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A novel fiber treatment applied to woven jute fabric/vinylester laminates

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Abstract

In this work, a novel fiber treatment consisting on an alkali treatment superimposed to biaxial tensile stress was successfully applied to woven jute fabric/vinylester laminates. The effect of treatment on the composites tensile properties was investigated at two different times of treatment. A significant improvement in stiffness was achieved by the composite treated with alkali under stress for 4 h. However, no significant differences between the stiffness of the untreated composite and the composites treated with alkali under stress for 24 h were found. On the other hand, irrespectively of the time of treatment, the composites with fabrics treated with alkali under stress showed the highest values of tensile strength. From results of fabrics tensile tests, compression shear tests and X-ray diffraction analysis, the improved tensile properties exhibited by the composites with treated fabrics could be attributed to structural changes of the fibers as well as to a change in the fiber/matrix interfacial properties.

Keywords: A. Polymer–matrix composites (PMCs); A. Laminates; B. Mechanical properties; B. Interface/interphase

1. Introduction

A pre-requisite of most engineering materials is that they have good stiffness and strength along with adequate toughness. Man-made composites usually fill this requirement, especially since they exhibit crack-stopping capability which makes them very attractive for structural or semi-structural applications.

On the other hand, 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, low cost, as well as their renewable and biodegradable nature [1]. However, natural fiber reinforced materials have substantially inferior mechanical and water resistance properties than conventional glass fiber reinforced composites.

In order to overcome these disadvantages, several treatments have been proposed in the literature [2–5]. In the case of natural fibers, in addition to the matrix/fiber interface modification, different changes on the interphase between elementary fibers, as well as on the roughness and density of the technical fibers could be also induced by chemical treatment [6,7]. Furthermore, other factors such as the orientation of microfibrils of cellulose within each elementary fiber which changes the fiber crystallinity play an important role [8]. On the other hand, the development of stress in fiber bundles is governed by the binder that separates the elementary fiber. Therefore, two kinds of interfaces should be taken into account in this case: one between fiber bundles and the matrix and the other between elementary fibers. All these factors complicate the dependence of mechanical properties on treatment methods [9,10].

The presence of polar groups in natural fibers is responsible for their good adhesion with thermosetting matrices. Several fiber treatments conducted on the natural fibers (i.e., alkali treatment, silane treatment, acetylation, and acrylonitrile treatment) exist. They not only modify the interphase but also produce morphological changes on the fibers as explained above.

From the economic point of view, alkali treatment appears the most viable one [1,11–14]. Removal of

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impurities such as waxes, pectin, mineral salts and some hemicellulose and lignin is promoted by that treatment. Extensive structural changes which strongly depend on the parameters of the alkali treatment (i.e., Na(OH) concentration, temperature and time of treatment) are also induced. The treatment conditions as well as the diversity of morphology and composition of natural fibers, make the evaluation of the effect of alkali treatment on the composite properties [15] very complicated. Therefore, differences in the literature for different fiber and matrix combinations and treatment conditions are frequently found.

One effect of alkali treatment is the modification of the crystallographic cell from cellulose I to cellulose II. Alkalization process also remarkably affects the surface morphology of natural fibers resulting in a more distinct exposition of the surface fibrils and a more pronounced surface relief. Due to the intra and interfibrillar swelling, the accessibility of fibers changes drastically [16,17]. Depending on the treatment condition, the alkali treatment produces a rough surface, hence the number of anchorage points increases offering a good fiber–matrix mechanical interlocking [18].

In the present work, a novel fiber treatment consisting on an alkali treatment superimposed to biaxial tensile stress was applied to woven jute fabric/vinylester laminates. The effect of the treatment on the tensile properties of the laminates was investigated at two times of treatment.

2. Experimental

2.1. Materials

Commercially available woven jute fabrics (Casthanal, Textil CIA, Brazil) were used as reinforcement. The matrix material was prepared from general purpose vinylester resin (Derakane Momentum 411–350 from Dow, kindly provided by Poliresinas San Luis, Buenos Aires, Argentina) and accelerator in a weight ratio of 1:0.05, respectively.

2.2. Fabric treatments

2.2.1. Washed

Jute fabrics were washed with distilled water and detergent solution, and dried until constant weight in an oven at 80 °C under vacuum before use. These simply washed fabrics will be referred here to as untreated fabrics.

