

# Surface Modification of Henequen (*Agave fourcroydes*) Fibers by Ultraviolet Curing with 2-Hydroxyethylacrylate and Ethylacrylate: Effect of Additives on Degradable Properties

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**ABSTRACT:** Henequen fibers were grafted with a double impregnating monomer 2-hydroxyethylacrylate (HEA) and ethylacrylate (EA) to improve the physico-mechanical properties. The fibers soaked in different concentration (1–10%) of monomer + MeOH solution along with photoinitiator Irgacure 907 [2%] were cured under ultraviolet (UV) lamp at different UV radiation intensities (2–14 passes). Concentration of monomer at different radiation intensities was optimized with extent of mechanical properties such as polymer loading, tensile strength, and elongation at break. Enhanced tensile strength (268%) and elongation at break (110%) were achieved by the polymer treated fibers than untreated virgin fibers. We observed that, henequen fibers treated by 3% EA showed better physico-mechanical

properties than those treated by 5% HEA. The tensile properties of henequen fibers treated by 3% EA can be enhanced by adding aioxysilane; 3-(trimethoxysilyl) propylmethacrylate additives with bulk monomer EA (3%). The degradability of the treated and untreated fibers due to accelerated weathering were also studied and it has been found that surface modified henequen fibers produced more resistivity towards different weathering conditions than untreated fibers. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 4000–4006, 2006

**Key words:** Henequen fiber; HEA; EA; enhanced physical properties; radiation degradation; degradable properties; UV curing

## INTRODUCTION

Natural fibers have recently attracted the attention of scientists and technologists because of the advantages that these fibers provide over conventional reinforcement materials, and the development of natural fiber composites has been a subject of interest for the past few years. The natural polymer is biodegradable, readily and abundantly available, and eco-friendly. The synthetic polymer, in contrast, is nondegradable, durable, and sustainable in the environment, causing pollution after its end uses. Environmentally conscious people want to therefore return to the use of natural polymers wherever possible in place of man-made synthetic polymers to minimize pollution of the environment. The main advantage of the synthetic polymer over the natural polymer is that it can be made to the requirements of the users, while the characteristic properties of natural polymers are

essentially unalterable. It is, therefore, necessary to improve the characteristic properties of these natural polymers chemically, so that they are improved with respect to durability, sustainability, and mechanical strength, but will, at the same time, be degradable and eco-friendly. In an attempt to induce such properties in natural polymers, various physical and chemical treatments have been adopted since ancient times. As a result, natural hard fibers are gaining attention as a reinforcing agent in thermoplastic materials.<sup>1–3</sup> Low density and highly reduced wear of the processing machinery may be mentioned as attractive properties, together with their abundance and low cost. These natural hard fibers possess mechanical properties that make them suitable candidates to reinforce thermoplastic resin.<sup>1</sup>

Henequen (*Agave fourcroydes*) is a close relative of the sisal plant (*Agave sisalana*). Henequen hard fibers have been successfully incorporated into elastomers and thermoplastics.<sup>2,4,5</sup> Henequen is the fiber obtained from a plant indigenous to Yucatan, Mexico. Its long, silky, white, strong fibers contain 78% cellulose, 10% hemicelluloses, 8% lignin, 2% wax, and 11% moisture content. The cell length of the fiber is

