ALKALI TREATMENT OF JUTE FABRICS: INFLUENCE ON THE PROCESSING CONDITIONS AND THE MECHANICAL PROPERTIES OF THEIR COMPOSITES

Exequiel S. Rodríguez and Analía Vázquez

Research Institute of Materials Science and Technology, INTEMA – Materials Department, Engineering Faculty, National University of Mar del Plata. J. B. Justo 4302, B7608FDQ, Mar del Plata, Argentina.
Corresponding Author's e-mail: anvazque@fi.mdp.edu.ar

ABSTRACT: The aim of this work is to evaluate the effect of the fibers alkali treatment on the injection processing. Woven jute preforms were used to prepare the composites using the vacuum infusion technique. The fibers were treated with NaOH (5 wt%) for 24 h at room temperature. Single filament tests showed that the treatment was detrimental for the mechanical properties of the fibers. The injection times increased in the treated jute preforms as a consequence of the increase in the exposed area and the flow resistance. The preform permeability decreased, also, in the tubular structure collapse of the fibers, which could reduce the capillary pressure. Flexural and impact properties of the treated jute composites decreased mainly in the lower mechanical properties of the fibers.

KEYWORDS: Natural fiber composites, vacuum infusion, alkali treatment, processing conditions, preform permeability, mechanical properties.

INTRODUCTION

The idea of replacing glass fibers with natural fibers in composite materials is increasing. The success of natural fiber composites (NFCs) depends on their ability to overcome their main disadvantages (low fiber/matrix adhesion, fluctuations in fiber properties, low thermal and fire resistance) and the possibility of using well-studied glass fiber reinforced plastics (GFRP) processing techniques. Different approaches have been used to improve NFCs performance.
Chemical treatments of the fibers [1, 2], chemical modification of the resins [3, 4] and coupling agents [2, 5] have been employed to enhance fiber/matrix adhesion. Although many chemical treatments have proved to be suitable for enhancing NFCs mechanical properties, it is difficult to select the best treatment for a particular fiber. The intrinsic differences of natural systems coming from diverse geographical regions and different harvesting, production and processing condition make the available results difficult to compare, even for the same kind of fiber. A relatively simple chemical treatment is the alkali treatment (mercerization), which has been successfully used in fibers like jute, sisal, hemp, palm or flax [1-2,5]. In cotton there is more than a century of experience in the use of mercerization to improve fibers performance.

Regarding the processing technique, in vacuum infusion , a liquid resin is introduced into a closed mold with the dry reinforcement inside. The closed system eliminates most of the fumes that are liberated in hand lay-up. These methods allow better control over part dimensions and fiber volume fraction. Hence, resin infusion methods overcome many of the limitations of wet lay-up processes and have low cost. This is compatible with its use with low-cost fibers. Among all the natural reinforcements, jute appears to be a promising fiber suitable for vacuum infusion, because it is relatively inexpensive and commercially available in the required form.

In this work, natural fiber composites were obtained by vacuum infusion using untreated and alkali-treated jute fabrics. The effect of the treatment on the mechanical properties of the fibers, the processing conditions (injection times, flow pattern, perform impregnation) and the mechanical properties of the composites was studied. Although many studies about chemical treatments of natural fibers can be found in the scientific literature, there is very few research in which its influence on the whole obtaining process (fiber and matrix, processing and final product) is analyzed. The intention of the work was not to obtain the best treatment conditions, but rather to make evident the effects of the treatment on fibers, composite properties and on the processing conditions.

EXPERIMENTAL

Commercial jute bi-directional fabrics were used as reinforcement. The matrix was an epoxy vinylester resin: Derakane 411-350 Momentum from Dow Chemical. VE resin containing 45 wt% styrene was used as purchased without removal of inhibitors. The initiating system was comprised of methyl ethyl ketone peroxide (MEKP, 1.5 wt%) together with cobalt nafthenate (0.6 wt%).

The fibers were washed with detergent (2 vol% in aqueous solution, 15% active matter) and then immersed in beakers with a solution of 5 wt% NaOH for 24 h at room temperature. After that, the fibers were washed thoroughly with distilled water to remove the excess of NaOH and dried at 70ºC for 24 h under vacuum. The major fibres constituents (α-cellulose, hemicellulose and lignin) of the untreated and alkaline-treated jute fibre samples were determined by chemical analysis following standard procedures. The results are listed in Table 1.