2.2.2. Treatment with alkali under biaxial tensile stress

Jute fabrics were treated with Na(OH) aqueous solution (5% w/v) for two different periods of time (4 and 24 h) with continuous shaking at 25 °C under biaxial tension. The elongation was kept at 1.2% during treatment using the especially designed device shown in Fig. 1. 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.

Then, the fabrics were washed with distilled water until all the sodium hydroxide was eliminated, that is the pH was neutral. Finally, they were dried at 80 °C until constant weight was attained.

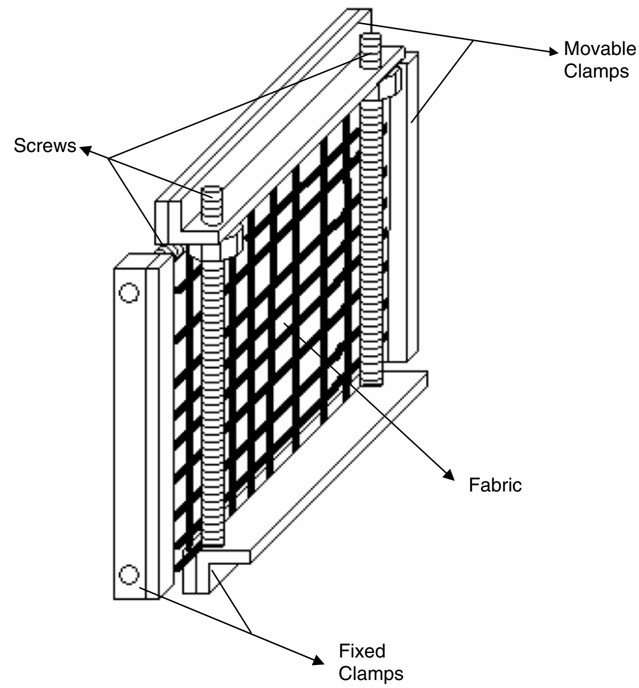

Fig. 1. Scheme of the especially designed device for the treatment of fabrics with alkali under biaxial tension.

2.3. Composites preparation

Each layer of jute fabric was pre-impregnated with matrix material 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 in a hydraulic press for 1 h at 80 °C. Then, the plaques were postcured 2 h at 80 °C followed by 2 h at 140 °C in an oven. Different composites with simply washed jute fabrics or with treated fabrics were prepared. For all composites, jute content was close to 40 wt%.

2.4. Tensile tests

2.4.1. Fabrics

The compliance of fabrics (initial displacement/load ratio) was obtained from tensile specimens directly cut from untreated and treated woven jute fabrics whose fineness is shown in Table 1. The distance between clamps

Table 1
Fineness of jute yarns

Yarn	Fineness (tex)	Std. deviation
Untreated	351.8	76.63
Alkali-tension 4 h	279.4	33.12
Alkali-tension 24 h	253.2	61.18
Alkali 24 h	261.4	41.05

was always kept at 150 mm and the sample width was 50 mm. The number of yarns by sample was counted and compliance values were referred to these values. Five measurements were done for each type of fabric. Tests were carried out in an Instron dynamometer 4467 at 2 mm/min crosshead speed.

2.4.2. Composites

Tensile specimens were machined from the compression-molded plaques of 3 mm thickness in accordance with ASTM D3039M-95 standard recommendations [19]. Uniaxial tensile tests were performed in the Instron dynamometer 4467 at 2 mm/min by using an incremental mechanical extensometer 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.

2.5. Compression shear tests (CST)

Although the short-beam test is the most commonly used method applied in shear strength measurements, it has been demonstrated to be inappropriate in many cases [20,21]. Therefore, to overcome the disadvantages of this test Lauke et al. [22,23] developed a compression shear test which allows subjecting in shear a predetermined plane. In this work, compression shear tests were performed in order to determine the apparent shear strength τ_D 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. [22,23] was used. Specimens were placed between two adjustable noses and loaded in compression as shown in Fig. 2. This loading promotes a shear-dominated failure at the midplane of the specimen leading to an estimation of apparent shear strength of the composite [22].

Tests were carried out at a crosshead speed of 1 mm/min on specimens which dimensions were 8 mm × 8 mm × 3 mm.