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2.2 mm and has spiral angle of 20°. The specific gravity of the untreated Henequen fiber is 1.3 with tensile strength of 510 MPa, modulus of 28 GPa, and specific modulus of 22.<sup>6,7</sup> Grafting of vinyl monomer onto cellulose fibers by the radiation method is one of the most effective methods to improve mechanical properties and reduce the water uptake tendency.<sup>8-10</sup> The use of ultraviolet (UV) radiation to cure cellulose materials offers several advantages, such as (i) curing occurs at ambient temperatures, (ii) reduced reaction time, (iii) continuous operation, (iv) improved monomer stability, (v) less atmospheric pollution, and (vi) increased design flexibility through process control. The criticism of the grafting field has been that very few commercial processes have evolved from this large volume of research. The onset of radiation curing has saved this situation, since the presence of concurrent grafting with curing can markedly influence the properties of the resulting product. A most important aspect of the occurrence of grafting with curing is that such a specific type of grafting process possesses both advantages and limitations.<sup>11</sup> Thus, when curing of monomers onto naturally occurring macromolecules like cellulose materials, the presence of grafting during curing can lead to problems in recycling of the finished product.<sup>12</sup> 2-hydroxyethylmethacrylate (HEMA) was used as a grafting monomer to improve the mechanical properties of jute fiber, coir, and henequen.<sup>13-18</sup> Although increased tensile properties and decreased degradable properties can be obtained for HEMA-treated henequen fibers but we feel that further enhancements of physicomechanical properties can be improved by using other monomers like 2-hydroxyethylacrylate (HEA) and ethylacrylate (EA) and rapid environmental degradation can be reduced. The present study is therefore designed to limited polymer loading (lower than HEMA) within the minimum level in the preparation of HEA-grafted henequen/EA-grafted henequen by using UV radiation. The physical-mechanical properties of the HEA- and EA-grafted henequen fibers were compared and the effect of addition of additives with the polymeric formulations on the physicomechanical and degradable properties of treated henequen fibers were also studied.

## EXPERIMENTAL

### Materials

Henequen (*Agave fourcroydes*) fibers were collected from Mexico. Monomer 2-hydroxyethylacrylate (HEA) and ethylacrylate (EA) were procured from E. Merck (Germany). Photoinitiator, Irgacure-907 [2-methyl-1-(4-methyl thiophenyl-2-morpholino-propanone-1)], procured from Ciba-Geigy, Switzerland was incorporated into the formulation to initiate free

radical polymerization reaction. Additive 3-(trimethoxysilyl)propylmethacrylate was purchased from Fluka (Switzerland).

### Methods

Henequen fibers were cut into small pieces (10–15 cm in length). The samples were washed with water to remove foreign materials and dried in an oven at 105°C for 20 h to remove moisture and stored in a dessicator. Different formulations were prepared using (1–10 wt %) bulk monomer HEA and EA and 2% (w/v) photoinitiator (Irgacure 907) in methanol (MeOH) in different pyrex test tubes, each tube containing a total of 20 mL solution. The compositions of formulations are given in Table I. The dried samples were soaked in the monomer methanol solutions for different periods of time varying in the range from 5 to 20 min. The samples containing monomers were directly irradiated under UV light (model IST Technik, Germany) at 254–313 nm wavelengths of radiation and 2 kW power at 50 A current. The UV radiation source contains a conveyor belt that moves around the mercury lamp and after moving once to the lamp it was considered one pass. The radiated (at different radiation doses) samples were kept 24 h for polymerization reaction. After 24 h of irradiation, the samples were subjected to various characterization tests. The cured samples were extracted in hot benzene for 48 h to remove homopolymer and to determine the amount of grafting (polymer loading) of the monomer. Grafting percentage was measured by the formulae

$$(\%) \text{ Grafting} = [(W_t - W_0)/W_0] \times 100,$$

where  $W_t$  is the weight of the treated dry fiber and  $W_0$  is the weight of the untreated virgin fiber. Tensile properties such as tensile strength (TS) and elongation at break (Eb) of both grafted and untreated virgin fibers were measured by INSTRON tensile

TABLE I  
Composition of Different Formulations (% w/w)

