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Tensile Response and Fracture and Failure Behavior of Jute Fabrics-Flyash-Vinylester Hybrid Composites

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Abstract: In this work, hybrid materials consisting on a vinylester matrix simultaneaously reinforced with jute woven fabrics and flyash particles were prepared. The tensile response and the fracture and failure behavior of these hybrid composites were investigated. Thermal stability of these materials was also studied. The aim was to obtain an environmentally friendly hybrid material with a good balance of tensile and fracture properties at relatively low cost. The effect of a novel treatment for the jute fabrics on the hybrids mechanical and fracture properties was investigated. The best balance of tensile and fracture properties was obtained for the hybrid consisting of fabrics treated with alkali under stress and fly ashes which also exhibited relatively high thermal stability.

Keywords: Natural fibers, Fly ash, Hybrid composite, Mechanical properties, Fracture

Introduction

A pre-requisite of most engineering materials is that they have good stiffness and strength along with adequate toughness. Man-made composites usually fulfill these requirements, especially because they exhibit crack-stopping capability which makes them very attractive for structural or semi-structural applications. For these applications, the knowledge of the material load-bearing capabilities is essential.

In addition, over the recent years there is an increasing interest in natural fibers as a substitute for glass fibers mainly because of their low specific gravity, workable specific modulus, low cost, as well as their non-abrasive, renewable and biodegradable nature [1]. Therefore, novel applications involving furniture as well as engineering end uses, such as building materials and structural parts for motor vehicles, were developed [2-4].

Mechanical properties of composites depend on several factors, such as content and orientation of the reinforcement, interfacial interaction and mode of testing [5], but in the case of natural fibers other factors can also influence those properties. Each natural fiber has different contents of cellulose, hemicellulose, and lignin as main compounds, as well as other compounds such as pectins and waxes [3]. The technical natural fiber is composed of several elementary fibers joined by cementitious material, such as pectin and lignin, and the fibers have a hole along the fiber length formed by the lumen. Such interphase between elementary fibers plays an important role because in this region longitudinal splitting can occur. The above factors complicate the evaluation of mechanical properties in the case of natural fiber composites.

On the other side, there is also an increasing trend of current industries to re-use their wastes, mainly as a result of economic and ecological concerns. Ashes are solid industrial wastes produced in the combustion of carbon and other fossil fuels which are mainly composed of a significant amount of SiO_2 and lower contents of other oxides (Al_2O_3, Fe_2O_3, Na_2O, MgO, K_2O, etc). They are cheaper and more environmentally friendly than conventional mineral fillers, therefore they represent an appealing alternative as reinforcement in polymers. This has been already the subject of many investigations [6-19].

On the other hand, hybrid materials imply a step beyond in the search for novel materials with improved mechanical properties and/or reduced cost for both advanced and standard applications [20]. They are made by the incorporation of several different types of reinforcement into a single matrix [14,21]. With this combination, it is possible to obtain a good balance of the desirable material properties simultaneously minimizing the undesirable ones. This cannot be developed in a single-reinforcement composite and allows designers to tailor composites to suit various requirements. When properties of the different reinforcements differ greatly, a hybrid effect is expected to exist for the hybrid reinforced composites [21].

The concept of hybrid materials is not new. Several authors have studied the use of a combination of reinforcements on various systems [14,21,22]. Recently, Cicala *et al.* [23]

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studied the hybridization of glass fibers with natural woven mats in a vinylester resin matrix for applications in the piping industry. They found that the hybrid fulfilled the mechanical requirements for these applications saving simultaneously cost and weight.

Most of the literature concerning hybrid composites is mainly focused on the combination of different types of fibrous reinforcements, however the combination of particulate reinforcements with natural fiber woven mats is almost a still a scarcely explored area.

In this work, hybrid materials consisting of a vinylester matrix simultaneously reinforced with jute woven fabrics and flyash particles were prepared. The mechanical response and the fracture and failure behavior of these hybrid composites were investigated. The aim was to obtain an environmentally friendly hybrid material with a good balance of tensile and fracture properties at relatively low cost.

Experimental

Materials

Flyashes kindly supplied by Industrias del Tablero S.A. (INTASA, Spain) were used as one of the reinforcement components. They were obtained from the biomass combustion and subsequently separated using a sieve of mesh 250. This results in a particle size distribution with a mean particle size between 10 and 20 μm.