2.6. X-ray diffraction analysis

Crystallinity index (CI) of fibers extracted from the treated and untreated fabrics was measured by wide-angle X-ray diffraction at room temperature. Samples were prepared by powder and subsequently laid on a glass sample holder.

A Phillips PW 1050/25 difractometer operating in a 2θ range between 5° and 70° was used. The scattering angle was varied in 1°/min steps. CuKα radiation was used as source.

Wide-angle X-ray diffraction curves were analyzed in order to determine the effect of the treatments with alkali under stress on the fiber crystallinity. Crystallinity index was obtained by the ratio of the area under crystalline peaks and the total area over the angular range of data [24].

2.7. Fracture surface analysis

Fracture surfaces of specimens broken in tensile tests were analyzed by scanning electron microscopy (SEM) after they had been coated with a thin layer of gold.

3. Results and discussion

3.1. Tensile tests

Experimental compliance (initial displacement/load ratio, v/P) values per yarn of untreated fabrics and fabrics treated with alkali under stress at two different treatment times are shown in Fig. 3. A significant decrease in the compliance of fabrics was found as a result of both alkali-tension treatments used, suggesting that these treatments induced structural changes in the fabrics some of which will be confirmed later from X-ray analysis. An increase in stiffness and strength of individual natural fibers by the treatment with alkali has been previously attributed to the change of cellulose I into cellulose II promoted by the alkali treatment that leads to a tighter packaging of the chains and an increase in the degree of molecular orientation [7].

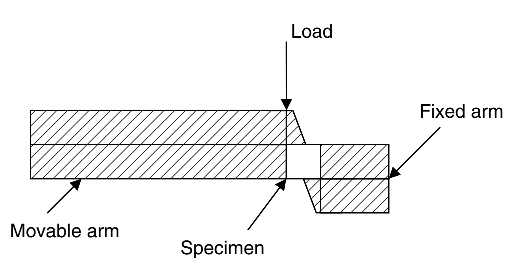

Fig. 2. Experimental setup for compression shear test.

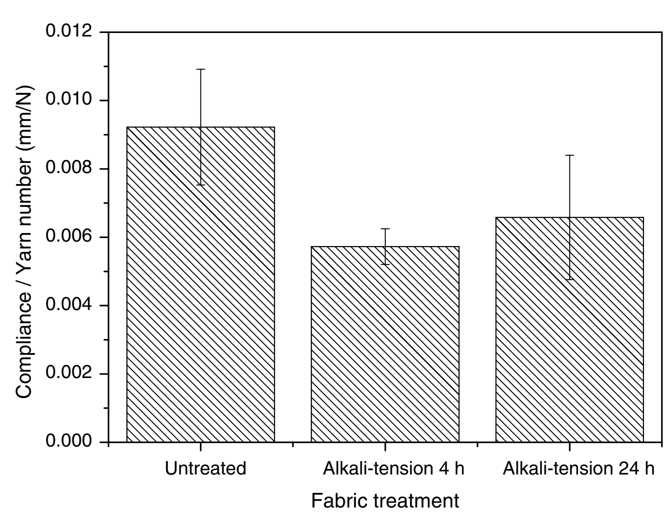

Fig. 3. Experimental compliance values per yarn of untreated fabrics and fabrics treated with alkali under stress during 4 h and 24 h.

Similar results have been also previously reported in the literature [5] for jute/epoxy quasi-unidirectional composites treated with alkali under different shrinkage conditions. These results have been explained in terms of the rupture of the alkali-sensitive bonds between different fiber components, the formation of new hydrogen bonds between some cellulose chains, changes in parts of the crystalline cellulose and changes in the orientation of the molecular chains as well as to a highly reduced fiber diameter.

In addition, Alvarez et al. [28] observed that stiffness values of sisal fibers exhibited a maximum with time of treatment with alkali. Hence, the higher value of the compliance of jute fabrics treated for 24 h compared to that for 4 h treated fabrics could be mainly related to the presence of a maximum in the fiber stiffness values with treatment time.

In uniaxial tensile tests (Fig. 4), all composites exhibited stress–strain curves with some degree of non-linearity before maximum load as a result of the development of incipient damage such as matrix cracking, fiber failure or fiber pull-out. Simultaneous fracture of fibers and matrix in all samples was evidenced by a marked drop of load after fracture.