Formulations	Monomer (%)	Methanol (%)	Photoinitiator (Irgacure 907) Additives (%)	
			(%)	(%)
1% HEA	1	97	2	–
3% HEA	3	95	2	–
5% HEA	5	93	2	–
10% HEA	10	88	2	–
1% EA	1	97	2	–
3% EA	3	95	2	–
5% EA	5	93	2	–
10% EA	10	88	2	–
3% EA	3	94.9	2	0.1
3% EA	3	94.8	2	0.2
3% EA	3	94.3	2	0.3

strength machine (model 1011, U.K.). The change in TS and Eb were expressed by tensile strength factor or tenacity factor ( $T_f$ ) and elongation factor ( $E_f$ ) that is,  $T_f = TS_t/TS_0$  and  $E_f = Eb_t/Eb_0$ , where  $TS_t$  and  $TS_0$  are the tensile strength of the grafted and untreated virgin samples,  $Eb_t$  and  $Eb_0$  indicates percent elongation of both treated and untreated coir fibers. Since henequen is a natural fiber, it has a tendency to rot due to water absorption, so water absorption property (water uptake) of both treated and untreated fibers were determined by placing the sample in a static water bath at room temperature, and water uptake for different swelling time were measured by the formulae

$$W_g = [(W_a - W_0)/W_0] \times 100$$

where  $W_g$  is the percent water uptake, and  $W_a$  and  $W_0$  are the weight of the samples before and after water treatment, respectively. The weight loss and the loss of the tensile properties by the impact of accelerated weathering test was performed at an alternating cycle of sunshine, dews and condensation with the help of an accelerated weathering tester (model Q.U.V, Q-Panel USA) for a period of 300 h. The cellulose possesses the tendency to degrade when buried in soil (having at least 25% moisture). For this purpose, treated and untreated henequen fibers were weighed individually and buried in soil for 72 days. After this period, samples were withdrawn carefully, washed with distilled water, dried at 105°C for 20 min, kept at room temperature for 24 h, and reweighed. Finally, the weight loss of various degraded samples and the loss of tensile properties were determined.

## RESULTS AND DISCUSSIONS

In the present investigation, the physicochemical properties of henequen fibers were improved by UV radiation through graft copolymerization using the bulk monomer HEA and EA. The effects of variation of radiation doses, monomer concentration and soaking time were evaluated with the extent of polymer loading, tensile properties, and water absorption behavior of the treated fibers. The degradable properties due to accelerated weathering and soil were also investigated. Most of the data presented in this report are average values of at least 10 samples, and the results obtained are within  $\pm 2\%$ .

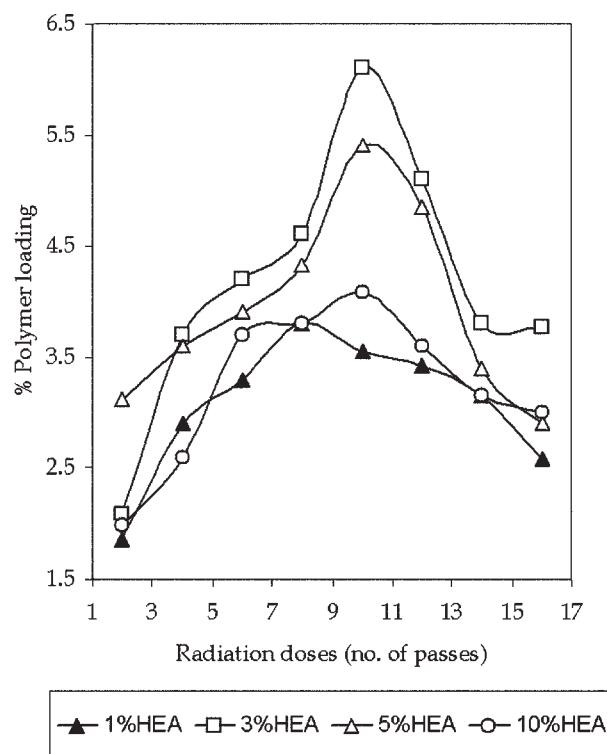
### Optimization of grafting condition for monomer HEA