The second reinforcement consisted of commercially available woven jute fabrics (Casthanal, Textil CIA, Brazil). Two types of fabrics were employed as detailed below.

The first type of fabrics were jute fabrics washed with distilled water and detergent solution, and dried until constant weight in an oven at 80 ^oC under vacuum before use. These simply washed fabrics will be referred to here as washed fibers.

The other type of fabrics were jute fabrics treated with Na(OH) aqueous solution (5 % w/v) for 4 hours with continuous shaking at 25 ^oC under biaxial tension. They will be referred here to as treated fabrics. During treatment, the elongation was kept at 1.2 % using a specially designed device. In this device, the fabric is placed between fixed and movable clamps on both directions (simultaneously). Two screws in each clamp allow applying a well-defined displacement. This alkali under tension treatment was explained in detail in a previous investigation [24]. These treated fabrics were washed with distilled water until all the sodium hydroxide was eliminated, that is the pH was neutral. Finally, fabrics were dried at 80 ^oC until constant weight was attained.

The matrix material was prepared from a general purpose vinylester resin (Derakane Momentum 411-350 from Dow, kindly provided by Poliresinas San Luis, Buenos Aires, Argentina).

Hybrids Preparation

Matrix and flyash were stirred in an ultrasonic bath at room temperature.

Each layer of jute fabric was pre-impregnated with the mixture of vinylester and flyash and placed one over the other in the mold by a hand lay-up technique, taking care to keep practically achievable tolerances on fabric alignment. Four layers were compression molded under a pressure of 50 MPa in a hydraulic press.

Different hybrid composites with simply washed jute fabrics or with treated fabrics were prepared. For all composites, jute content was close to 40 wt % and flyash content was about 30 wt %. The final nominal thickness of the plaques was 3 mm.

The results of this investigation were compared with previous results on vinylester composites independently reinforced with jute fabrics or flyash particles [24-26].

The hybrids were cured with MEKP (Metil Etil Ketone Peroxide) as catalyst and cobalt naphthenate (CoNap) as accelerator in weight fraction of 1 and 0.5 %, respectively at room temperature. Then, all plaques were postcured for 2 h at 140 ^oC in an oven.

Thermogravimetric Analysis

Dynamic thermogravimetric measurements were performed by using a thermo- balance Shimadzu TGA 50 (Japan). Temperature programs for dynamic tests were run from 30 ^oC to 950 ^oC. Samples between 10 to 20 mg were heated at a heating rate of 10 ^oC/min. TG/DTG tests were carried out under air atmosphere (30 m*l*/min).

Mechanical Characterization

Uniaxial tensile tests were performed in an Instron dynamometer 4467 at 2 mm/min in accordance with ASTM D638-92 standard recommendations. An incremental mechanical extensometer was used to measure actual elongation during the tests. Stress-strain curves were obtained from these tests and Young's modulus and tensile strength values were determined from these curves.

Flexural tests were also carried out in three-point bending in accordance with ASTM D790-93 standard recommendations in the Instron dynamometer 4467 at 2 mm/min. Flexural modulus values were determined from the initial slope of the load-displacement plots.

Single-edge notched bend (SENB) specimens were also cut out from the plaques. Sharp notches were introduced by sliding a fresh razor blade into a machined slot. Crack-to-depth (a/W), thickness-to-depth (B/W) and span-to-depth (S/W) ratios were always kept equal to 0.5, 0.5 and 4, respectively.

Fracture characterization was carried out in three-point-bending in the Instron dynamometer at 1 mm/min by following ASTM 5045-93 Standard recommendations.

Energy release rate values at initiation (G_{IC}) were obtained.

As the hybrids exhibited a semi-controlled propagation way, i.e., load did not suddenly drop to zero after the critical load was reached, the work of fracture (W_f) was also determined. It was calculated from the total area under the load-displacement curve (U_T) divided by twice the area of the fracture surface (since two new faces are created), as follows [27,28]:

$$W_f = U_T / 2B(W - a) \quad (1)$$

Where B is the thickness (mm), W is the sample width (mm) and a is the notch lenght (mm).

The work of fracture was defined simply as the total energy consumed to produce a unit area of fracture surface during the "complete" fracture process.