Fig. 5 presents Young's modulus values for the composites with fabrics treated with alkali under stress. The results obtained in a previous work [25] for untreated, silicone-treated, alkali-treated and acetylated composites are also included in Fig. 5 for comparison. It is clearly observed in this figure, that a significant improvement in stiffness was achieved by the composite treated with alkali under stress for 4 h (22% modulus improvement compared to the untreated composite). It is also important to note that superimposed tensile stress during treatment with alkali for 24 h led to a 12% modulus improvement respect to the same treatment without stress. However, no significant differences between the stiffness of untreated composite and the composite treated with alkali under stress for the longest time (24 h) were found.

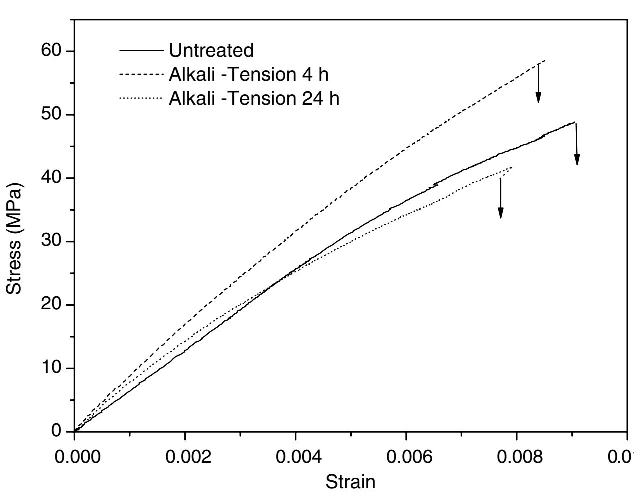

Fig. 4. Typical stress–strain curves for untreated composites and the composites treated with alkali under stress for 4 h and 24 h.

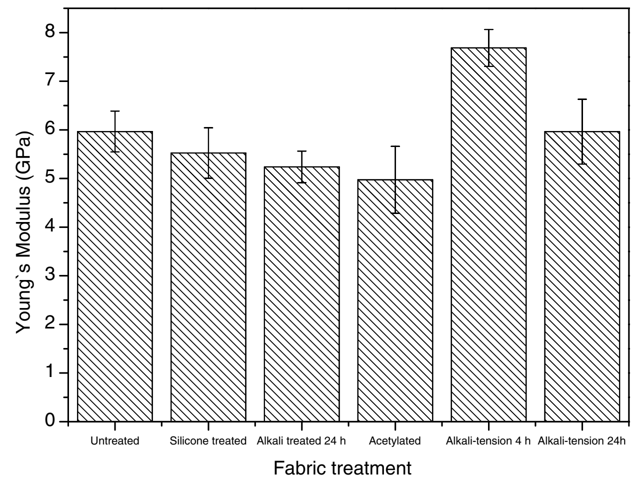

Fig. 5. Young's modulus values for the different composites investigated.

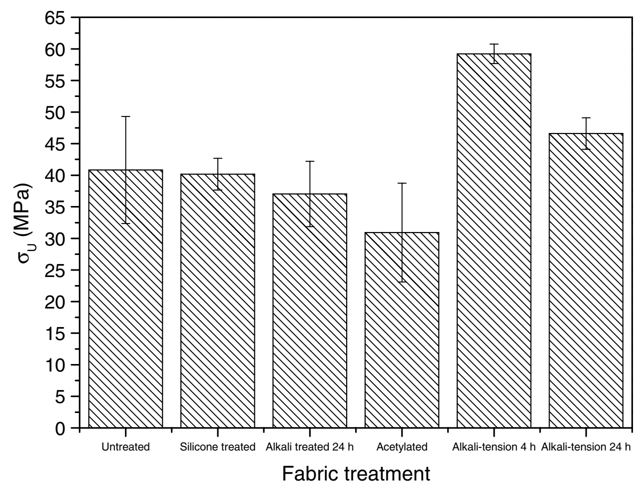

Fig. 6. Tensile strength values for the different composites investigated.

On the other hand, Fig. 6 shows tensile strength values for the different composites investigated. Irrespectively of the time of treatment, the composites with fabrics treated with alkali under stress showed the highest values of strength even for the longest time of treatment.

