

# Aspects of Alkali Treatment of Sponge Gourd (*Luffa cylindrica*) Fibers on the Flexural Properties of Polyester Matrix Composites

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**ABSTRACT:** Sponge gourd (*Luffa cylindrica*) forms a natural mat that deviates the crack path in brittle thermoset resin matrix composites, leading to a controlled fracture mode and increasing the toughness of the composite. The use of luffa as reinforcement is, however, restricted by a very weak fiber–matrix interface. In this work, luffa fibers were alkali-treated at two temperatures, with varying alkali concentrations. Although the surface analysis shows that the treatments promote a clear removal of the outer surface layer of the fibers with the exposition of the inner fibrillar

structure and the consequent increase of the fiber surface area, only a secondary increase on the mechanical properties was obtained. The slight increase observed was attributed only to mechanical interlock. Long pullout lengths and neat fiber beads were the main features observed at the fracture surface of the treated and untreated fiber composites. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 87: 1927–1932, 2003

**Key words:** mechanical properties; biofibers; composites

## INTRODUCTION

The use of natural fiber-reinforced composite materials is, nowadays, steadily increasing. The biodegradability of natural fibers, as well as the fact that they are a renewable resource spread worldwide, is a significant advantage to these materials.<sup>1–4</sup> The main disadvantages of the use of natural fibers as a reinforcement element in composites are well established and discussed in the literature.<sup>2,5–8</sup> One can highlight, for example, the presence of a weak fiber–matrix interface as a common characteristic for these composites that can strongly reduce their applicability.<sup>7–10</sup>

Weak interfaces can be used to advantage, however, to increase the toughness of composites.<sup>11</sup> This is the particular case when sponge, or towel, gourd (*Luffa cylindrica*) is used in hybrid glass fiber–luffa polyester composites.<sup>12</sup> Luffa fibers form a natural mat that deviates the crack path, leading to a controlled fracture mode of a composite and increasing the composite's toughness. The use of luffa alone to reinforce a polyester matrix is, however, restricted by a very weak fiber–matrix interface. Therefore, surface treating

these fibers could be a valid alternative to enhance their use in composites.

In this work, the effect of the common mercerization treatment to improve the strength of a luffa fiber–polyester matrix interface is reported. The effects of temperature and concentration of the NaOH solution were studied, and their effects on the fiber surface morphology and the composite's flexural strength were analyzed and are discussed.

## EXPERIMENTAL

The sponge gourd is a subtropical plant abundant in China, Japan, and other countries in Asia as well as in Central and South America. Its fruit has a fibrous vascular system that forms a natural mat when dried. A study on the structure and morphology of the dried sponge fruits was reported elsewhere.<sup>12</sup> In this work, the dried sponges were washed in flowing water, dried, and cut into strips 120 mm long and 25 mm wide. The area weight of this mat was 0.078 g/cm<sup>2</sup> and the fibers' diameters typically varies from 270 to 400 μm.<sup>12</sup> These strips were treated before the fabrication of the composites.

Mercerization is one of the most used treatments for natural fibers.<sup>13–16</sup> It has been successfully applied to enhance fiber–matrix adhesion in such different systems such as jute–epoxy<sup>15</sup> and woodflour–unsaturated polyester resin<sup>16</sup> composite materials. It consists of the immersion of the fibers in a solution of sodium

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**TABLE I**  
Specification of the Variables Used in the  
Mercerization Treatment

| NaOH<br>(% in weight) | Temperature of treatment |       |
|-----------------------|--------------------------|-------|
|                       | 23°C                     | 100°C |
| 0                     | ✓                        |       |
| 1                     |                          | ✓     |
| 5                     | ✓                        | ✓     |
| 10                    | ✓                        |       |
| 20                    | ✓                        |       |
| 40                    | ✓                        |       |
| 60                    | ✓                        |       |

hydroxide, NaOH. What could vary in the treatment from one composite to another are the experimental setup of the treatment, namely, the time of immersion and temperature of the solution, and the NaOH concentration in the solution. One can find very different values for all these parameters in the literature. For example, the common treatment used for textile fibers is done for 1 h at ambient temperature using a solution of 40%, in weight, of NaOH. Table I shows the treatments used in this work, where one can see that only the concentration of the solution and the temperature were varied. The duration of the treatment was maintained constant for 1 h. After the treatments, the fibers were copiously washed in distilled water.