In order to measure the apparent shear strength, compression shear tests were also performed on the different hybrids. The apparent shear strength τ_D was calculated as P_{max}/A, where P_{max} is the maximum measured load and A is the shear area. For these measurements, the special apparatus developed by Lauke *et al*. [29,30] was used. In this set-up, the specimen is placed between 2 adjustable load noses and pure shear stress is applied by compression [29]. These tests were carried out at a crosshead speed of 1 mm/min on specimens which dimensions were 8×8×3 mm.

Fracture Surface Analysis

In order to study the failure mechanisms operative in the different hybrids investigated, fracture surfaces of specimens broken in uniaxial tensile tests were analyzed by scanning electron microscopy (SEM) after they had been coated with a thin layer of gold.

Results and Discussion

Thermogravimetric Analysis

Figure 1(a) and (b) presents the derivatives results of the mass percent as a function of temperature for the hybrids compared with their respective composites and the matrix filled with flyash. Figure 1(a) corresponds to the composite with washed fibers and Figure 1(b) to the composite with alkali-treated fibers. The decomposition peak in the range of 250 °C and 300 °C corresponds to the thermal depolymerization of hemicellulose, lignin and the glycosidic linkages of cellulose [31] The peak at about 350 °C represents the decomposition of α-cellulose for most of the fiber reinforced samples. Finally, the peak at about 440 °C is related to the decomposition of the vinylester matrix [32] and is present in all samples. Furthermore, degradation of lignin is known to occur first and at a slower rate than the other components [33] having a broader peak between 200 °C and 500 °C. Therefore, this peak probably overlaps the other peaks.

For the composites with alkali-treated fabrics, the main peak shifted to a higher temperature due to the partial removal of lignin and hemicellulose promoted by the alkali

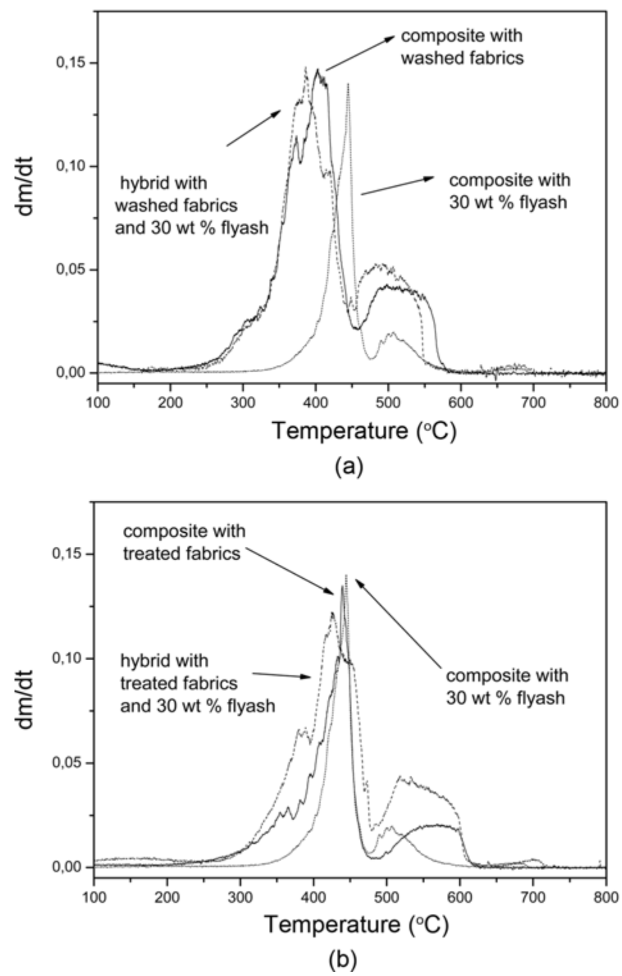


Figure 1. DTGA curves for the composites and hybrids with different fiber treatments; (a) with washed fabrics and (b) with alkali-tension treated fibers. The composite with 30 wt % flyash is also included in both graphs.

treatment [33-35].

It should also be noted that the main degradation peak temperature does not exhibit significant differences among those for the hybrids and those for their corresponding composites (composites reinforced with treated or untreated fabrics), suggesting that hybridization does not significantly affect the materials thermal stability. However, the alkali treatment does lead to a shift of the peaks to higher temperatures improving the thermal stability of the materials.