Table 1. Jute chemical composition

Component (%)	Treated	Untreated
α-Cellulose	80.3	71.1
Hemi-cellulose	9.6	15.9
Lignin	7.6	11.8
Other components (ash, water, pectins)	2.5	1.2

Composites with 30 vol% of reinforcement were prepared using the vacuum infusion technique. Injections were conducted on an acrylic mold with a 4mm x 10mm x 450mm rectangular cavity. The resin enters through an injection point located on one side of the mold and the suction point was located in the opposite side. Two injections were performed for each kind of composite. Molded plaques were cured at 80ºC for 2 h, and post-cured at 110ºC for 3 h.

RESULTS AND DISCUSSION

- Effect of the alkali treatment on the fiber properties

After treatment, semicrystalline and amorphous portions in the fibers, such as hemicellulose, lignin and other alkali-soluble fraction, were preferentially removed. The following effects of the alkali treatment on natural fibers have also been reported in the literature:

- It cleans fiber surface removing impurities, waxy substances and natural oils. It also produces a rough surface topography, facilitating mechanical interlocking which leads to an improvement in fiber-matrix adhesion.

- Produces fiber fibrillation, i.e. axial splitting of the elementary fibers (or microfibers) that constitute the elementary fiber [3, 6]. This process leads to a decrease in fiber diameter, increasing the aspect ratio and the effective surface area available for wetting by a matrix in a composite. There is also an increase in fiber density as a consequence of the collapse of its cellular structure.

- Increases the number of free hydroxyl groups in the fiber surface, which improves the adhesion with resins like polyester or vinylester.

Also, there are more physical and chemical changes in the fibers affect the tensile modulus and the strength in an opposite manner. In favor: the decrease in spiral angle and the increase in crystallinity, the better rearrangement of the fibrils along the load axis and the increment in the fibers aspect ratio. Against: removal of lignin and hemicellulose, which play a cementing role, transferring the stress to the microfibrils. Therefore, the alkali treatment is an effective procedure to improve the mechanical properties of natural fibers, but its success depends on many variables, so it is very difficult to predict the effect over a particular natural system.

In order to evaluate the effect of the treatment on the mechanical properties of the fibers used in this work, single filament tests were performed. Table 2 shows the results for untreated and treated fibers. Both tensile strength and modulus showed an important drop after the alkali treatment. The treatment used in this work produced an excessive extraction of lignin and hemicellulose, probably with damage in the ultimate cells walls. The decrease in elastic modulus is an evidence of the loss of cementing material in the fibers. Mannan and Talukder [7] obtained similar results for dewaxed and delignificated jute. The fibers were treated with 17.5% NaOH for 1h at room temperature. On the other hand, Gassan and Bledky [8] reported an important increase in modulus and strength of jute yarns treated with a dense caustic soda solution (25 wt%) for 20 min under isometric conditions (the fibers are not allow to shrink). When the shrinkage is avoided, the tensile stresses developed in the fibers produce microfibrills orientation that is responsible for the increase in modulus. In slack conditions (free shrinkage) the authors reported a decrease in fibers modulus. Other authors [6, 9] have reported improvement in jute composites mechanical properties after fibers alkali treatment with solutions in the range of 5 to 10 % o NaOH for about 4h at room temperature.

Table 2. Single filament tests results

Fiber	Modulus (GPa)	Strength (MPa)
Untreated	30 ±14	505 ±165
Alkali-treated	12,2 ± 5,2	326 ±150

- Effect of the alkali treatment on the processing conditions

In reference to the processing conditions, the pressure difference needed to obtain an injection time (t_{inj}) of 60 seconds was calculated using Equation 1, derived from Darcy's law for constant pressure flow. The porosity (ϕ) and the resin viscosity (μ) at the temperature of each injection were used for the calculations. The permeability values for each porosity ($K_{preform}$) were calculated using the Carman-Kozeny model (Equation 2) and the constants for the model (n, C) were taken from previous work of untreated and non-washed jute [10]. The obtained injections times were 95 and 90 seconds for the untreated jute fabric and 135 and 128 seconds for the treated one. The processing conditions for two injections are summarized in Table 3.

$$\Delta P = \frac{L^2_{mold} \; \mu\phi}{2 \, K_{preform} \; t_{inj}} \tag{1}$$

$$K_{preform} = \frac{\phi^{n+1}}{C(1-\phi)^n} \tag{2}$$

where, ΔP is the pressure difference driving the flow, L_{mold} is the flow front position.