#### Polymer loading

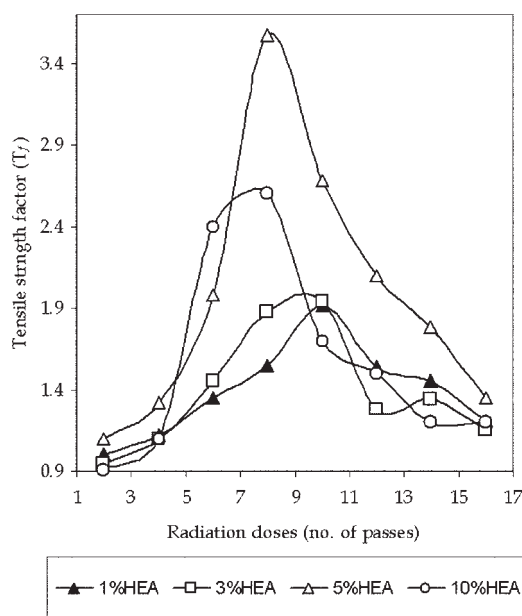
Polymer loading (PL) of the grafted henequen fibers were determined at different HEA concentrations

(1–10%) soaked for 10 min and irradiated at various UV doses (represented by number of passes). PL values are graphically represented in Figure 1 against UV doses as a function of monomer concentration. We observed that PL values for HEA cured samples increases with radiation doses up to a certain level and after attaining maximum values it started to decrease. From the figure, we have observed that maximum PL is obtained when the henequens were cured 10 passes of UV radiation. The PL values decrease for the samples cured with radiation doses more than 10 passes. The decreases of PL values at higher radiation doses (more than 10 passes) could be caused due to the radiation degradation of polymer at higher UV doses.<sup>18</sup>

On the other hand, we observed that the PL values for 1% HEA in methanol-treated fibers are relatively low and it increases as the monomer concentration in methanol are increased to 3% but again decreases for 5% HEA or more in methanol. The highest PL value (6.11%) was obtained by the sample treated with 3% HEA after 10 passes of UV radiation. At concentration lower than 3% the PL is less because vinyl monomer promotes rapid propagation reaction with the help of photoinitiator leading to network polymer structure through graft copolymerization reaction via their double bonds.<sup>16</sup> As monomer concentration is raised to 3%, the amount of residual



**Figure 1** Polymer loading (%) of grafted henequen fiber concentration against radiation doses (no. of passes) as a function of HEA concentration.



**Figure 2** Tensile strength represented by tensile factor ( $T_f$ ) of grafted henequen fiber concentration against radiation doses (no. of passes) as a function of HEA concentration.

unsaturation is also increased with the consequence of faster rate of formation of three-dimensional network structure causing restricted mobility. For more HEA (5% or more) in methanol decreases the PL values, which could be associated with the fact that the homopolymerization reaction between monomer and monomer-radicals is dominant than that of the monomer and fiber cellulose reaction.

### Tensile properties

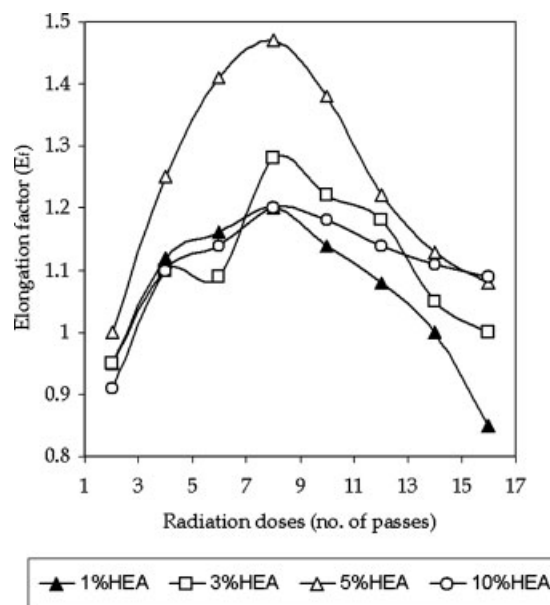
Tensile properties of the fibers are expressed by two factors, tensile strength factor ( $T_f$ ) and elongation factor ( $E_f$ ), which is the ratio of tensile strength and elongation at break of untreated and treated henequen fiber. The tensile strength of untreated fiber was found to be 350 MPa and elongation at break was found at 17%. The values of  $T_f$  and  $E_f$  are shown in Figures 2 and 3, where  $T_f$  and  $E_f$  are plotted against number of passes as a function of monomer concentration. Figure 2 showed that the tensile strength increases with radiation doses and after attaining maxima further increase of radiation dose decreases the  $T_f$  values. The highest tensile strength ( $T_f = 3.57$ ) was obtained after 8 pass of radiation by 5% HEA-grafted fiber. It has been observed that the highest tensile strength was obtained by the sample having PL value of 5.4%, while the highest PL containing sample yielded much lower tensile strength ( $T_f = 1.94$ ). The decrease of  $T_f$  at higher PL value could be due to the formation of excess three-dimensional crosslinked structure in the grafting zone of the cellulose, which form insubstantial polymer prod-

