Effect of Alkali Treatment on the Flexural Properties of *Hildegardia* Fabric Composites

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Received 24 March 2005; accepted 15 August 2005
DOI 10.1002/app.23522
Publication online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: A uniaxial natural fabric of *Hildegardia populifolia* was treated with 5% sodium hydroxide solution for 1 h, and the resulting changes were analyzed by polarized and scanning electron microscopic techniques. The untreated and treated *H. populifolia* fabric was reinforced in epoxy and toughened with 10% polycarbonate. The variation of the flexural strength and flexural modulus with different fabric contents and fiber orienrations was studied. The effect of sodium hydroxide and a silane coupling agent on the flexural properties of the composite was also studied. It was observed that the flexural properties increased on alkali treatment and when the coupling agent was used. The morphology of the cryogenically fractured surfaces indicated good bonding between the matrix and the reinforcement when a coupling agent was used. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 102: 1297–1302, 2006

Key words: natural fabric; epoxy resin; polycarbonate; blend matrix; flexural properties; morphology; alkali treatment

INTRODUCTION

Natural fibers as reinforcement have advantages such as low cost, high strength, environmentally friendly nature, and easy dispersion in polymer composites. They are also biodegradable. In the last decade there has been a renewed interest in natural fibers as a substitute for glass, carbon, and synthetic fibers, because natural fibers are less dense and cheaper than conventional fibers, and may be easily recycled. At present, the trend is slowly changing towards using natural fibers as reinforcements. Composites made with natural fiber reinforcements, known as "green composites," were developed by several workers[1–10] using sisal, banana, bamboo, coir, pineapple leaf fiber, and so forth. However, being hydrophilic, natural fibers need to be treated first to make them more compatible with hydrophobic thermosets and thermoplastics. Several researchers reported improvement in mechanical properties of cellulose fibers when alkalized at different NaOH concentrations. Bisanda and Ansell[11] applied an aqueous 0.5*N* NaOH solution on sisal fiber, while Sreekala and coworkers[12] and Geethamma and coworkers[13] used 5% NaOH to remove surface impurities on oil palm fibers and short coir fibers, respectively. Mwaikambo and Ansell[14] treated hemp, jute, sisal, and kapok fibers with various concentrations of NaOH. Varada Rajulu et al.[15,16] studied the properties and effect of alkali treatment of the lignocellulosic natural fabric *Hildegardia populifolia*. Recently, Babu Rao[17] developed *H. populifolia* fabric/polycarbonate-toughened epoxy composites and studied their performance. He reported that the mechanical properties of the matrix improved substantially on reinforcing it with this natural fabric. Roy and Sarkar[18] studied the physical and mechanical properties of jute fibers after alkali treatment. They reported improvement in these properties after alkali treatment. Hill and Abdul Khalil[19] studied the effect of acetylation on the mechanical properties of coir and oil palm fiber-reinforced polyester composites. They reported an increase in interfacial shear strength between the fiber and the matrix after acetylation of coir. Mannan and Munir[20] characterized jute fibers treated with soap–glycerol micelles. They reported improvement of the mechanical properties after treatment. In the present work, we studied the effect of alkali treatment, coupling agent, fiber content, and orientation of the fabric on the flexural properties of the natural fabric *H. populifolia*-reinforced polycarbonate-toughened epoxy composites.

EXPERIMENTAL

Materials

The epoxy resin Araldite LY-556 and hardener HY-951 (M/s Hindustan Ciba-Geigy) and polycarbonate (Cal-