In all of the injections conducted, the obtained injection time was higher than the estimated. This could be attributed to the decrease in the fabric permeability. The permeability values used in the calculations correspond to "as received" jute. When the fibers are washed, the coatings added to facilitate the woven/weaving procedure (potato starch and waxes) are eliminated and the fiber surface became rougher. Then, there is an increment in the exposed area and therefore in the flux resistance. Injection time was higher in the mercerized fabric because the treatment produced fiber fibrillation, i.e. axial splitting of the elementary fibers (or microfibers). This process led to a decrease in fiber diameter, increasing the exposed area even more than in the only washed fibers.

Table 3. Main processing variables

Variable	Injection with untreated jute	Injection with treated jute
Porosity	0.70	0.69
Kpreform (m^2)	4.25 x 10-9	4.63 x 10-9
ΔP (hPa)	625	570
μ (Pa.s)	0.310	0.263
t_{inj} estimated (s)	60	60
t_{inj} actual (s)	95	135

There is another factor that can increase injections times: the decrease in capillary pressure. During the injection, the resin can flow either among the bundles that constitute the preform (macroflow) or inside the bundles, i.e. among the fibers filaments (microflow) [11]. In natural fibers the possibility of resin flow inside the fibers lumen also exists. The driving force for the microflow is the capillary pressure developed in the intratow region that should be added to the static pressure in Darcy's law in order to have more accurate results. But if the collapse and compacting of the fibers structure occurs during mercerization, the microscopic flow is inhibited and the volumetric flow rate should decrease. There is no agreement in the literature on the real effect of the microflow on the injection times. Dungan and Sastry [12] claimed that the microflow decrease the overall permeability by increasing

penetration times due to the required wetting of the tows. The evidence for their proposal was the higher values of the saturated (or wet) permeability in comparison to the unsaturated (or dry) permeability. Pillai and Advani [13] explored the effect of delayed impregnation of fiber tows in porous media constituted of woven fiber mats. They modeled this phenomenon by the inclusion of a "sink" effect (or negative source term) in the equation of continuity for the flow in the intertow regions. On the other hand, other authors [14, 15] found that the capillary pressure can produce a wicking flow that is faster than the macroflow, obtaining unsaturated permeability values higher than the saturated ones. Chang and Morgan [16] explained that the wicking flow could be faster or slower than the macroflow depending on the relative values of capillary and viscous forces. The last ones dominate at high impregnation velocities, i.e., low porosities or high injection pressures. In the case of this work, the injection pressure is relatively low so a positive contribution of the capillary pressure to the permeability is expected, resulting higher in the untreated jute fabric than in the alkali treated one.

- Effect of the alkali treatment on the composites properties

The results of the mechanical properties of the composites are summarized in Table 4. In general, mechanical performance of a fiber composite basically depends on the strength and toughness of the matrix, and efficiency of interfacial stress transfer [5]. Specifically, the modulus does not depend on interface adhesion. The elastic stiffness is defined as the strain approaches to zero; thus the degree of adhesion has no bearing on the elastic stiffness of the system. The decrease in flexural modulus observed in the alkali treated jute composites compared to the untreated ones, is a consequence of the decrease in fibers modulus. The same trend was observed for the flexural strength.

Table 4: Mechanical properties of the composites

Property	untreated jute composite	treated jute composite
Flexural modulus (GPa)	6.6 ± 0.5	5.5 ± 0.2
Flexural strength (MPa)	103 ± 6	83 ± 6
Impact Energy (J/m)	56.5 ± 2.4	47.2 ± 4.2

The impact tests showed that the impact energy of the composites decreased when the jute fibers were treated. This result was predictable considering the factors that affect energy consumption during the impact:

- fiber/matrix adhesion: as explained before, an increase in the interfacial strength, which is expected after treatment, reduces energy consumption by avoiding fiber pull-out. In this case, the fracture of the material takes place with little change in the cracking plane, breaking the fibers instead of pulling them out.

- Fibers mechanical properties: their decrease after treatment is obviously unfavorable for the impact resistance.