uct that decreases the tensile strength of the treated fibers. The enhancement in elongation due to polymer treatment of henequen fibers are depicted in Figure 3. The highest elongation ( $E_f = 1.47$ ) is obtained by the same sample that yielded the highest tensile strength.

From the discussion above, it is evident that highest  $T_f$  and  $E_f$  values are attained at 8 passes of radiation with 5% monomer (HEA) concentration. So this condition is considered optimum for henequen fibers treated with HEA.

### Optimization of grafting condition for monomer EA

Henequen fibers are soaked in different concentration (1–10%) of EA + MeOH solution and irradiated under UV radiation with different intensities. The values of PL,  $T_f$ , and  $E_f$  are given in Table II as a function of EA concentration at different UV intensities. We have observed that the highest PL (6.18%) is achieved by the samples treated with 3% EA at about 10 passes of UV radiation and after attainment of maximum PL, it started to decrease as described in the previous section. Studying the tensile properties of the treated fibers we have found that TS increases with UV doses up to 10 passes irrespective of monomer concentration, attain maxima, and then decreases with UV doses. Decrease of TS at higher radiation may be caused by two factors; at higher radiation doses, bombardment of monomer increases and many of them may initiate homopolymerization rather than grafting, thus increasing the homopoly-



**Figure 3** Elongation at break represented by elongation factor ( $E_f$ ) of grafted henequen fiber concentration against radiation doses (no. of passes) as a function of HEA concentration.

**TABLE II**  
**Different Physicomechanical Properties of EA-Grafted Henequen Fiber at Different UV Radiation Doses (Represented By Number of Passes)**

Properties	No. of pass	1%	3%	5%	10%
		EA + MeOH	EA + MeOH	EA + MeOH	EA + MeOH
% PL	2	1.98	2.88	3.20	2.10
	4	2.20	3.54	4.66	2.45
	6	2.35	4.8	5.32	3.50
	8	2.88	5.53	<b>6.01</b>	4.45
	10	<b>3.20</b>	<b>6.18</b>	5.50	<b>5.22</b>
	12	2.90	4.69	3.29	4.75
	14	1.99	2.8	1.98	4.6
$T_f$	2	1.40	1.48	2.15	1.80
	4	1.68	1.66	2.48	2.30
	6	1.86	1.85	3.37	2.90
	8	1.88	2.38	<b>3.52</b>	<b>3.14</b>
	10	<b>2.78</b>	<b>3.68</b>	2.78	3.08
	12	1.68	2.54	1.96	2.16
	14	1.35	1.64	1.80	2.04
$E_f$	2	0.90	0.98	1.18	0.90
	4	0.98	1.14	1.24	1.10
	6	1.1	1.28	1.36	1.28
	8	1.18	1.46	<b>1.52</b>	<b>1.37</b>
	10	<b>1.25</b>	<b>1.57</b>	1.42	1.29
	12	1.09	1.33	1.30	1.24
	14	1.0	1.24	1.19	1.12