Composites were fabricated with a volume fraction of fibers of 30%, by placing the fibers inside a mold and casting the preformulated resin. The volume fraction of the fibers was measured using digital image analysis. This volume fraction was shown previously to give composites with a fracture mode already controlled by the fibers.<sup>17</sup> An orthophthalic polyester resin (Polylite T-208, Resana, Brazil) mixed with 2 wt % of a methyl ethyl ketone catalyst was used as the matrix. This resin was used as received, without any purification. The mold was subjected to a pressure of 13 MPa to aid in the removal of entrapped air bubbles and also to produce composites with uniform thickness. This fabrication procedure proved to produce composites with a low content of voids ( $V_v \leq 3\%$ ), as determined by analyzing the obtained microstructure using digital image analysis. The composite plates obtained were 120 mm wide and 120 mm long, with a thickness of 5 mm. The composites were cured at room temperature and were left at rest for 7 days before the machining of the test specimens. This procedure was adopted to guarantee uniformity on resin curing.

The effect of the treatments on the mechanical behavior of the composites was evaluated using the three-point bending flexure test, as recommended by the ASTM D790-00 standard. All tests were done at room temperature. Specimens 100 mm long, 25 mm wide, and 5 mm thick were tested under a constant crosshead speed of 1 mm/min with a span-to-depth

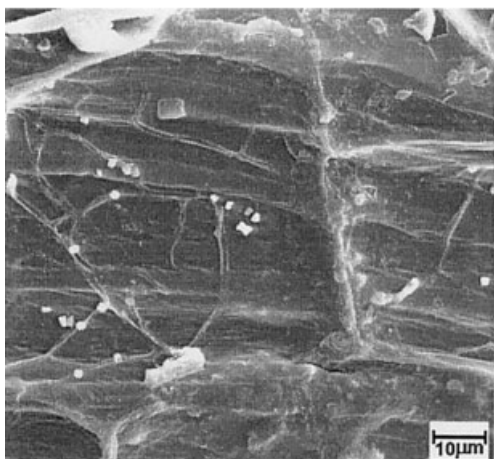
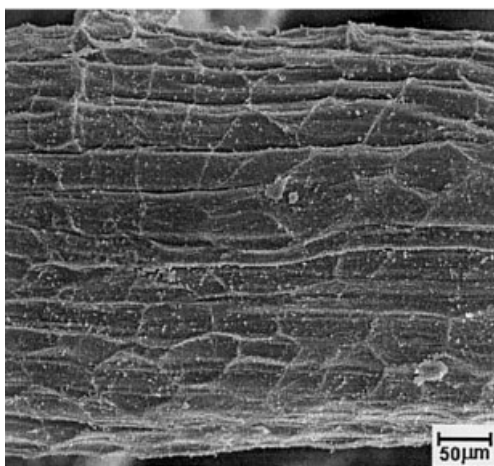
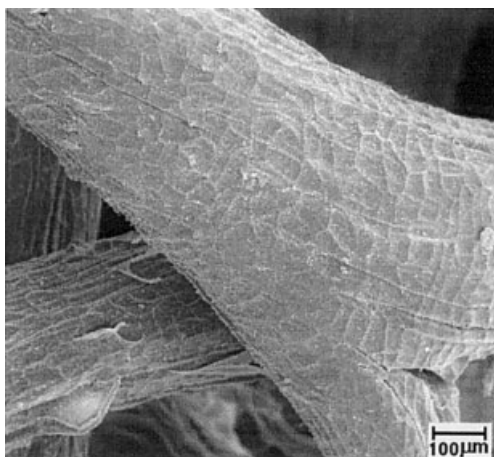
ratio of 16. At least five specimens were tested for each condition analyzed. The treated and untreated luffa fibers, as well as the fracture surface of the composites, were analyzed by scanning electron microscopy (SEM). The SEM analysis enables a detailed study of the fiber surface morphology and could be used to evaluate aspects related to the fiber–matrix interface interaction. This analysis was done on gold-sputtered specimens using secondary electron imaging and an electron-beam voltage ranging from 15 to 20 kV. Energy-dispersive X-ray analysis (EDS) was also performed on the surface of the untreated fibers.