Uniaxial Tensile Behavior

The hybrids showed some non-linear behavior up to fracture, as a result of the development of incipient damage such as, fiber failure, particle debonding, matrix cracking and/or fiber pull-out as it will be shown later. Simultaneous fracture of fibers and matrix in all samples was evidenced by a marked drop of load after fracture. This behavior is not significantly different from the behavior displayed by the

Table 1. Young's modulus, tensile strength, fracture energy values and flexural modulus for the matrix and the different hybrids investigated

	Young's modulus E (GPa)	Tensile strength σ_u (MPa)	Critical energy release rate G_{IC} (kJ/m^2)	Work of fracture w_f (kJ/m^2)	Flexural modulus E_f (GPa)
Vinylester matrix	3.27 ± 0.41	39.45±3.37	0.48±0.02	0.16±0.01	3.22±0.07
30 wt % flyash-vinylester composite [26]	4.03 ± 0.12	44.00±4.05	0.91±0.09	0.48±0.04	3.98±0.02
Washed jute-vinylester composite [36]	5.97 ± 0.42	40.83±8.47	5.17±1.64	3.02±0.31	4.29±0.29
Alkali-treated jute-vinylester composite [24]	7.68 ± 0.38	59.2±1.54	4.15±1.54	1.70±0.57	7.26±0.15
Hybrid with washed fabrics	6.54 ± 0.66	42.19±5.19	4.82±0.31	1.96±0.13	6.59±0.20
Hybrid with alkali-treated fabrics	6.27 ± 0.60	42.78±1.38	5.62±0.41	2.01±0.11	6.19±0.25

composite independently reinforced with washed fibers or alkali-tension treated fabrics reported in a previous investigation [24].

Tensile parameter values (Young's modulus and ultimate strength) for the hybrid materials with washed fabrics and with fabrics treated with alkali under biaxial stress are presented in Table 1 along with their deviations. The values for the fiber-reinforced composite with only washed fabrics or alkali-treated fabrics and the flyash-reinforced composite, originally reported in References [24,26,36], are also included in Table 1 for comparison. These materials will be referred to as composites in the following text.

It can be observed in Table 1, that within the wide scatter of experimental values which was not unexpected for this kind of materials, the hybrids obtained in this work showed higher values of stiffness and tensile strength than the vinylester matrix or the composite with only washed fabrics. In addition, the vinylester-flyash composite exhibited a lower value of tensile modulus compared to the other reinforced materials. This was not unexpected as the main reinforcement effect in Young´s modulus comes from the fibers and not from the particles. However, that composite still retained a relatively high value of strength as a result of some kind of interaction between vinylester and flyash [26].

Nevertheless, the significantly high values of tensile parameters obtained of the composite with alkali-tension treated fabrics could not be attained from hybridization. Particularly, in the case of the hybrid with alkali-treated fabrics under stress, the beneficial effect from the woven fabric incorporation to the vinylester resin on tensile properties reported before [24,36] and also shown in Table 1, was counteracted by the addition of the particulate reinforcement, mostly due to processing problems associated with the increase in the resin viscosity [28]. Insufficient wetting of the matrix due to the presence of flyash in the hybrids hindered matrix penetration inside elemental fibers and thus, decreased the material capability to withstand loads.

Flexural Behavior

Table 1 also shows the flexural modulus of the matrix, the composite with 30 wt % flyash, the composites with washed fabrics and with alkali-treated fibers under stress, and the hybrids. In agreement with our results of tensile stiffness, the hybrids showed improved flexural modulus respect to the matrix, the composite with flyash or the composite with simply washed fabrics.

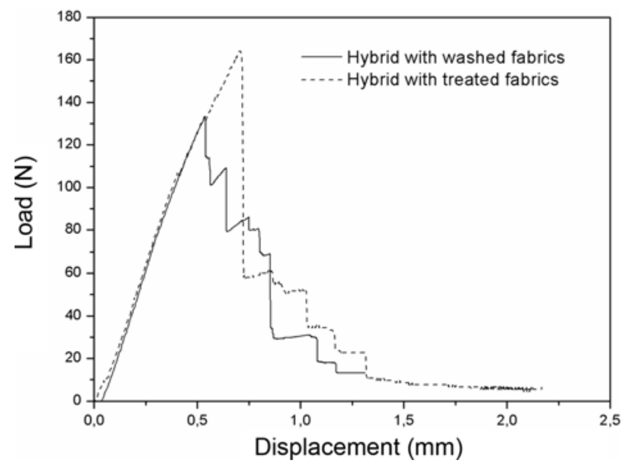


Figure 2. Typical load-displacement curves obtained in three-point-bending fracture tests for the hybrid with washed fabrics and for the hybrid with fabrics treated with alkali under stress.

