

# Effect of Fiber Treatment on the Mechanical Properties of LDPE–Henequen Cellulosic Fiber Composites

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**ABSTRACT:** The degree of mechanical reinforcement that could be obtained by the introduction of henequen cellulosic fibers in a low-density polyethylene, LDPE, matrix was assessed experimentally. Composite materials of LDPE–henequen cellulosic fibers were prepared by mechanical mixing. The concentration of randomly oriented fibers in the composite ranged between 0 and 30% by volume. The tensile strength of these composite materials increased up to 50% compared to that of LDPE. There is also a noticeable increase in Young's modulus for the composite materials that compares favorably with that of LDPE. As expected, the addition of the fibers decreases the ultimate strain values for the composite materials. The thermal behavior of the LDPE–henequen cellulosic fibers materials, studied by differential scanning calorimetry, DSC, showed that the presence of the fibers does not affect the thermal behavior of the LDPE matrix; thus, the interaction between fiber and matrix is probably not very intimate. Preimpregnation of the cellulosic fibers in a LDPE–xylene solution and the use of a silane coupling agent results in a small increment in the mechanical properties of the composites, which is attributed to an improvement in the interface between the fibers and the matrix. The shear properties of the composites also increased with increasing fiber content and fiber surface treatment. It was also noted that the fiber surface treatment improves fiber dispersion in the matrix. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **65**: 197–207, 1997

**Key words:** fiber treatment; LDPE–henequen cellulose fiber composites; thermal treatment; mechanical properties; adhesion

## INTRODUCTION

The preparation of composite materials with different polymeric matrices and reinforcing fibers has received widespread attention lately. The main advantage of these composites lies in their relative low weight compared to their high mechanical properties and resistance to severe environmental conditions. These materials have a broad spectrum of applications as structural components. In the particular case of long-fiber composites, they have been used in a variety of struc-

tural applications, and the prediction of their expected mechanical properties at given orientations and configuration of the fibers has been extensively studied.<sup>1,2</sup> However, composite materials based on short fibers or particles still pose problems that make the prediction of their effective mechanical properties difficult. The most common problems in the prediction of mechanical properties arise from the uneven distribution and orientation of the fibers inside the matrix that are difficult to control by the processing methods commonly used in industry such as extrusion, compression, and injection molding of thermoplastic materials.<sup>3</sup> It has been proven that another important factor is the degree of adhesion

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between fiber and matrix, which plays an important role on the final mechanical properties of the composite material.<sup>3-6</sup>

An interesting alternative for reinforcing polymeric matrices with short fibers is the use of cellulosic fibers, which can be obtained from different renewable resources, mainly wood.<sup>7-14</sup> Micromechanical considerations point out that cellulosic fibers can act as a reinforcement for at least certain thermoplastic and thermosetting materials.<sup>14</sup> Other advantages of cellulosic fibers stem from their relative low cost, their tensile resistance, and elastic modulus, which on a per-weight basis compare favorably with those of other fibers such as glass, aluminum, and carbon and are more readily available than other organic fibers such as polyester, nylon, or kevlar. Moreover, the composites can be produced using the normal processing equipment used for thermoplastic polymers processing, without the problems such as machine wear that are the drawback of some other synthetic and metallic fibers.<sup>11,15-18</sup>

In this work the degree of mechanical reinforcement that can be achieved when cellulosic fibers obtained from henequen (*Agave fourcroydes*) are used in the preparation of composite materials with a low-density polyethylene (LDPE) matrix is reported. These studies span the reinforcement achieved when the reinforcing fibers are used without or with surface treatment as well as the effects that thermal annealing has on the composite material mechanical properties.

## METHODS AND MATERIALS

Henequen cellulosic fibers were extracted from henequen whole fibers through an alkaline digestion process followed by bleaching with sodium hypochlorite. The conditions for the cellulosic fibers extraction were reported before.<sup>19</sup> The physical properties of henequen cellulosic fibers have been reported recently.<sup>20</sup> Low-density polyethylene, LDPE, was obtained from PEMEX (20020) and used as received.

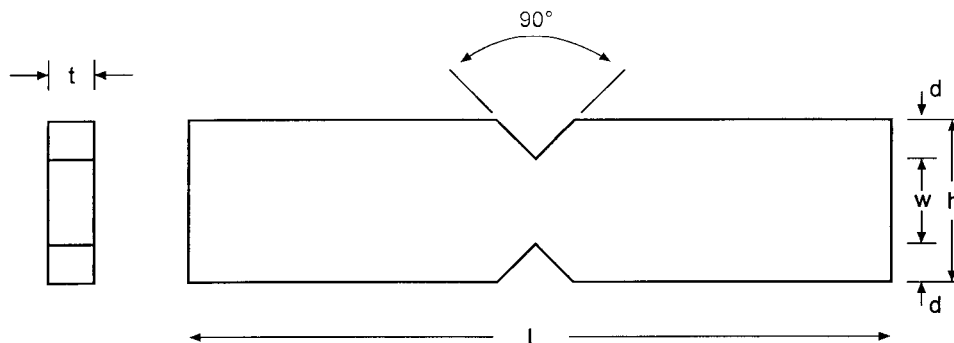
Three different types of surface treatment for henequen cellulosic fibers were used for the preparation of the composite materials. In the first case, the fibers were used without any treatment. A second surface treatment consisted of preimpregnation of henequen cellulosic fibers with a solution containing 2% by weight of LDPE in xylene at 130°C under reflux for 1 h conducted under nitrogen atmosphere. A third set of materials was