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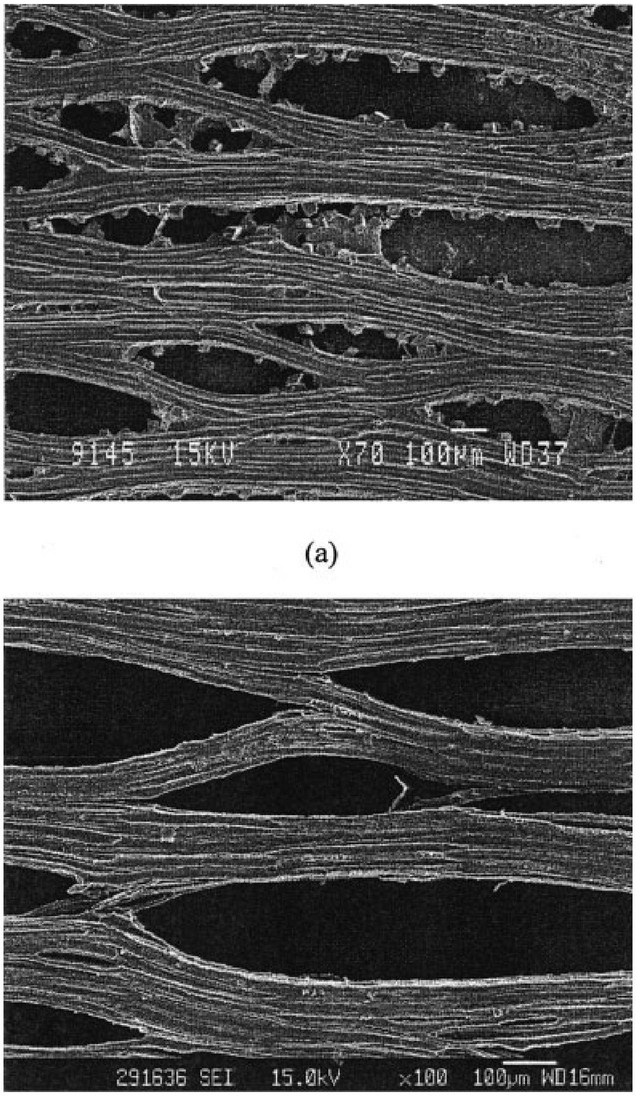


(a)

(b)

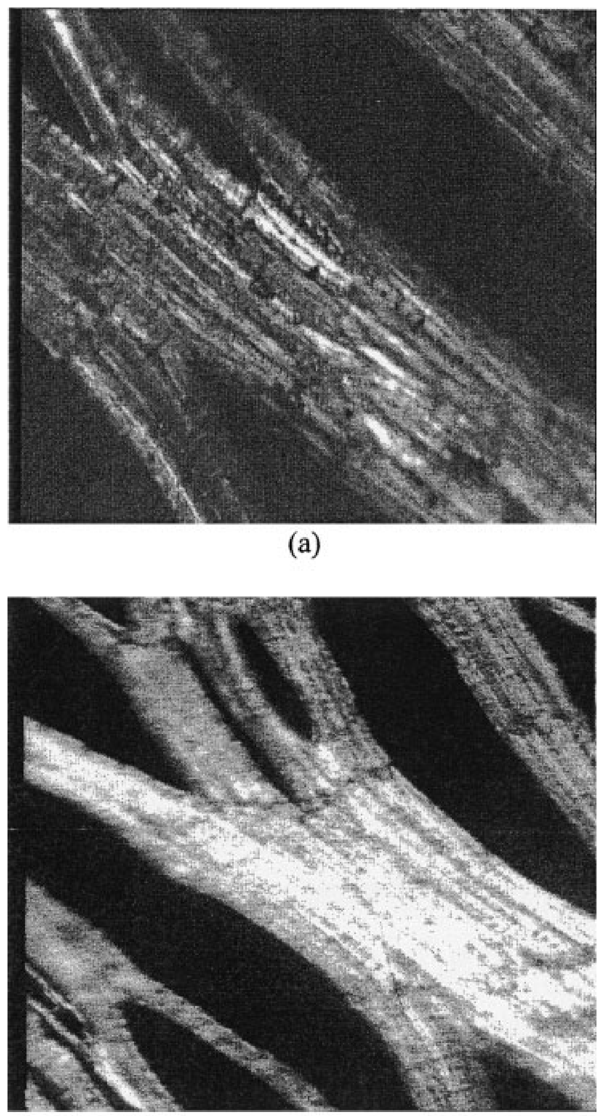
Figure 1 Scanning electron micrograms of (a) untreated and (b) 5% NaOH-treated *H. populifolia* fabrics.

ibre 201-15 grade of M/s Dow Chemicals Company) were used as the matrix components. The main feature of this epoxy system is its excellent mechanical and dynamic strength.