Fracture Behavior

Typical load-displacement curves for the two hybrids investigated (the hybrids with washed fabrics and the hybrid with fabrics treated with alkali under stress) are presented in Figure 2. They were obtained in three-point-bending fracture tests.

As it can be observed in Figure 2, the hybrid containing alkali-treated fabrics presented more marked drops of load after the maximum value than the hybrid with washed fabrics which exhibited a more gradual drop of load up to complete fracture. This effect could be attributed to the better conservation of the fiber structure promoted by the alkali treatment under stress. In an earlier work [24], the effect of the treatment of jute fibers with alkali under biaxial stress was analyzed. It was concluded that this treatment improves stiffness and strength of the fibers and promotes a better conservation of the fabric structure in comparison to the conventional alkali treatment or the simply washing of fabrics. In the hybrids with treated fabrics investigated here, the conservation of the fabric structure leads to the existence of zones rich in matrix that allow the crack to easily propagate and zones rich in fibers that stops the crack propagation, with a few larger steps in the load-displacement records. In the hybrids with washed fabrics, on the other hand, the presence of jute yarns randomly distributed within the matrix leads to a more gradual crack propagation pattern.

Critical energy release rate (G_{IC}) and work of fracture (W_f) values for the hybrids are also presented in Table 1 along with their deviations. G_{IC} and w_f values for the composites with washed fabrics, with treated fabrics or with 30 wt % flyash, originally reported in References [26] and [24], are also included in this table for comparison.

It can be observed in Table 1, that G_{IC} values for the hybrids with washed fabrics and flyash were in the same range of values of the composite with only washed fabrics or with treated fabrics, whereas these values were significantly higher than the values for the composites with 30 wt % flyash and even much higher than the matrix value. In addition, the hybrid with treated fabrics exhibited the highest value of fracture energy at initiation. However, no significant differences between work of fracture values (proportional to the area under the load-displacement records) for the two hybrids investigated were found. These results can be attributed to two counteracting effects: the higher fiber strength promoted by the treatment with alkali under stress which contributed to both initiation and propagation steps on one hand, and the more gradual decrease of load exhibited by the hybrid with washed fabrics during crack propagation which mostly contributed to the w_f values, on the other hand.

In agreement with our tensile results, no significant synergistic effect from the incorporation of a particulate reinforcement in a jute-fabric reinforced composite could be attained. As mentioned before, the beneficial effect obtained from the incorporation of the woven fabric into vinylester on the material fracture behavior [37] was counteracted by the addition of the particulate reinforcement which led to an increase in the resin viscosity and hence, to insufficient wetting of the elemental fibers by the matrix with the subsequent detrimental effect on the material crack stopping capability.

Compression Shear Tests

A common way to measure interfacial properties in composite materials is the compression shear test (CST) [29,30,38]. This method is widely used to evaluate the apparent shear strength which is a sensitive quantity to know the composite consolidation, and thus interfacial adhesion.

Values of apparent shear strength for the composites with washed fabrics or treated fabrics and for the hybrids investigated in this work are presented in Table 2, along with their deviations.

The two hybrids exhibited higher apparent shear strength values than the composite with washed fabrics or treated fabrics, being the value for the hybrid with treated fabrics slightly higher than that for the hybrid with washed fabrics. Therefore, the improved value of G_{IC} for the former could also be attributed to a better interfacial adhesion in that material.