prepared from cellulosic fibers modified with a silane coupling agent (A-172 Union Carbide) applied at a concentration of 1% by weight in carbon tetrachloride at 70°C with dicumyl peroxide as the catalyst. Henequen cellulosic fibers were dried in a convection oven for 1 h at 105°C to eliminate their moisture content prior to mixing. The fibers treated with the silane coupling agent and LDPE solution were dried at 55°C under vacuum for 24 h. Mixing of the pure and treated cellulosic fibers with LDPE was carried out by mechanical mixing in a 60 mL Bambury mixing chamber (Plasticorder PL330, G.W. Brabender) at 160°C. The mixing process was performed in the following order. First, one-half of the total LDPE was placed inside the mixing chamber in about 1 min at 10 rpm; then, henequen cellulosic fibers were added over a period of 3 min. After this, the other half of the LDPE was placed inside the mixing chamber and the mixing speed was increased to 30, 50, 70, and 100 rpm in 1-min intervals, respectively, following a procedure similar to that suggested by Shen<sup>3</sup> for the mixing of cellulosic fibers and thermoplastic materials. Other mixing procedures were also tried, but the best fiber distribution was obtained using the method described above. The fiber distribution was assessed visually using an optical microscope. The concentrations of cellulosic fibers in the composite materials used in these experiments were up to 30 vol % of fibers.

The mixture of LDPE-henequen cellulosic fibers was laminated by compression molding into thin sheets 150 × 150 × 1.0 mm. The lamination was performed at 160°C in a laboratory press with temperature control (Carver Laboratory Press). The pressure used was 3000 psig for 5 min and then it was cooled down under pressure to 35°C in 5 min. After lamination, test coupons type IV were cut according to ASTM-D638-82a method. Iosipescu shear test samples were cut following the dimensions shown in Figure 1. All samples were tempered at 50°C for 1 h prior to mechanical testing unless otherwise stated.

Tensile properties were determined according to the norms of the ASTM-D638-82a method, using an Instron 1125 Universal Testing Machine equipped with a 500 kg load cell and a crosshead speed of 5 mm/min. The test were performed in at least five samples for each composite material with different concentrations of henequen cellulosic fibers with and without surface treatment.

Shear properties were determined using a Wyoming modified Iosipescu shear test fixture using the same loading rates as for the tensile tests.



**Figure 1** Iosipescu shear test coupons dimensions used in all shear tests are  $L = 76.6$  mm,  $h = 19.3$  mm,  $W = 12$  mm,  $t = 2$  mm, and  $d = 3.65$  mm. The notch radius was equal to 0.79 mm.

Due to the fact that the samples yielded under shear loads, it was necessary to precondition some of the samples at  $-80^{\circ}\text{C}$  prior to loading in order to analyze the failure surface at the interface. This preconditioning increased the yield strength of the matrix, thus allowing an interfacial failure rather than matrix failure. The observation of failure surfaces of shear test specimens was carried out by scanning electron microscopy, SEM, in a Zeiss DSM 940A scanning electron microscope.

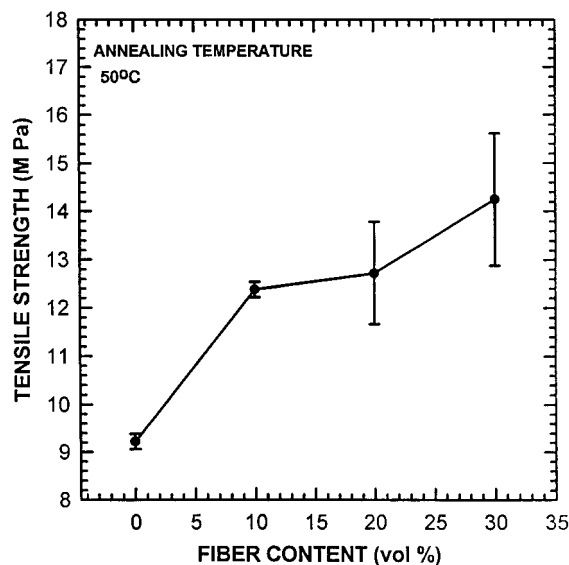
Thermal analysis was performed by differential scanning calorimetry in a DSC-2C (Perkin-Elmer Co.). The composite materials were scanned from 50 to  $200^{\circ}\text{C}$  at a rate of  $20^{\circ}/\text{min}$ . Annealing of the composite materials at 50, 60, 70, and  $80^{\circ}\text{C}$  for 1 h was performed to assess its effect on the LDPE-henequen cellulosic materials mechanical and thermal properties.

## RESULTS AND DISCUSSION

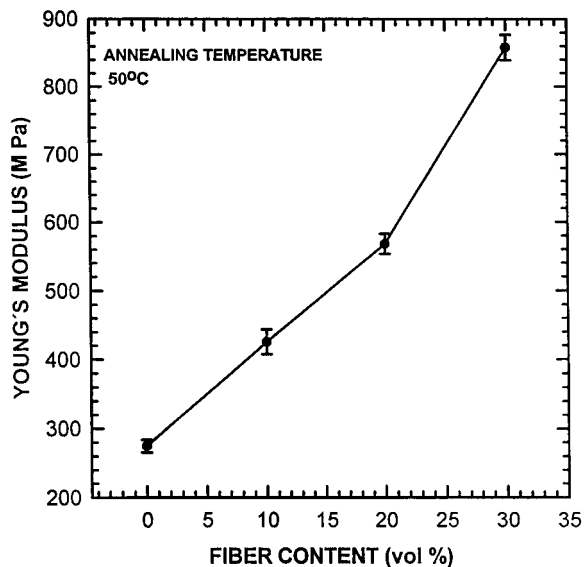
The degree of reinforcement obtained with the introduction of henequen cellulosic fibers in the LDPE matrix can be visualized by the tensile strength presented by the composite material (Fig. 2). The addition of cellulosic fibers increases the maximum tensile strength of the materials. The increase is also dependent on fiber concentration because the average ultimate stress increases as the fiber concentration increases for all concentrations tested here. It is also interesting to notice that the dispersion of the ultimate stress values becomes larger as the concentration of fibers increases in the material. This could be attributed to a nonhomogeneous distribution of the fibers inside the matrix. It should be pointed out that the obtention of a homogeneous fiber distribution

could be considered a problem when dealing with short-fiber composites. However, as it will be mentioned later, fiber surface conditioning plays a major role in getting a better dispersion of the fibers in the matrix.