Araldite LY-556 is a clear liquid with a molecular weight of 450 g mol^{-1}, and viscosity at 25°C in the range of 9000 to 12,000 MPa s^{-1}.[21] The epoxy in the resin varies between 5.2 and 5.45 Eq kg^{-1}. It has a shelf life of three years if the product is stored in a dry place at a temperature range of 18–25°C. For achieving longer shelf life, the lid of the container is closed immediately after transfer of the material. The hardener HY-951 is a clear liquid with a viscosity at 25°C in the range of 5000–11,000 MPa s^{-1}. It has a shelf life of 2 years if the product is stored in a temperature range of 18–25°C. The polycarbonate has a molecular weight of 30,000 g mol^{-1}, and a melt flow rate (at 300°C, 1.2 kg weight), of 15 g/10 min. The heat deflection temperature at 264 psi load for an unannealed sample is 127°C, and the Vicat softening point is 120°C/h/kg.

The naturally occurring fabric *H. populifolia* was used as the reinforcement. This fabric was obtained from Kadiri Taluk of the Anantapur District, Andhra Pradesh, India. This fabric exists under the bark of the trunk and branches of the tree. The fabric is removed from the branch part of the tree. The tree belongs to the "Sterculiaceae" family. The average length, width, and thickness of the fabric were found to be 300–700 cm, 70–100 cm, and 0.18 mm, respectively.

The extracted fabric, which was naturally woven, was dried in the sun for a long period of time until it



(a)

(b)

Figure 2 Polarized optical micrograms of (a) untreated and (b) 5% NaOH-treated *H. populifolia* fabrics.

TABLE I
Flexural Strength and Modulus of the Matrix and Composites with Different wt % of the Natural Fabric *H. populifolia* Reinforced in an Epoxy/Polycarbonate (10%) Blend Matrix with (WCA) and without (WOCA) Coupling Agent (orientation angle $\theta = 0°$)

wt % of fabric	Flexural strength (MPa)				Flexural modulus (GPa)			
	Untreated fabric		Treated fabric		Untreated fabric		Treated fabric	
	WOCA	WCA	WOCA	WCA	WOCA	WCA	WOCA	WCA
15	122.38	136.68	145.11	167.59	26.98	34.75	46.24	50.29
20	130.74	145.92	157.76	178.2	31.54	40.66	48.35	57.41
25	135.59	153.68	167.65	185.95	39.76	43.05	52.71	60.37
30	141.02	159.2	170.26	194.53	44.39	48.76	59.55	64.98
35	155.86	165.91	176.19	200.35	50.12	54.31	65.34	69.53
40	159.61	170.42	192.3	215.73	53.85	59.76	67.89	74.33
Matrix	85.57				19.31			

was completely dry. The weaving appears to be intermingled. The dry fabric was treated with 5% aqueous solution of NaOH to remove the soluble hemicellulose and lignin. After this, the fabric was washed with water and dried in the sun.

Preparation of the samples

For making the composite, a die steel mold with 150 × 30 × 3 mm dimensions was used. The mold cavity was coated with a thin layer of aqueous solution of polyvinyl alcohol, which acted as a good releasing agent. To make the blend, the epoxy resin was added with polycarbonate dissolved in dichloromethane. The solvent was removed by degassing in vacuum for about 1 h. The hardener was added to this in stoichiometric ratios. A thoroughly mixed mixture containing 10% polycarbonate was used as the matrix. The composite was made by room temperature curing. To ensure complete curing, the composite and matrix sheets were post cured at 100°C for 3 h. Composites, with silane coupling agent treated fabric, were also prepared. For studying the effect of fiber orientation on the tensile properties of the composites, specimens with the fiber axis making angles of 0, 15, 30, 45, 60, 75, and 90° with respect to the stress direction were prepared.

Characterization

The flexural properties were studied using a flexural tester supplied by M/s PSI sales (P) Ltd., New Delhi. The fabric was observed both in a polarized optical microscope (Leitz Vario Orthomat 2) and a scanning electron microscope (JEOL JSM 820). The fabric was gold coated before subjecting it to SEM analysis. The surface of the cryogenically fractured and gold-coated composites was observed using scanning electron microscopy.