3.2. Compression shear tests (CST)

Values of apparent shear strength for untreated and alkali-tension treated composites are shown in Fig. 7. Irrespective of the time of treatment used, a slight improvement in the apparent shear strength was achieved by the alkali treatment with superimposed tensile stress and thus in the interfacial adhesion. The connection between layers of a composite (called consolidation) affects mechanical properties. Since shear loads induce delamination between layers, the quantity interlaminar shear strength, as estimated by the compression shear test [22], is likely to relate to consolidation, and therefore serves as a measure of interfacial adhesion. An increase in the OH concentration due

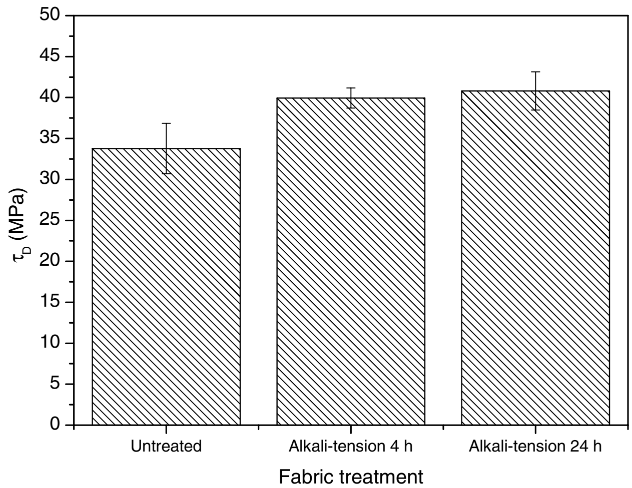

Fig. 7. Apparent shear strength values for untreated and alkali-tension treated composites.

to the changes in the spiral angle and higher exposition of OH, when cellulose I changes to cellulose II [8] is expected from the alkali treatment. The fiber with a higher amount of OH groups would become more compatible with the vinylester matrix and therefore, a higher ability to transfer shear stresses from the matrix to the fibers might develop.

3.3. X-ray diffraction analysis

The degree of crystallinity of the different treated fibers is shown in Fig. 8. A marked increase in the crystallinity was achieved by the treatment with alkali under stress. However, it is well established in the literature [26] that physicochemical properties are not only determined by the degree of crystallinity but also by the distribution pattern of crystallite size. In addition to the observed increase in the crystallinity of the fibers as a result of the treatment with alkali under stress, a conversion of cellulose I into cellulose II should be expected as mentioned before as well as a change in the crystallite size [27].

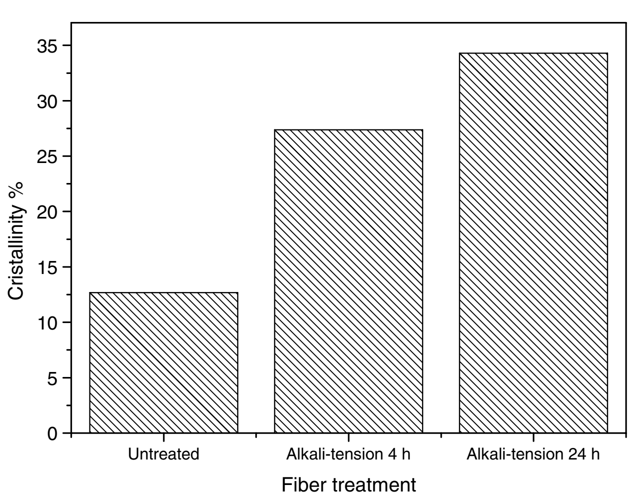

Fig. 8. Crystallinity index (CI) of the different treated fibers.

Hence, from the results of fabrics tensile tests, compression shear tests and X-ray diffraction analysis, the improved tensile properties exhibited by the composites with treated fabrics, could be attributed to structural changes of the fibers as well as to a change in the fiber/matrix interfacial properties in the case of the strength.