The inclusion of henequen cellulosic fibers in LDPE also affects the Young's modulus of the material. As in the case of the ultimate stress, the modulus increases as the concentration of henequen cellulosic fibers increases in the composite material (Fig. 3). Composite materials with a 30% by volume of cellulosic fibers have a modulus that is, on the average, 200% larger than the values observed in LDPE. The inclusion of henequen cellulosic fibers also results in a decrease on the

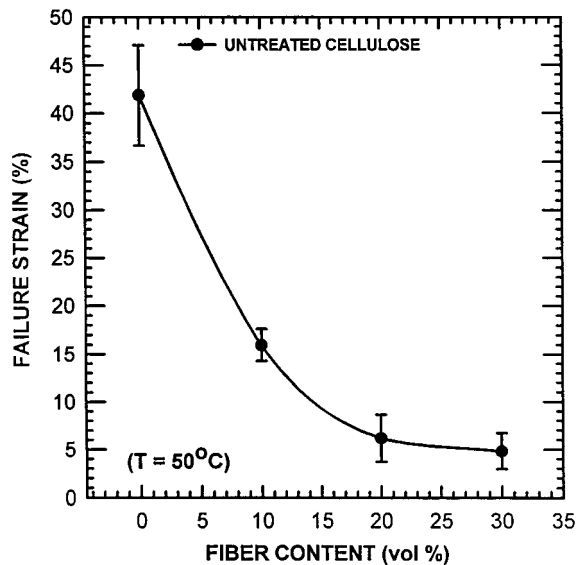


**Figure 2** Tensile strength of LDPE-henequen cellulosic fiber composites for several henequen cellulosic fibers contents.

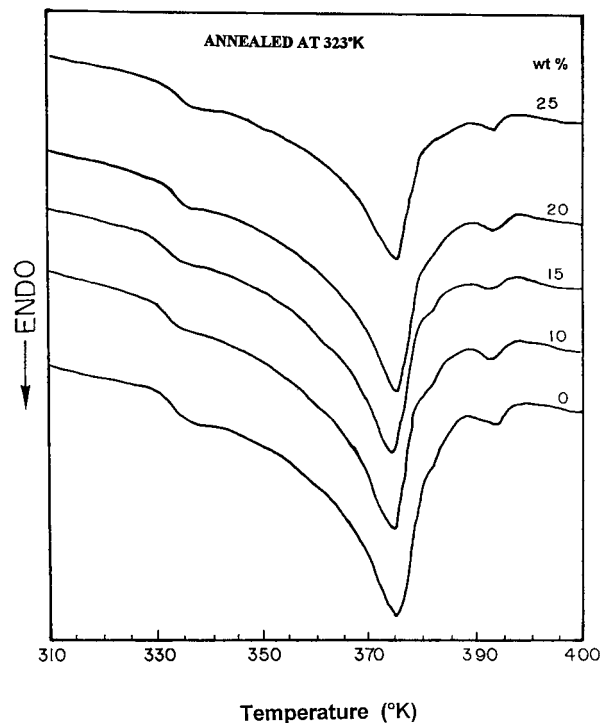


**Figure 3** Young's modulus of LDPE-henequen cellulosic fiber composites for several henequen cellulosic fibers contents.

failure strain of the LDPE composite materials. In Figure 4 it can be observed that the strain values decrease and that they do not show a large dispersion at high-fiber concentration as in the case of the tensile strength. It was also observed that as the fiber concentration increases in the composite material the strain decreases rapidly until it reaches an almost constant value.



**Figure 4** Failure strain of LDPE-henequen cellulosic fibers composites for several henequen cellulosic fibers contents.

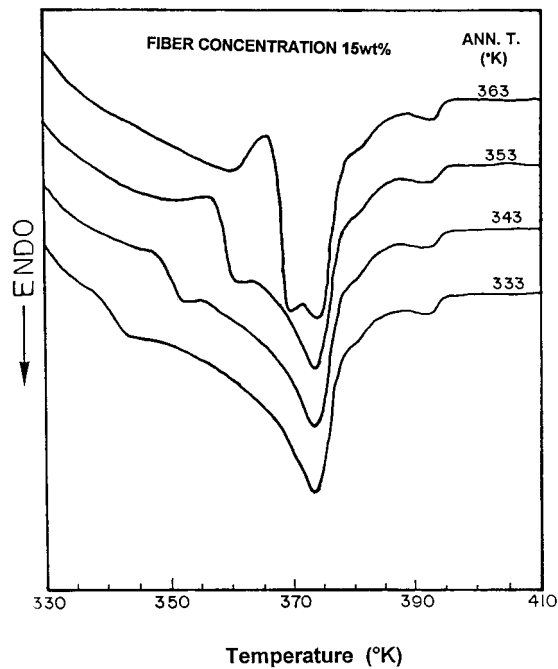


**Figure 5** DSC thermograms of the fusion of LDPE and LDPE-henequen cellulosic fiber composites.

#### Thermal Measurements

Thermograms of LDPE obtained by DSC place the fusion of this polyolefin between 330 and 400 K with a maximum in the endothermic peak at 375 K and a small shoulder around 395 K. The inclusion of henequen cellulosic fibers in the LDPE does not seem to affect the crystallinity of LDPE because no displacement of the initial peaks was observed in respect to the LDPE without fibers, as can be seen in Figure 5.