RESULTS AND DISCUSSION

Surface analysis of the fabric

The scanning electron micrograms of untreated and 5% alkali-treated *H. populifolia* fabric are presented in Figure 1. The untreated fabric [Fig.1(a)] shows a white layer on the surface of the fabric, which may be due to the hemicellulose, lignin, and the bonding material. This layer is absent in the microgram of the alkali-treated fabric [Fig.1(b)], confirming the elimination of hemicellulose. From these figures, it is clearly evident that the fabric is in a knitted form, with several void regions between fibers. Such geometry allows for the penetration of the resin in the void regions and, as a result, the bonding between the matrix and the fabric is expected to be good. Such an observation was also made by Narasimha Chary[22] and Rama Chandra Reddy[23] in the case of alkali-treated bamboo fibers. They reported a flow of the liquid resin into these micropores (enhanced by alkali treatment) by capillary action and the subsequent solidification by crosslinking, thus facilitating good bonding.

The polarized optical micrograms of untreated and 5% alkali-treated *H. populifolia* fabric are presented in Figure 2. The microgram image of the untreated fabric is diffuse [Fig.2(a)]. This may be the result of a presence of an amorphous hemicellulose layer on the surface of the fabric. The microgram image of the alkali treated fabric is, however, sharp [Fig.2(b)]. This may be due to the elimination of hemicellulose upon alkali treatment. Moreover, the surface of the alkali-treated fabric was found to be rough when compared to that of the untreated one.

Flexural properties

The flexural strength and modulus values of the composites with varying content of untreated and alkali treated fabric, both in the absence and presence of the silane coupling agent, are presented in Table I. For

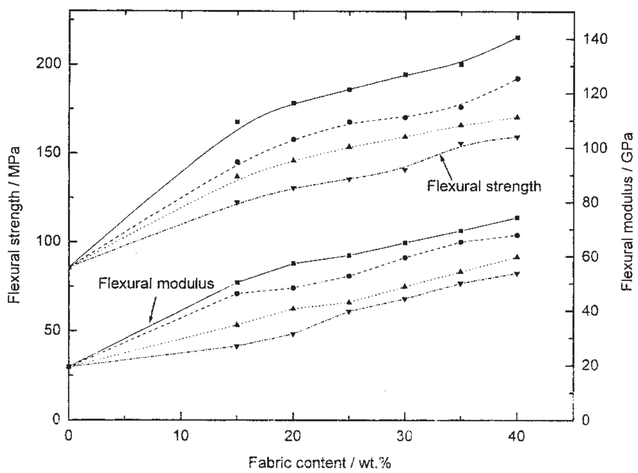


Figure 3 The variation of flexural strength and modulus as a function of *H. populifolia* fabric content in reinforced epoxy/10% polycarbonate blend matrices, with and without coupling agent (▼ = untreated without coupling agent; ▲ = untreated with coupling agent; ● = treated without coupling agent; ■ = treated with coupling agent).

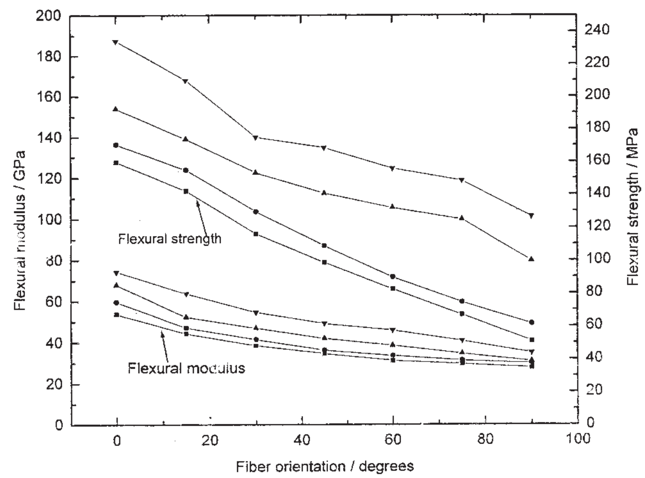


Figure 4 The variation of flexural strength and modulus as a function of *H. populifolia* fabric orientation in reinforced epoxy/10% polycarbonate blend matrices, with and without coupling agent (▼ = untreated without coupling agent; ▲ = untreated with coupling agent; ● = treated without coupling agent; ■ = treated with coupling agent).

comparison, the values for the matrix are also presented in the same table. The variation of flexural strength and modulus with different fabric contents are presented in Figure 3. Both the flexural strength and modulus increase nonlinearly with fabric content. The values are also higher when the fabric was treated with an alkali, and a coupling agent was used. Enhanced bonding between the reinforcement and the matrix as a result of alkali treatment, as well as the presence of a coupling agent, may be responsible for the increased flexural properties. Table I shows that the flexural strength and modulus of the composites are higher than those of the matrix.