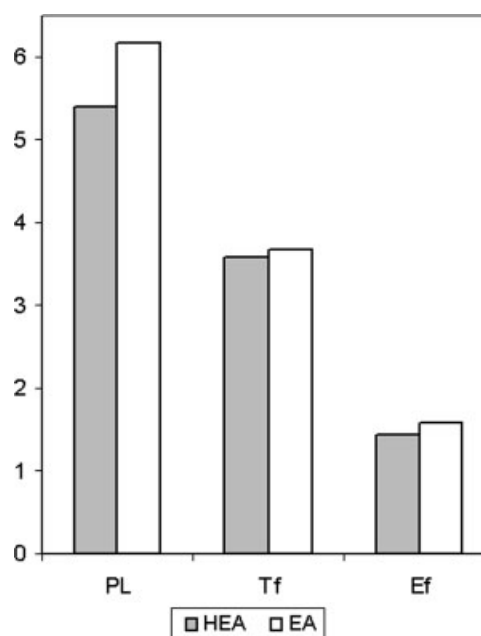
mers. The second factor could be the degradation of cellulose occurs at higher UV doses, which change the morphological structure of cellulose and decreases the strength.<sup>19</sup> However, the highest TS ( $T_f = 3.68$ ) is obtained from the sample treated with 3% EA at 10 passes of radiation. At higher EA concentration, radical-radical recombination processes may be dominating, creating more homopolymer rather than monomer-cellulose backbone reaction. It could also suggest that the swelling of the cellulose backbone in methanol monomer solution is insufficient due to low methanol concentration. This may cause a number of reacting sites at the cellulose backbone and would thus continue to reduce the active sites. The enhancement of elongation was found to maximum for the sample treated with 3% EA after 10 passes of radiation and the minimum is given by the sample treated with 1% EA.

Figure 4 shows the comparison of the physical properties between 5% HEA-grafted and 3% EA-grafted Henequen fibers and we have observed that better physicomechanical properties were obtained by the sample treated with 3% EA monomer at 10 passes of UV radiation dose that is soaked for 10 min in monomer methanol solutions. This condition is taken as optimum for further studies on henequen fiber including inclusion of additives into the formulated solutions.

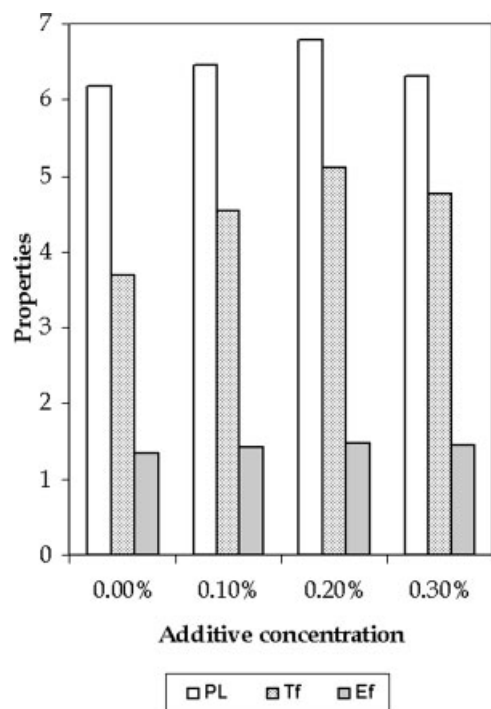
#### Effect of additives on physicomechanical properties of henequen fiber

Henequen fibers were treated with 0.1–0.3% of aloxysilane; 3-(trimethoxysilyl)propylmethacrylate incor-

porated into 3% EA containing solution as shown in Table I. The henequen samples were impregnated into the formulations having additive silane and cured with 10 passes of UV doses (which was optimized for monomer EA) and the properties of the cured samples were measured. PL and tensile properties are graphically presented in Figure 5 with respect to additive concentrations. We have observed