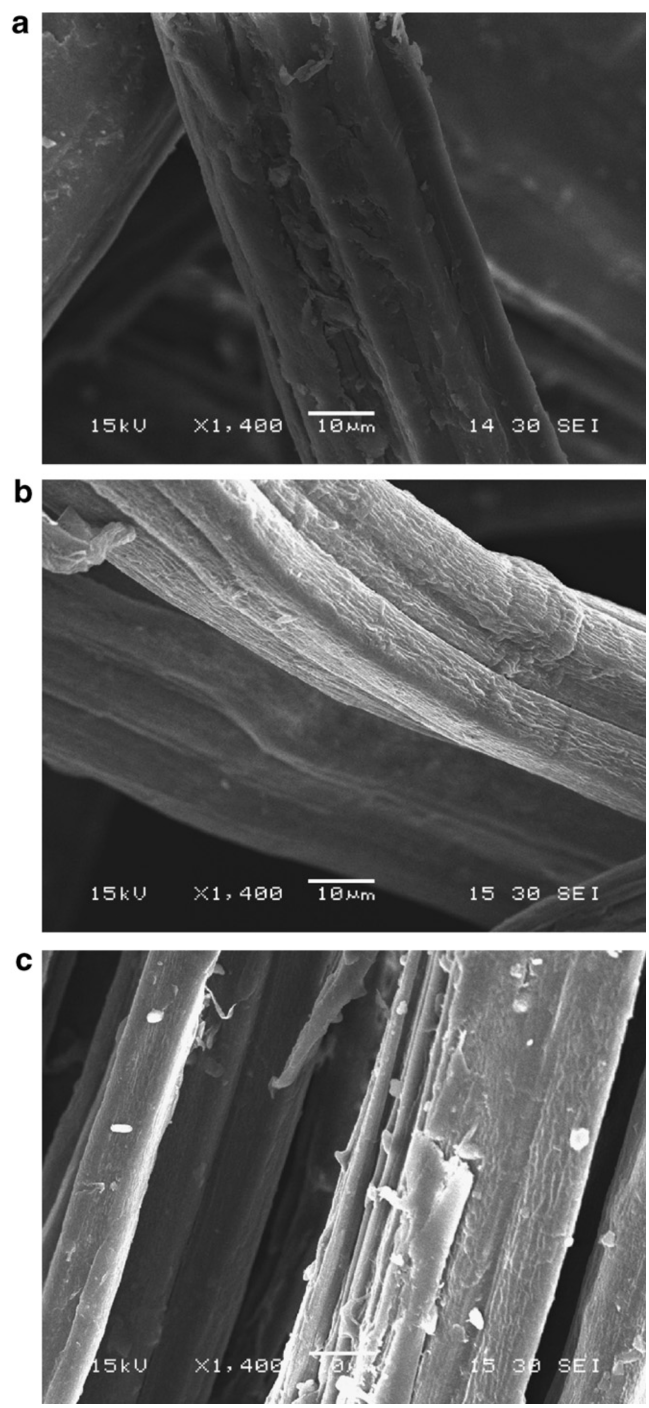

Fig. 9. SEM micrographs of jute fibers: (a) untreated fibers, (b) fibers treated with alkali under biaxial tensile stress for 4 h and (c) fibers treated with alkali under biaxial tensile stress for 24 h.

On the other hand, interface quality should not be expected to affect stiffness results as it is determined at low loads where the interface is still intact [20].

3.4. SEM analysis

SEM micrographs of untreated jute fibers and the fibers treated with alkali under biaxial tensile stress are shown in Fig. 9. It can be observed in this figure that in untreated fibers the cementicious material between the fibers cells which holds the structure together is clearly seen. In addition, as the time of treatment increases individual ultimate fibers become more visible and they also appear more fibrillated.

The alkali treatment leads to fiber fibrillation. This phenomenon has been reported to increase the effective surface area available for contact with the matrix, reduce the fiber diameter and produce a rougher surface. As a result of these, fibrillation may promote better mechanical interlocking [29]. In our case, better adhesion was found in alkali-tension treated fiber composites compared to the untreated one but no significant differences between composites treated at different treatment times were found. The longer time of treatment for the composite treated under 24 h tension did not lead to further increase in adhesion compared to the 4 h treated composite as a result of the increased damage induced onto the fibers treated for the longest time.