## RESULTS AND DISCUSSION

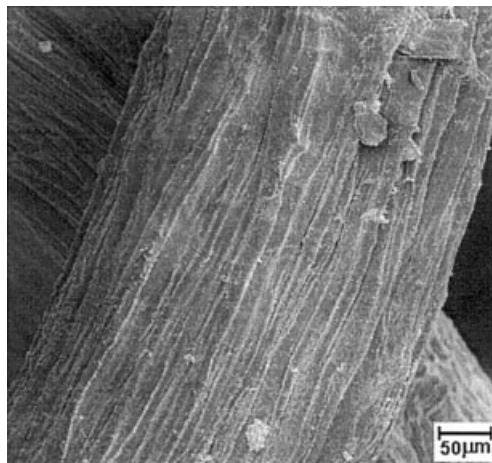
The surface morphology of untreated luffa is shown in Figure 1. Figure 1(a) highlights the natural ramification of these fibers, which produces a continuous fiber–matrix interface path. Figure 1(b) shows the characteristic cellular structure found at the surface of natural fibers. This outer surface is composed of parenchyma cells like, for example, sisal fibers.<sup>18</sup> From Figure 1(c), one can see the finer fiber structure inside a parenchyma cell. The white dots spread over the surface were identified by energy dispersive X-rays as potassium–calcium-rich structures. Similar features, although silicon-rich ones, identified as tyloses, were observed at coir fibers.<sup>13,19</sup>

Figure 2 shows the overall aspect of the treated fibers. One can see that the surface of the fibers was modified, with the removal of the outer cellular layer. The internal fibrillar structure of the fibers is exposed, therefore increasing the surface area for contact with the resin matrix. Although for the solutions with higher NaOH concentration a complete split of the fibers could sometimes be observed, the analysis performed only with SEM is not suitable to qualitatively identify which treatment promotes the better modification at the surface of the fibers. In fact, many of the topographic aspects observed are very similar for the different treatments performed.

The experimental results obtained for the composites with fibers treated at room temperature are shown in Table II. One can see that the treatments produced only a minor increase in the flexural strength, and the best results were obtained using the 5% NaOH solution. This same behavior was observed for the variation of the elastic modulus, as is also shown in Table II. The trace of the load-deflection curves of the composites (Fig. 3) also shows that the treatment with the 5% NaOH solution produces a different fiber–matrix interaction. As shown in Figure 3, the typical fiber-controlled fracture mode observed for untreated luffa composites [Fig. 3(a)] was also common to all treatments [e.g., Fig. 3(b)] except for the 5% one [Fig. 3(c)]. One can see in Figure 3(c) that a sharper rupture occurred, indicating that the propagating crack is not being effectively deviated from its path by weak in-



**Figure 1** Aspects of the untreated *L. cylindrica*: (a) general view, showing the natural ramification of the fibers; (b) cellular structure at the surface of the fibers; (c) one surface cell with characteristic white dots spread all over the cell surface.