The flexural strength and modulus of the composites, in which the fiber orientation was changed, are presented in Table II. For comparison, the values for the matrix are also presented in the table. A fabric content of 40 wt % was used for all orientations. The variation of flexural strength and modulus of these composites are presented in Figure 4. It shows the results for both untreated and alkali-treated fabric in the absence and presence of a silane coupling agent. It is clearly evident that both flexural strength and modulus are a maximum when the orientation angle (θ) is zero and a minimum when $\theta = 90°$ to the stress direction. The values for the other angles are intermediate between the maximum and the minimum.

It is also clear that the alkali treatment and the use of a coupling agent enhance the flexural properties. These may increase the bonding between the fabric and the matrix, and as a result, the properties improve. However, the bonding has to be confirmed by other methods, such as scanning electron microscopy (SEM).

TABLE II
Flexural Strength and Modulus of the Matrix and Composites with Different Orientations of the Natural Fabric *H. populifolia* Reinforced in an Epoxy/Polycarbonate (10%) Blend Matrix with (WCA) and without (WOCA) Coupling Agent (40 wt % Fabric Content)

Fabric orientation (degrees)	Flexural strength (MPa)				Flexural modulus (GPa)			
	Untreated fabric		Treated fabric		Untreated fabric		Treated fabric	
	WOCA	WCA	WOCA	WCA	WOCA	WCA	WOCA	WCA
0	159.61	170.42	192.30	215.33	53.85	59.76	67.89	74.33
15	142.07	154.8	173.77	210.02	44.36	47.14	52.41	63.81
30	115.95	129.57	153.07	174.94	38.56	41.5	46.95	54.67
45	98.68	108.79	140.57	168.38	34.61	36.29	41.99	49.22
60	82.41	89.70	131.89	155.76	31.25	33.65	38.49	45.97
75	66.83	74.52	124.70	148.21	29.59	31.33	34.65	40.78
90	50.97	61.54	99.62	126.56	27.79	29.92	30.95	35.14
Matrix	85.57				19.31			