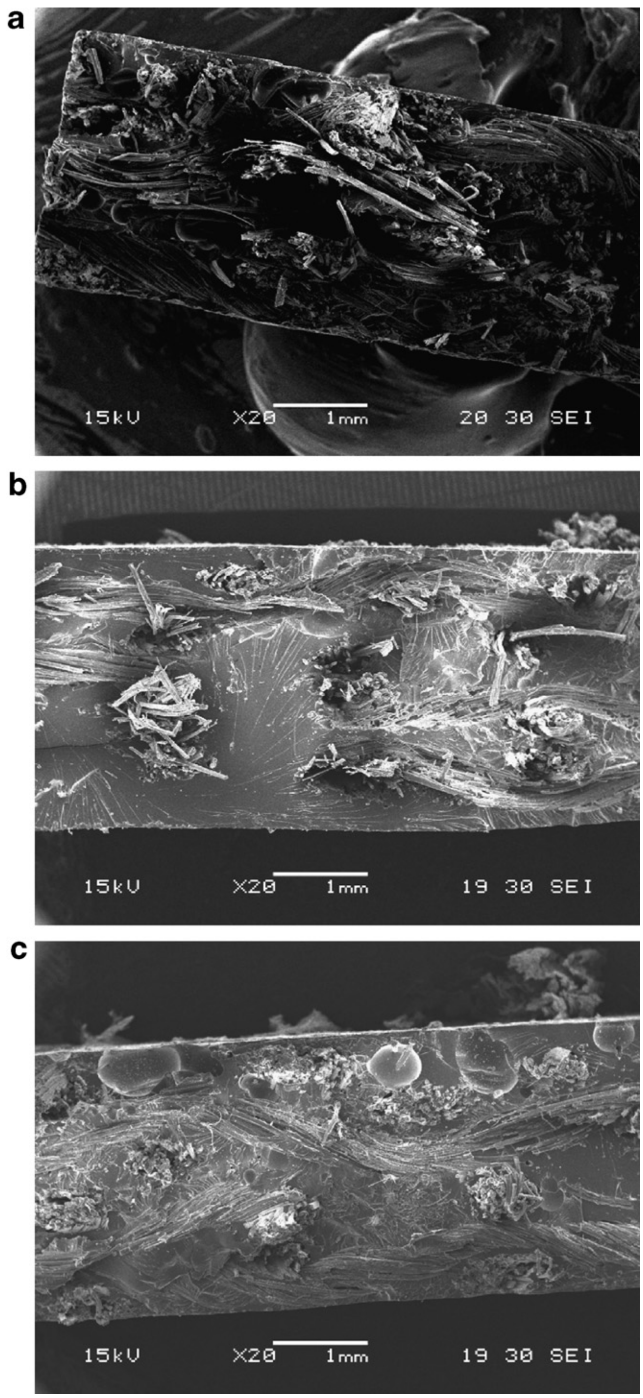

Fig. 10. SEM micrographs of composites samples tested in tension: (a) untreated composite, (b) composite treated with alkali under biaxial tensile stress for 4 h and (c) composite treated with alkali under biaxial tensile stress for 24 h.

Therefore, the detrimental effect of the alkali treatment time under tensile stress on tensile properties could be due to the damage induced onto the fibers by that treatment. It would counteract the improvement of interfacial properties and the increase in fiber crystallinity observed in Figs. 7 and 8, respectively.

On the other hand, the composites with fabrics treated with alkali under stress (Fig. 10) exhibited more tighten fabric structures than untreated composites and well-defined jute and matrix zones were clearly observed. In contrast, in the untreated fabric composite jute yarns were randomly distributed in the whole material (Fig. 10a) thus contributing to the worse tensile properties exhibited by this material in the loading direction (warp or weft direction of the jute fabric) compared to the alkali-tension treated composite.

4. Conclusions

In this work, a novel fiber treatment consisting on an alkali treatment superimposed to biaxial tensile stress was successfully applied to woven jute fabric/vinylester laminates.

A significant improvement in stiffness was achieved by the composite treated with alkali under stress for 4 h. However, no significant differences between the stiffness of the untreated composite and the composite treated with alkali under stress for 24 h were found. On the other hand, for both times of treatment, the composites with fabrics treated with alkali under stress showed the highest values of strength even for the longest time of treatment.

From the results of fabrics tensile tests, compression shear tests and X-ray diffraction analysis, the improved tensile properties exhibited by the composites with treated fabrics could be attributed to structural changes of the fibers as well as to a change in the fiber/matrix interfacial properties in the case of the strength.

Further work is in progress in order to determine the optimum conditions of the alkali under stress treatment to obtain better mechanical properties.

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