**Figure 2** Alkali-treated fibers. General view showing that the outer surface layer was removed; 5% NaOH solution.

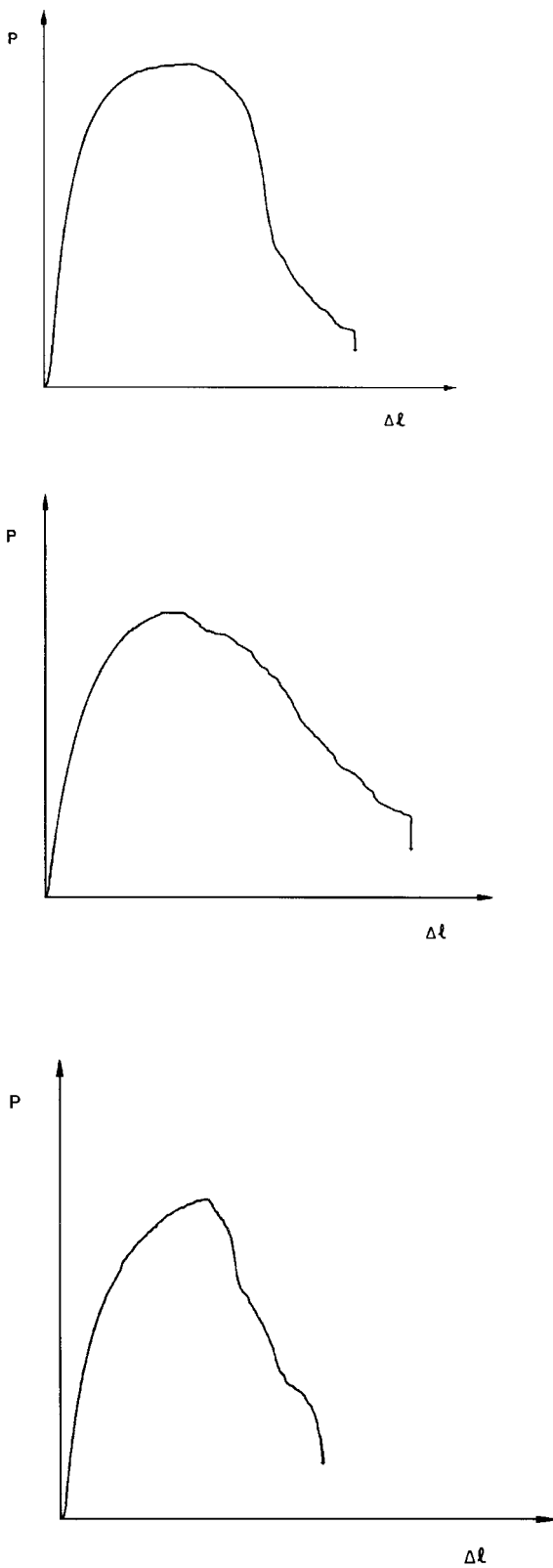
terfaces as in the other composites. It is clear, however, from the figures quoted in Table II, that the treatment used did not promote a strong interface, because, as said before, only a minor increase on the mechanical properties was obtained.

The analysis of the fracture surface of the composites corroborates the results presented above. As shown in Figure 4, the fracture surface of these composites is characterized by the presence of extensive fiber pullout, even for the composites fabricated with the luffa fibers treated with the 5% NaOH solution. Therefore, one can say that a weak interface is still the dominant parameter controlling the mechanical behavior of these composites. Nevertheless, as also shown in Figure 4, the fraction of fibers broken at the fracture plane is higher for the 5% NaOH treatment. As is well known, smaller pulled-out lengths are related to a better interface strength.<sup>11,20</sup>

As shown in Figure 5, completely broken fiber-matrix interfaces were observed for all the composites, which is clear evidence that the room-temperature treatment performed was not able to develop a proper chemical coupling between the luffa fibers and the polyester resin, as would be expected from theoretical arguments.<sup>9</sup> From the results obtained, it seems that only a physical modification occurs at the fiber sur-