Annealing of LDPE or LDPE-henequen cellulosic fiber materials for 1 h at different temperatures did show some differences in the above observed behavior. The annealing of LDPE and LDPE-cellulosic fiber materials results in a crystallinity change that depends on the annealing temperature. As shown in Figure 6, when samples of any of the composite materials or LDPE alone are annealed at 323 K there appears a small inflection at 330 K in the endothermic peak. If the annealing temperature is 333 K, the inflection point moves to 338 K and there is an increase in the size of the inflection. When the annealing temperature moves to 343, 353, and 363 K, the inflection shoulder displaces to 346, 356, and 366 K, respectively. Moreover, the small inflection or



**Figure 6** Effect of annealing temperature on the endothermic transition of LDPE and LDPE-henequen cellulosic fibers composites.

shoulder that appears at an annealing temperature of 323 K, annealed at 363 K becomes a peak approaching the value of the maximum for fusion in the endotherm of LDPE. This suggests that a crystallinity change is induced in the LDPE as the annealing temperature increases. Based on the heat of fusion for LDPE and the composite materials, annealed for 1 h in the DSC under a nitrogen atmosphere, the changes in crystallinity of the annealed samples were assessed using the value for the heat of fusion for a fully crystalline PE, 69 cal/g, reported in the literature<sup>21</sup> and compared to the experimental values obtained from the DSC. Table I gives the crystallinity of LDPE annealed between 323 and 363 K (50 and 90°C, respectively) as evaluated from the heat of fusion. Annealing between 50 and 90°C of LDPE produces a decrease in crystallinity of 14% as assessed here.

It was also noticed that the heat of fusion, measured in cal/g, for the different LDPE-henequen cellulosic fibers composite materials diminishes as the concentration of fibers increases in the material. This result was expected, because, in a weight basis, there is less LDPE in the composite than in the pure polymer. The heat of fusion for LDPE-henequen cellulosic fiber materials de-

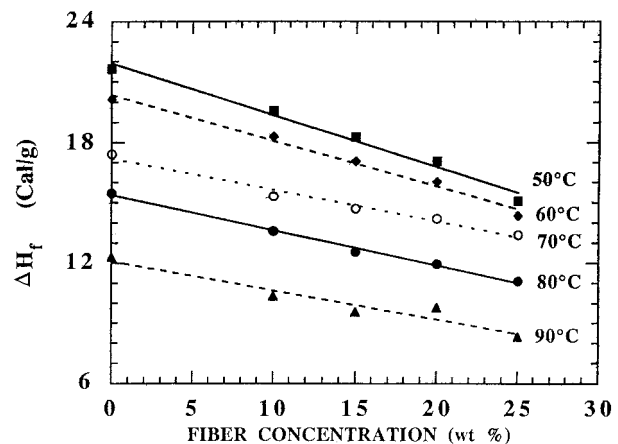
**Table I** Change in LDPE Crystallinity with Annealing Temperature

Annealing Temperature (°C)	$\Delta H_f$ (cal/g)	Crystalline Fraction
50	21.8	0.31
60	20.1	0.29
70	17.4	0.25
80	15.5	0.22
90	12.2	0.17

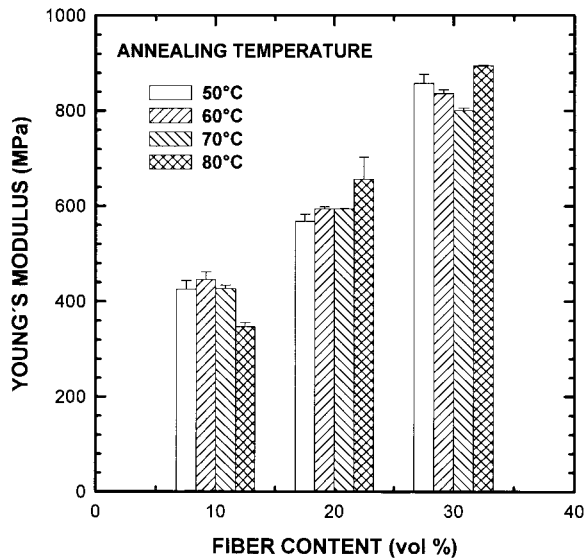
creases proportionally with the concentration of fibers following the form:

$$\Delta H_{fc} = \Delta H_{fp}(1 - x_f) \quad (1)$$

where  $\Delta H_{fc}$  and  $\Delta H_{fp}$  are the heats of fusion of the composite material and LDPE, respectively, and  $x_f$  is the weight fraction of henequen cellulosic fibers in the composite material. The behavior of the experimental heat of fusion for different concentrations of henequen cellulosic fibers, annealed between 50 and 90°C, according to eq. (1), is given in Figure 7. In this figure, the straight lines correspond to the values calculated from eq. (1), taking the  $\Delta H_{fp}$  of pure LDPE annealed at each temperature, and the symbols represent the experimental values that are an average of three experiments. It is seen that experimental values agree closely with the behavior expected from eq. (1). The above results imply that the presence of the cellulosic fibers in the LDPE matrix does not affect the ability of the polymer to crystallize and



**Figure 7** Variation of the heat of fusion for LDPE and LDPE-henequen cellulosic materials as the concentration of fibers increases in the composite.