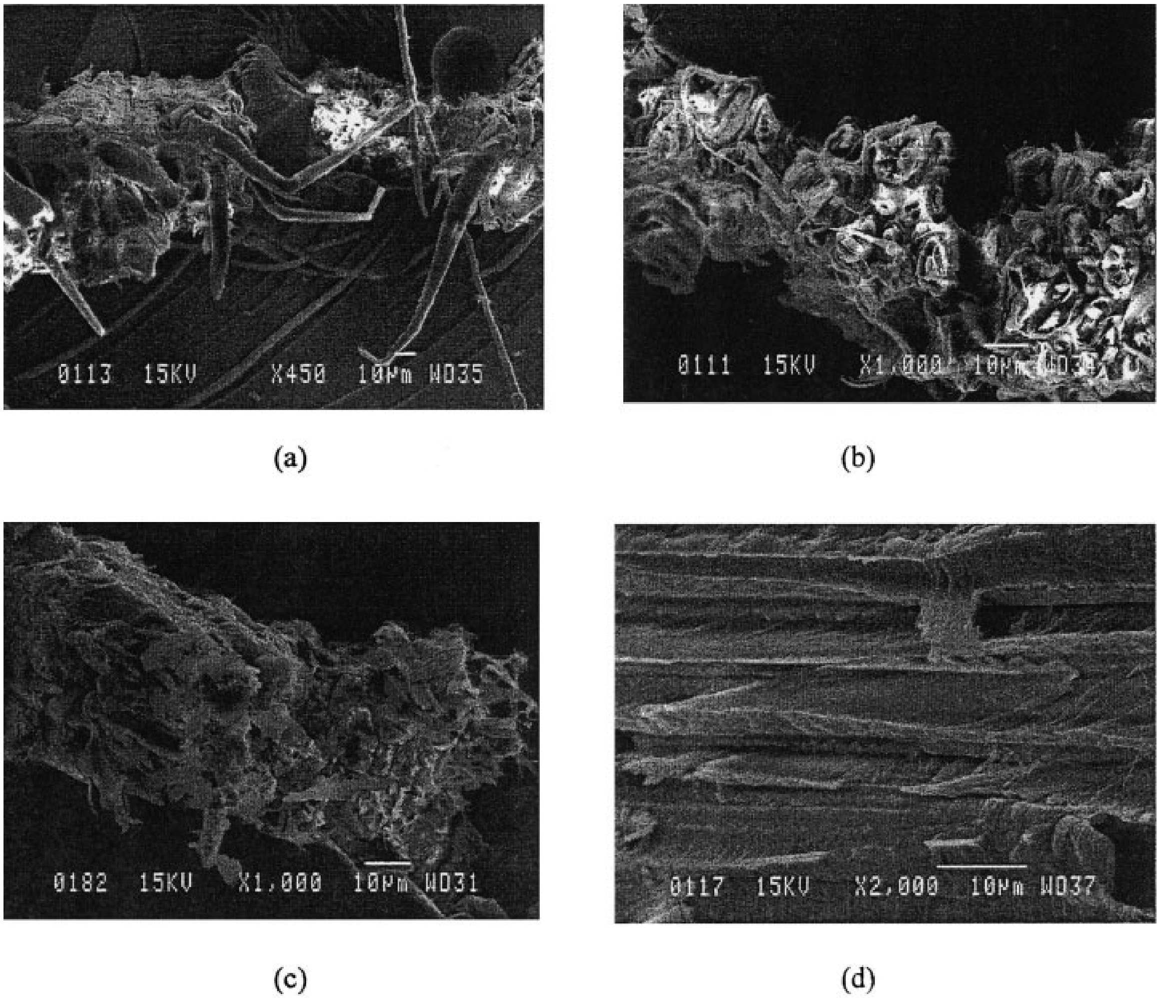


Figure 5 Scanning electron micrograms of *H. populifolia*-reinforced epoxy/10% polycarbonate blend matrices: (a) untreated without coupling agent, (b) untreated with coupling agent, (c) treated without coupling agent, (d) treated with coupling agent.

Morphology of the composites

The scanning electron micrograms of the untreated and treated *H. populifolia* fabric-reinforced polycarbonate (10%)-toughened epoxy composites, with and without coupling agent, are presented in Figure 5. The pull out of the fibers from the matrix is observed from Figure 5(a). This indicates poor adhesion between the reinforcement and the matrix when the untreated fabric in the absence of a coupling agent was used. Figure 5(b) shows that some fibers are broken and not pulled out. When the untreated fabric was used in the presence of the coupling agent, there was an improvement in the bonding between the fibers of the *H. populifolia* fabric and the matrix.

The scanning electron micrograms of the fractured surfaces of alkali-treated *H. populifolia* fabric reinforced in polycarbonate-toughened epoxy composites in the absence and presence of a coupling agent are presented in Figure 5(c) and Figure 5(d), respectively. From Figure 5(c) it is observed that the matrix formed a skin on the fibers when the fabric was treated with alkali, even in the absence of a coupling agent. This observation indicates improved bonding between the reinforcement and the matrix, even in the absence of a coupling agent, when the fabric was treated with alkali. Varada Rajulu et al.[15,16] reported that, when the *H. populifolia* fabric was treated with alkali, the hemicellulose and some part of the lignin were removed and, as a result, the fibers became thin. The density of the fabric also increased with alkali treatment. The surface of the fibers in the fabric became rough with the removal of hemicellulose and lignin. However, when a coupling agent was also used, it is observed that skin formation is enhanced, indicating an improvement in the bonding between the components of the composite. These observations are in line with the observed flexural properties.

CONCLUSIONS

The morphology of the untreated and alkali-treated natural fabric *H. populifolia* was investigated by scanning electron microscopy and polarized optical microscopy. Untreated and alkali-treated fabrics were used as a reinforcement in a polycarbonate-toughened epoxy matrix in the absence and presence of a coupling agent. The flexural properties of these composites are found to increase with alkali treatment and in the presence of the coupling agent. The scanning electron micrograms of the fractured surfaces of the composites reveal enhanced bonding between the reinforcement and the matrix when the fabric was treated with alkali and a coupling agent was used.

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