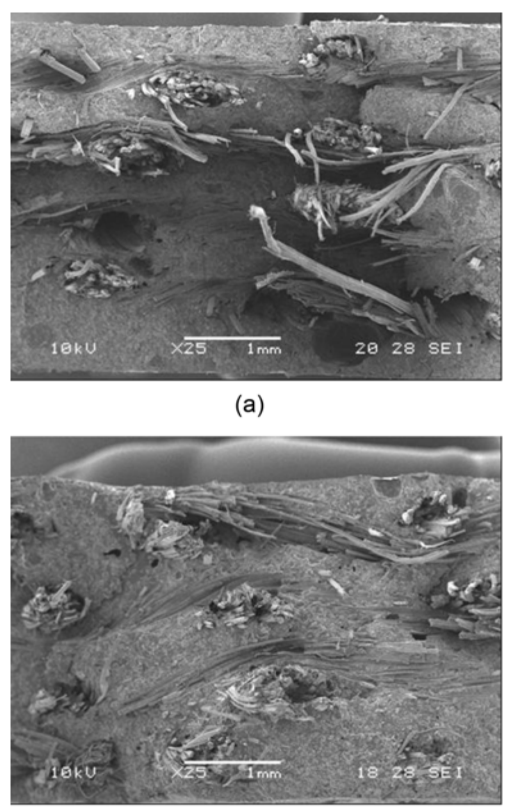
Table 2. Interlaminar shear strength τ_D for the different composites investigated

	Interlaminar shear strength, τ_D (N/mm^2)
Washed jute-vinylester composite [24]	33.78 ± 3.08
Alkali-treated jute-vinylester composite [24]	39.94 ± 1.23
Hybrid with washed fabrics	45.74 ± 1.35
Hybrid with alkali treated fabrics	48.66 ± 1.14

Fracture Surface Analysis

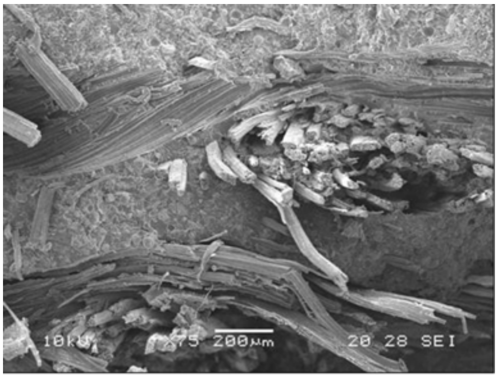
In order to analyze the failure mechanisms operative in the hybrids investigated, scanning electron microscopy (SEM) was performed on the fracture surfaces of samples broken in uniaxial tensile tests.

Figure 3(a) and (b) shows SEM micrographs of the fracture surfaces of the hybrid with washed fabrics and the hybrid with alkali-tension treated fabrics, respectively.



(b)

Figure 3. SEM fractographs for: (a) the hybrid with washed fabrics and (b) the hybrid with alkali-tension treated fibers.



(a)

(b)

Figure 4. Closer views of Figure 3.

As it can be observed in Figure 3, the hybrid with treated fabrics presents a higher conservation of the fabrics structure showing the entire yarn whereas the hybrid with washed fabrics exhibits untighted yarns where individual fibers can be clearly observed.

Closer views of Figure 3 are shown in Figure 4(a) and (b). It is seen in this figure, that the main failure mechanisms of the hybrids investigated were found to be yarn pull-out and debonding of ash particles from the vinylester matrix. In addition, it is important to note that the pull-out length for the hybrid with treated fabrics was notably shorter than that for the hybrid with washed fabrics (compare Figure 4(a) with Figure 4(b)). This is in agreement with the results of compression shear tests which indicated that the former material had better interfacial adhesion. The hybrid with treated fabrics also exhibited the highest critical energy release rate value (G_{IC}) while no significant differences were found between work of fracture values for the two hybrids. Therefore, it appears that interfacial adhesion mainly affected crack initiation rather than the propagation step in the hybrids investigated here.

Conclusion

The tensile and fracture and failure behavior of jute fabrics-flyash-vinylester hybrid composites was investigated.

The effect of a novel treatment of the fabrics with alkali under superimposed biaxial tensile stress and the microstructure on the hybrids tensile and fracture properties was analyzed.

The material with the best combination of mechanical (stiffness and tensile strength) and fracture properties (critical energy release rate and work of fracture) was found to be the hybrid with fabrics treated with alkali under biaxial stress. This was the result of a better interfacial adhesion and improved stiffness and strength of the jute fabrics promoted by that treatment. In addition, this material also exhibited improved thermal stability.

In this work, an environmentally friendly material having a good balance of tensile, fracture and thermal properties at relatively low cost was obtained.

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