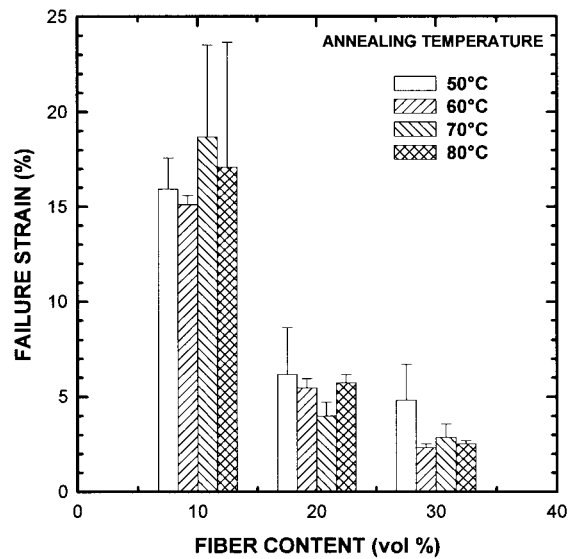


**Figure 8** Effect of annealing temperature on the elastic modulus of LDPE–henequen cellulosic fiber composites.

that the observed differences in crystallinity came from the thermal treatment imposed on the LDPE in the composites.

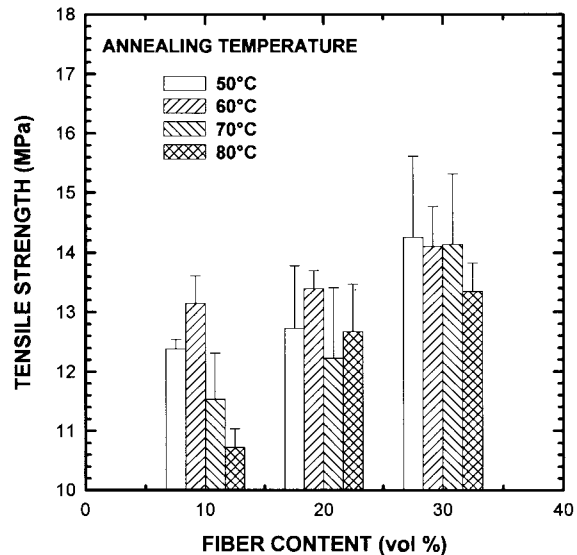
#### Dependence of Composite Properties on Annealing Temperature

The composites were compression molded at high temperature and then cooled rapidly to room temperature before testing. This thermal gradient suddenly imposed on the composite material induces large thermal stresses on the fiber as a result of contractions and change of morphology of the matrix upon cooling from the melt temperature to room temperature, that to a certain extent, complicates the state of stress at the fiber–matrix interface. The effect of thermal stresses upon the effective properties of the composite material is studied by controlled annealing of the specimens. Figure 8 shows the elastic modulus of untreated cellulosic fibers composites that were annealed at 50, 60, 70, and 80°C for 1 h. It is observed that only small variations on the modulus value are obtained for temperatures up to 80°C. For the failure strain, only small decrements are observed with increasing fiber content (Fig. 9). The effect of annealing is more pronounced on the tensile strength (Fig. 10), because for higher annealing temperatures and large fiber concentrations the tensile strength in general increases. However, the composites annealed at 80°C tend to have a



**Figure 9** Effect of annealing temperature on the failure strain of LDPE–henequen cellulosic fiber composites.

lower tensile strength. This result can be attributed to the change in morphology, loss of crystallinity in the matrix by the thermal treatment, although it is less than one MPa for the composites with the larger concentration of fibers. It should be remembered that the residual stresses resulting from the sudden cooling and the disparity in the thermal expansion coefficients between fiber and matrix results in compressive strains

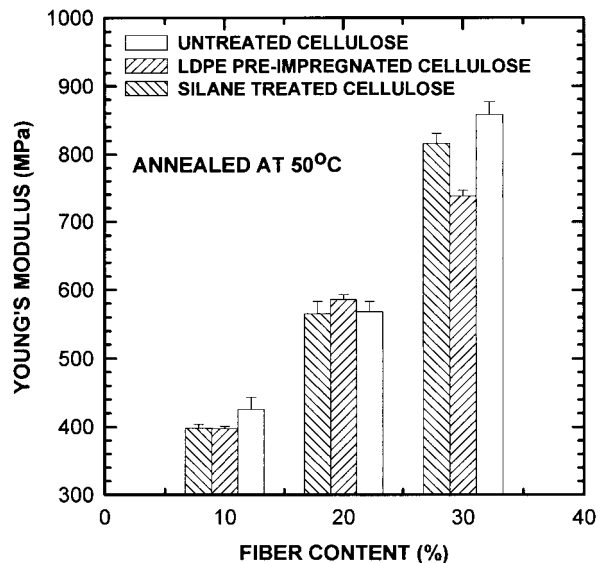


**Figure 10** Effect of annealing temperature on the tensile strength of LDPE–henequen cellulosic fiber composites.

upon the fibers. The magnitude of the residual strain due to the thermal mismatch can be estimated from the value of  $(\alpha_f - \alpha_m) \Delta T$  in which  $(\alpha_f)$  is the longitudinal thermal expansion coefficient of the fiber,  $(\alpha_m)$  is the thermal expansion coefficient of the matrix, and  $\Delta T$  is the difference between the annealing temperature and room temperature. It should also be noted that in this case the stress transfer from matrix to fiber is predominantly by frictional stresses. Thus, any modification of matrix morphology would modify the state of stress acting upon the fiber; therefore, affecting the fiber–matrix interface strength. It should also be noted that the annealing temperature improves the composite tensile strength with increasing fiber content.