**TABLE II**  
Flexural Mechanical Properties as a Function of Alkali Concentration—Room-temperature Treatment

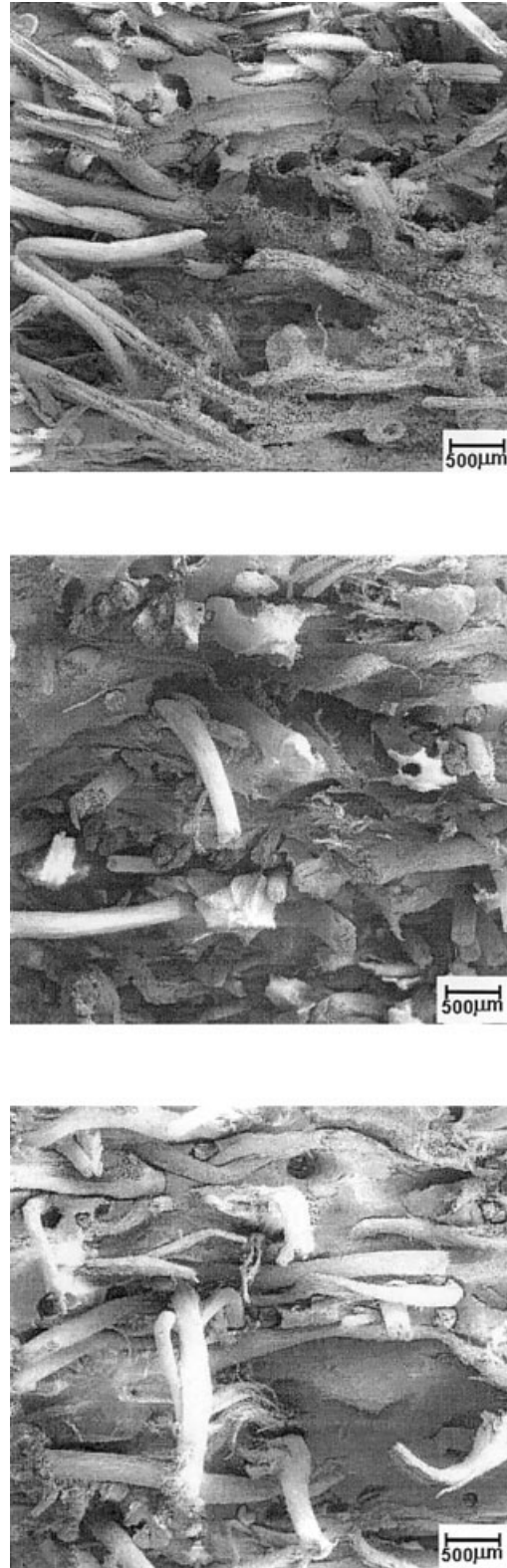
| NaOH (% in weight) | Modulus of rupture (MPa) | Elastic modulus (GPa) |
|--------------------|--------------------------|-----------------------|
| —                  | 41.6 ± 17.2              | 2.63 ± 0.91           |
| 5                  | 46.4 ± 10.3              | 3.22 ± 1.05           |
| 10                 | 41.1 ± 9.5               | 2.41 ± 0.48           |
| 20                 | 41.7 ± 11.1              | 2.51 ± 0.91           |
| 40                 | 29.8 ± 12.3              | 1.83 ± 1.01           |
| 60                 | 38.8 ± 3.2               | 2.30 ± 0.16           |



**Figure 3** Schematic load-deflection curves of the luffa-polyester composites: (a) untreated luffa fibers; (b) treated fibers, 10% NaOH; (c) treated fibers, 5% NaOH.

face. The slight increase on the mechanical properties obtained could, therefore, be due only to the increase of the fiber surface area, as shown in Figure 2, and the

increase of mechanical interlocking. In fact, a very good contact was developed between the treated fibers and the resin matrix, as revealed by the sharp



**Figure 4** Fracture surface of the composites showing, typically, long fiber pullout: (a) untreated fiber; (b) 5% NaOH; (c) 40% NaOH.

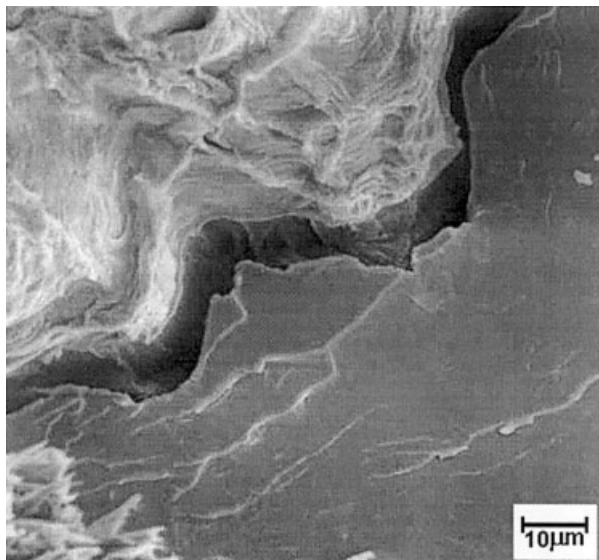


Figure 5 Common aspect of the luffa-matrix interface.

