angles on treated fibers give an indication of enhanced hydrophobicity.

Curing of treated fibers was carried out at different times and temperatures (Fig. 10). At 60°C, the adsorbed layers were undercured and virtually unaffected with increasing drying time, while crosslinking in the deposited layers occurred at higher temperatures. At 110°C, the increased drying time reduces the amount of adsorbed layers by the loss of residual moisture and methacrylate functionality through oxidation or polymerization.<sup>24</sup> The optimum efficiency of treatment was obtained at 80°C for 4 h of drying.

#### **Composite Properties**

The effect of adsorbed coupling agents on the mechanical properties of sisal and polyester composites was reported in earlier articles.<sup>13–15</sup> An improvement of ~ 15–33% in tensile strength and ~ 21–29% in flexural strength was observed with

respect to untreated sisal composites. The improvements are thought mainly due to the responsibility of chemisorbed coupling agent layers on the fiber surface involved in the chemical bond formation with unsaturated polyester resin.<sup>15</sup> As a result, enhanced adhesion and good wetting between surface-modified fibers and the resin matrix have been obtained. However, the role of silane coupling agent is restricted only to processing benefits by the way of working as a lubricant, as evident in the minimal effect on wetting and spectroscopic data discussed earlier. On the contrary, the physisorbed coupling agents onto fiber surface could have a different role from the chemisorbed layers, forming the copolymer between the physisorbed layer and the resin matrix at the fiber-matrix interface. It was reported that higher the content of physisorbed coupling agent layers on the fiber, the lower the strength of composites.<sup>28</sup> Under wet conditions, a decrease of 30-44% in tensile strength and 50-70% in flexural strength has been observed. However, the



Figure 6 Schematic representation of N-substituted methacrylamide-sisal fiber interaction.

strength retention of surface-treated composites (except silane) is higher than those of untreated composites. It is thought that surface modifica-



**Figure 7** Effect of pH on the adsorption of N-substituted methacrylamide onto sisal fiber (initial concentration of treating solution, 50 mg/g; treatment time, 30 min).

tion of fiber caused a comparably better hydrophobic interphase. It is noticed that N-substituted methacrylamide-treated fiber composites exhibited better properties under both dry as well as wet conditions, as compared to other treated fiber-composites.<sup>14,15</sup> These findings are also supported by the fractographic evidence, such as fiber pull-out and/or fracture, fiber swelling and/or



**Figure 8** Variation of polymer adsorbed as a function of treatment time (pH 9; concentrations of treating solution, 50 mg/g).





Figure 9 Adsorption of N-substituted methacrylamide onto sisal fiber at different concentrations of treating solution (pH 9; treatment time, 30 min).

splitting, fiber pull-out clean surface and/or matrix adherence.

#### CONCLUSIONS

80

Treating sisal fibers by water-soluble N-substituted methacrylamide resulted in a significant decrease of nondispersive component of surfacefree energy, thus with better spreading of resin on the fiber surface as compared to other treatments.



Figure 10 Retention of adsorbed N-substituted methacrylamide on the fiber surface at different drying times and temperatures (pH 9; initial concentration of treating solution, 50 mg/g; treatment time, 30 min).

Heterogeneity of the compound onto fiber, as observed in IR microscopic examination, may diminish the overall performance of composites while using sisal fiber as reinforcement in the polymer matrices. The effective results could be obtained only by regular deposition of adsorbed layers under properly adjusted treating conditions, such as pH, concentration of treating solutions, treatment time, drying time, and temperature. It is thus concluded that surface treatment of sisal is necessary prior to introducing in polymer matrices for making building materials with improved performance.

Table III Properties of Sisal Fibers Treated with Different Percentages of N-Substituted Methacrylamide (Using Water as the Probe Liquid for Contact Angle Data)

Concentration (1% by Wt of Fiber)	Amount Adsorbed (mg/g)	Moisture Content (%)	Water Absorption (%)	Contact Angle (°) (Advancing)	Work of Adhesion (mJ m <sup>-2</sup> )
Control	_	12.77	123	54.80	1.14
0.2	25.10	5.44	7.59	76.12	0.90
0.5	39.90	4.21	7.69	72.78	0.94
1.0	48.10	2.25	7.88	76.00	0.90
2.0	54.50	4.14	3.94	76.75	0.89
3.0	58.80	5.70	6.75	77.98	0.87

Surface tension of water, 72.8 mN/m.

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