- Fibers spiral angle: Pavithran et al. [17] found that fibers with high spiral angle formed composites with higher toughness than those with small spiral angles. Even though the comparison was done among different kind of fibers, the decrease in spiral angle that takes place during alkali treatment should be detrimental for the composites toughness.

- Analysis by scanning electron microscopy

In order to have a better understanding of the effects of alkali treatments on the jute composites, SEM micrographs were obtained form the fractured surface of the composites. Figures 1a-c-e and 1b-d-f shows the micrographs for the untreated and treated jute composites respectively. Fibrillation and diameter reduction is evident in the alkalized jute (Fig. 1b). Fig. 1d and 1f also show the damage in the cell walls and a rougher fiber surface. Also, the collapse of fibers lumen can be seen. This is responsible for the decrease in capillary pressure and therefore in preform permeability. On the contrary, the untreated jute (Fig. 1c and 1e) shows open lumens, which in many cases are full of resin (marked with arrows). Both composites showed good fiber-matrix adhesion and low pull-out, which is consistent with the results obtained in previous work [18].
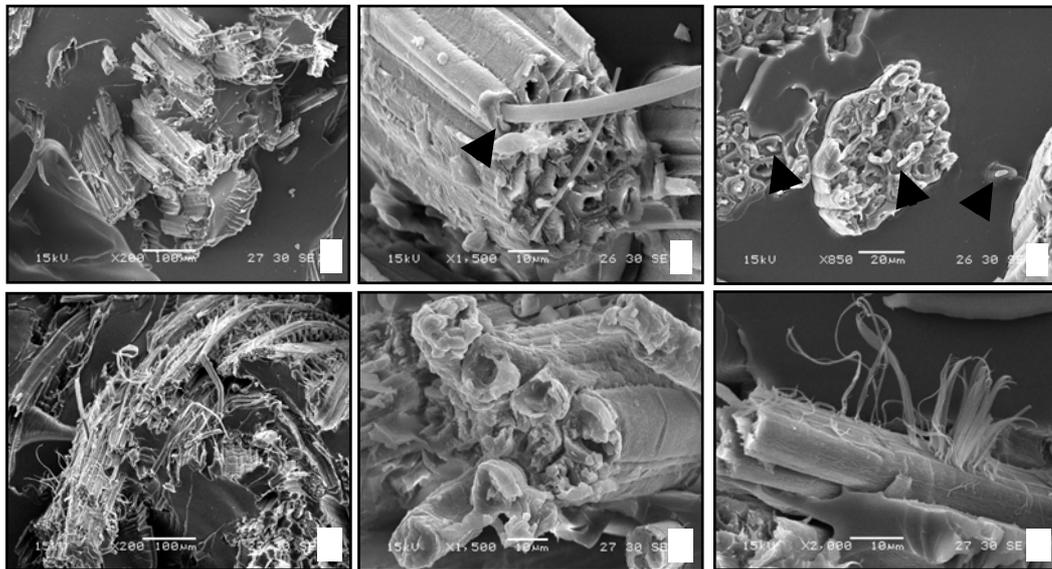

Fig. 1 SEM micrographies of the fracture surface of the composites

CONCLUSIONS

As a consequence of the severe alkaline treatment that was applied to jute fibers in this work, the following effects could be seen in fiber properties and processing conditions:
- The longer injections times obtained for the treated jute preforms were due the more compact and rougher fiber surface that produced the treatment. A lower capillary pressure that leads to lower preform permeability was expected as a consequence of the fiber lumen collapse that could be observed by SEM.
-The alkali treatment produced a drop in both tensile strength and Young's modulus of the fibers. This was attributed to the damage induced in the cell walls and the excessive extraction of lignin and hemicellulose, which play a cementing role in the structure of the fibers.
- The treatment also produced fibrillation, i.e. splitting of the microfibers that compound the technical fiber. This process leads to diameter lowering and higher dispersion in strength values. The high dispersion that presented the modulus values was in part due to deviations of

the cylindrical model for the fibers used in the diameter determination: optical micrographs showed that some of the fibers were split, had ramifications and presented a tape form rather than cylindrical.

- The composites processed presented a brittle behavior with lower flexural and impact properties in the case of the composites reinforced with treated jute. The drop in the mechanical properties of the fibers was the main cause of these results.

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