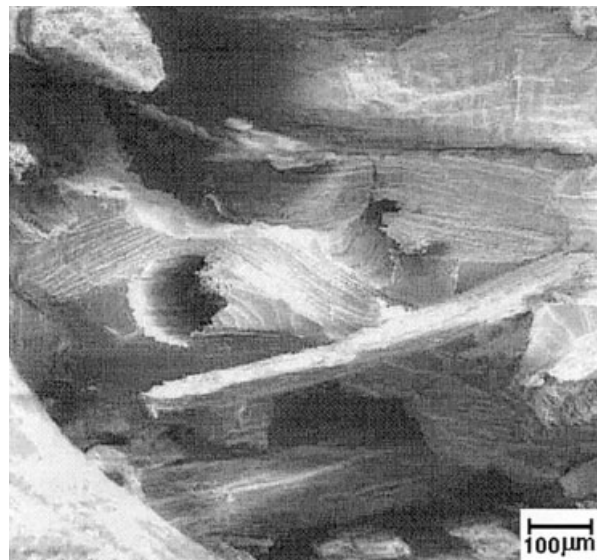


Figure 6 Common aspect of the fracture surface for the composites with fibers treated at 100°C. Pulled-out fibers and sharp fiber imprints left at the resin matrix are the two main features observed.

fiber beads observed at the fracture surface. These neat fibers imprints are, nevertheless, another sign of weakly bonded fibers.<sup>21</sup>

The results for the composites with fibers treated at 100°C are shown in Table III. Although only two different NaOH solutions were analyzed, a similar trend was obtained (i.e., only a secondary increase was obtained for the mechanical properties evaluated). The topographic features observed at the fracture surfaces of these composites were also similar to the ones found for the composites treated at room temperature. Figure 6 shows, as an example, a fractured specimen where fiber beads and pulled-out fibers can be seen.

Although the alkali treatment produced only a secondary increase on the flexural mechanical properties of this luffa-polyester composite, the values obtained for the flexural strength are encouraging, since they are on the same level of other mat or chopped fiber-reinforced natural fiber composites with a similar volume fraction of fibers, like coir-polyester (29–41 MPa),<sup>13</sup> jute-polystyrene (45–55 MPa),<sup>22</sup> and sisal-phenolic (23–64 MPa).<sup>23</sup>

CONCLUSIONS

Common to other natural fibers, the mercerization treatment produced strong morphological changes on

TABLE III  
Flexural Mechanical Properties as a Function of Alkali Concentration (100°C)

| NaOH (% in weight) | Modulus of rupture (MPa) | Elastic modulus (GPa) |
|--------------------|--------------------------|-----------------------|
| —                  | 41.6 ± 17.2              | 2.63 ± 0.91           |
| 1                  | 44.9 ± 7.3               | 2.52 ± 0.85           |
| 5                  | 47.1 ± 8.3               | 3.02 ± 1.14           |

the surface of luffa fibers. The outer surface of the fibers was completely removed, exposing the inner fibrillar structure. The solution with 5% NaOH provides composites with the best flexural mechanical properties. Nevertheless, only a secondary increase on the properties was obtained. This slight increase was attributed solely to mechanical interlock due to the increase of fiber roughness and the increase of the contact area of the fibers. The fractographic analysis corroborates the lack of a chemically bonded interface. The strength attained by these composites was, however, comparable to other common natural fiber-resin matrix composite materials, showing the feasibility of luffa as reinforcement to resin matrix composites.

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