### Effect of Interfacial Adhesion on the Composite Strength

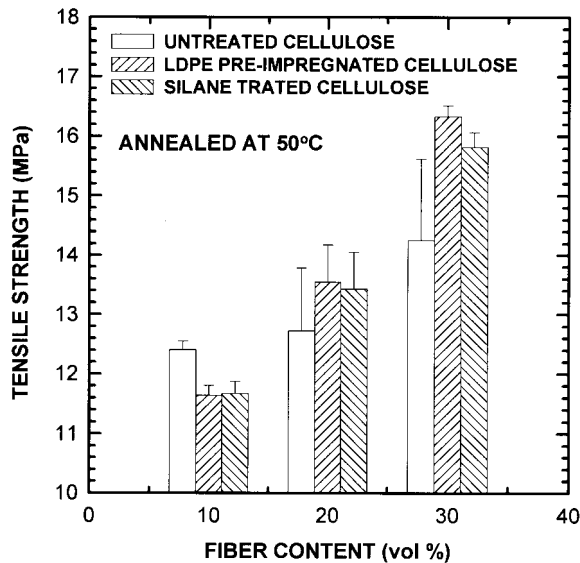
To study the effect of adhesion at the fiber–matrix interface, several approaches were taken. It is known that the hydrophilic cellulosic fibers have no adhesion to the hydrophobic thermoplastic matrix.<sup>22,23</sup> It is also known that the high viscosity of the matrix during composite fabrication hinders the proper fiber impregnation and, therefore, results in a poor fiber–matrix interaction. Then, the first approach was to perform a preimpregnation of the cellulosic fibers with the matrix material in solution. It is expected that this will result in an almost null-strain field after solvent evaporation, but there will be a better wetting of the fibers. A second approach for improvement of the fiber–matrix was the use of a silane coupling agent. Figure 11 shows a comparison of Young's modulus for untreated, LDPE pretreated and silane-treated cellulose fiber reinforced composites annealed at 50°C. Only some variations are observed at 30% by volume fiber concentration. The tensile strength of composites made with untreated cellulose fibers does not show a linear response with increasing fiber content. There is not a noticeable increase of strength for fiber contents between 10 and 20%, but at higher fiber contents there exists a more noticeable increase (Fig. 12). The fiber surface treatment seems to improve the materials tensile strength, and its behavior follows a linear behavior with increasing fiber content, and, the differences on tensile strength that are observed between both the preimpregnated and the silane-treated fiber are not considerable. On the other hand, it is noticed that the fiber surface treatment has an influence on fiber dis-



**Figure 11** Effect of different surface treatments of the cellulosic fiber on Young's modulus of LDPE–henequen cellulosic fiber composites.

persion in the matrix also. Such influence appears as lower experimental errors for the strength value as compared to those for the untreated fibers. Strain at failure values for both fiber surface treatments show a slight increase at all fiber contents (Fig. 13). Better results are found for the composites made with preimpregnated fibers.

It has been reported<sup>24</sup> that silane coupling agents increase the strength of the composite due to a better fiber–matrix adhesion. This is especially true for off-axis strength properties such as transverse tensile and flexural strength and shear strength. Observation of failure surface of Iosipescu shear load test samples give a good indication of the importance of fiber matrix adhesion on strength results. Figure 14 shows a series of scanning electron microscope photographs for composites tested under shear loads and for two different fiber contents (20 and 30% by volume). The main difference between the composites stems from the fiber surface conditions that were modified to yield different types of fiber–matrix interactions. In Figure 14(a), the untreated fibers appear to be free of any matrix material adhering to them. This is a clear indication of poor fiber–matrix adhesion. Also, there seems to be considerable matrix tearing. This reveals that considerable fiber pullout occurs for the untreated fibers. In the case of the LDPE preimpregnated fibers [Fig. 14(b)], there seems to be better fiber–matrix adhesion. Despite the fact that there is fiber

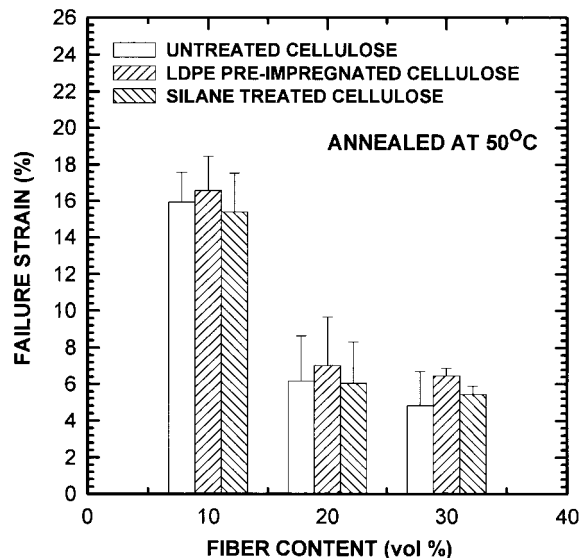


**Figure 12** Effect of different surface treatments of the cellulosic fiber on tensile strength of LDPE–henequen cellulosic fiber composites.

pullout, the fiber is coated with the matrix polymer. Another feature of this fiber treatment is on the matrix failure mode, because appreciable matrix failure by shear yielding rather than tearing is observed. It can be inferred that mechanical interlocking and friction are responsible for the observed composite strength increment. Figure 14(c) shows the failure surface for the composite where the fibers had been treated with the silane coupling agent. In this case, for both fiber content composites, it is possible to observe that the fibers are still coated with the matrix and that the matrix failed by shear yield flow and tearing from the fiber. If the matrix failure mode of the untreated fiber composite is compared to the preimpregnated fiber composite, it can be seen that it changes from a tearing mode to shear yielding. Then, from these matrix failure modes, it can be said that there exists a higher force transfer capability at the fiber–matrix interface in the composites with fibers treated by preimpregnation and a silane coupling agent. It should also be pointed out that the adhesion between fiber and matrix did not fail, when the fiber surface was treated by preimpregnation or the silane coupling agent, and that the limiting factor for composite strength, in this case, is the matrix yield strength. It is then reasonable to expect an upper bound in the strength increment for this fiber–matrix combination, especially for transverse and off-axis loading of the composite.

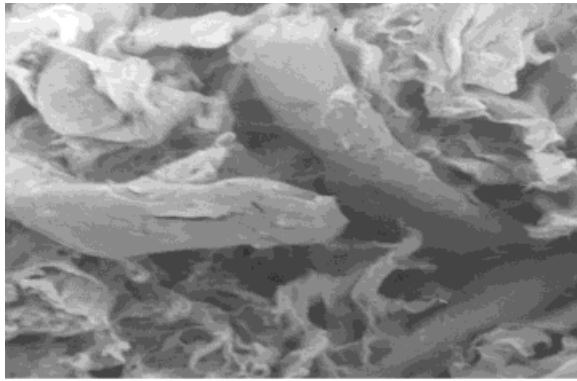
This qualitative assessment is confirmed by the results obtained in the Iosipescu shear load tests. Figure 15 shows the shear yield strength for untreated, preimpregnated, and silane-treated fiber composites with different concentrations of fibers. It can be seen that there is a small increment in the shear strength in the composites made with the surface-treated fibers, and also that at fiber concentration of 30% by volume there is a twofold increase in the shear yield strength of the composite material as compared with those that contain 10% by volume of cellulosic fibers. The shear yield strength increase follows a linear tendency with fiber concentration.

It has been shown for the untreated fibers that annealing of the composites has an effect on the composite mechanical properties. In the case of the preimpregnated fibers and silane-treated fibers, this thermal treatment enhances the effect of the strong fiber–matrix interaction. Figure 16 shows marginal effect on Young’s modulus of composites annealed at 60°C. In Figure 17, the strength increases with the thermal treatment for the two fiber-treated composites. This is especially noticeable for the composite with a 30% by volume of fibers. Again, slight changes in morphology seem to be beneficial to the strength values, and they improve noticeably with surface treatments at high-fiber concentrations. The failure strain, shown in Figure 18, increases for the surface-treated fibers due to the thermal treat-

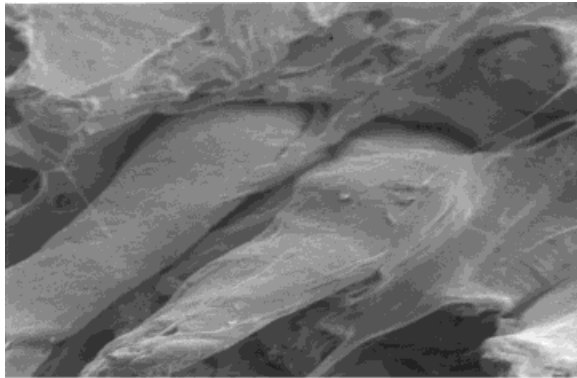
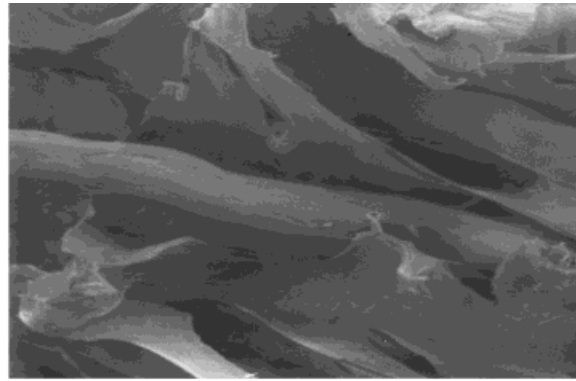


**Figure 13** Effect of different surface treatments of the cellulosic fiber on failure strain of LDPE–henequen cellulosic fiber composites.

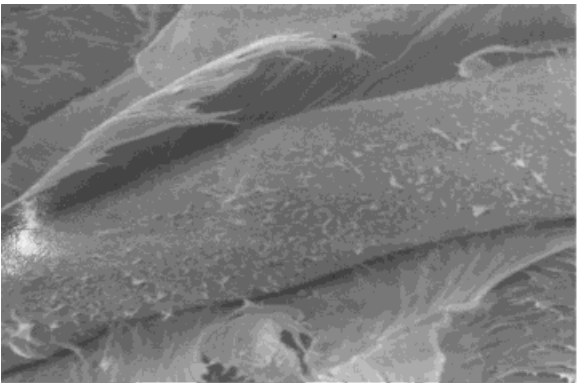
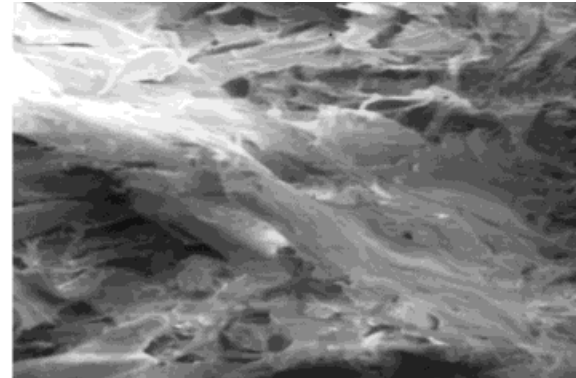




(a)



(b)



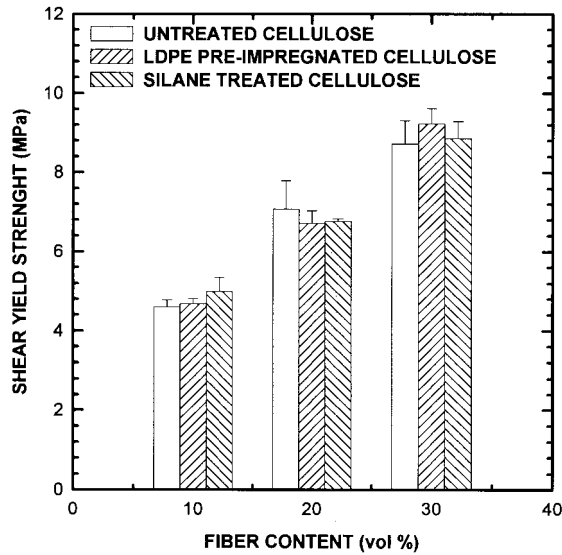
(c)

**Figure 14** Micrographs of the failure surface of shear test coupons for a LDPE–henequen cellulosic fiber composite with 20 and 30% by volume of cellulosic fiber. (a) Untreated cellulose; (b) LDPE preimpregnated cellulose; and (c) silane-treated cellulose.

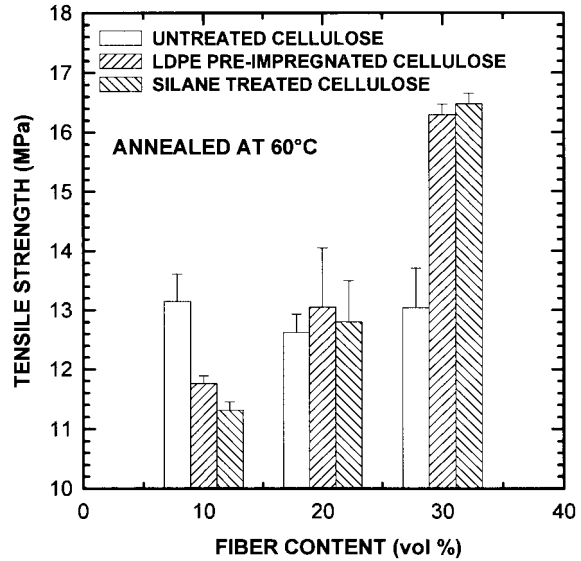
ment. This behavior can be explained in terms of an increase of adhesion between fiber and matrix induced by the morphological changes of the matrix. The reduction of crystallinity in the matrix improves the adhesion, especially when the cellulosic fibers had surface treatment.

## CONCLUSIONS

The mechanical properties of LDPE–henequen cellulosic fibers composite materials were assessed. The concentration of fibers in the composite material ranged between 0 and 30% by volume.



**Figure 15** Comparison of shear strength of LDPE–henequen cellulosic fibers composites with different treatments and as a function of fiber composition.

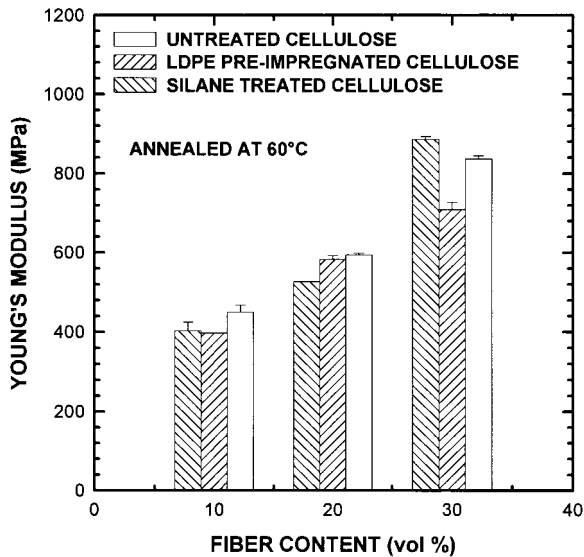


**Figure 17** Effect of annealing at 60°C on the tensile strength of LPDE–henequen cellulosic fiber composites with fibers subjected to surface treatment.

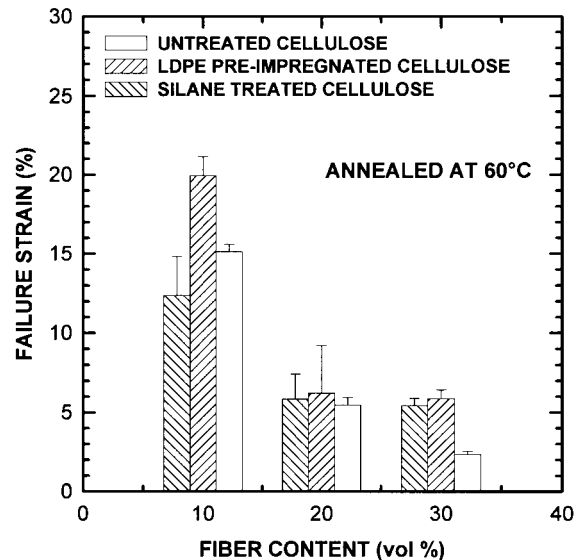
It has been shown that the inclusion of henequen cellulosic fibers in a LDPE matrix produces composites with superior mechanical properties. The tensile strength supported for the material increases up to 50% with respect to LDPE. The Young’s modulus also increases up to 200% at the largest concentration of cellulosic fibers tested. The ultimate strain diminishes as the concentration of fibers increases. There was also the prob-

lem of a large scatter in the mechanical properties measured experimentally as the concentration of cellulosic fibers in the LDPE increased. This scatter diminished considerably when the fibers were surface treated.

Thermal measurements showed that the inclusion of the fibers in the LDPE matrix does not affect the crystallinity of the material. Annealing



**Figure 16** Effect of annealing at 60°C on the Young’s modulus of LDPE–henequen cellulosic fiber composites with fibers subjected to surface treatment.



**Figure 18** Effect of annealing at 60°C on the failure strain of LDPE–henequen cellulosic fiber composites with fibers subjected to surface treatment.

at different temperatures results in a decrease of LDPE crystallinity. However, heat of fusion of LDPE is not affected by the presence of the fibers because it decreases linearly with concentration of henequen cellulosic fibers.

Iosipescu shear yield strength tests and scanning electron microscopy of the failure surfaces show that there is an improved adhesion between fiber and matrix when the cellulosic fibers are pre-impregnated with LDPE or treated with a silane coupling agent. Annealing of the composite also enhanced the effect of the level of adhesion between fiber and matrix.

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