**Figure 4** Comparison of physical properties between HEA-grafted henequen fibers and EA-grafted fibers.



**Figure 5** Effect of additive concentration on the properties of grafted henequen fibers.

that addition of 0.2% additives to the monomer solution increases  $T_f$  value of 20% solution and  $E_f$  value up to 25%, while PL increases only 10% from the parent value. This additional increase of physical properties could be caused by the fact that alkyl-siloxane undergo hydrolysis, condensation, and bond formation reaction to form silanol. Silanol undergoes polysiloxane through polymerization reaction. The OH groups of polysiloxane, reacts with the OH groups of cellulose back bone through condensation reaction that produces silanized-henequen complex. The resulted three dimensional network structures lead to higher mechanical properties of the fibers.

### Water uptake

Water uptake by the treated and untreated henequen samples was monitored at 25°C. The results are shown in Figure 6 against soaking time (minutes) in water. All the samples attained maximum water uptake within initial 30 min and then soaking of water stops for silanized sample, while the untreated sample still continues to soak water slowly. The untreated samples showed the highest water uptake of 35.5%, the silanized treated sample yielded a value of 17.2, and 3% EA treated sample showed 19.6% water uptake after 80-min soaking in water. The reduced water uptake by the treated samples is due to the fact that the monomer and additive reacts with the OH group of cellulose through graft copolymerization reaction and hence reduces the hydro-

philic nature of the henequen fiber and also polymer fills the void space of the treated fiber.

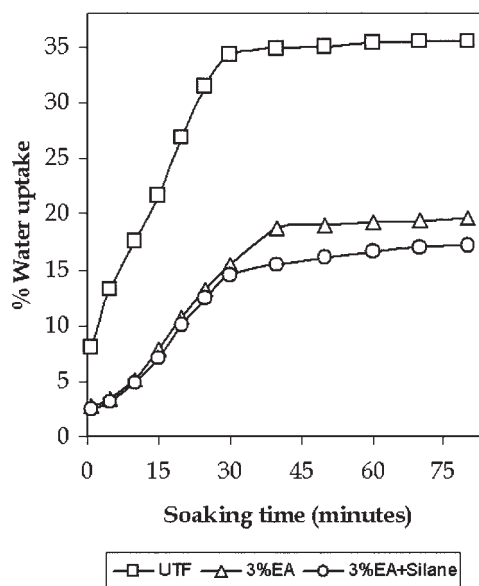
### Degradable properties of grafted henequen fibers

#### Simulating weathering effect

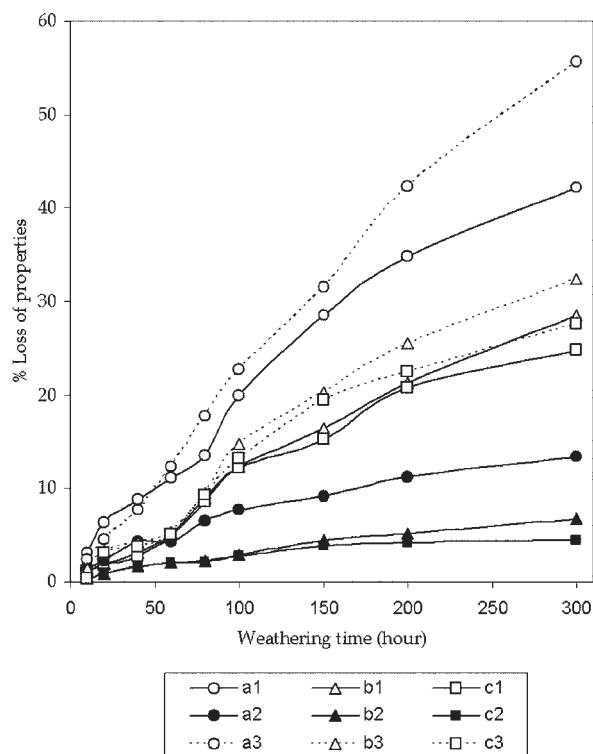
Untreated henequen fiber, 3% EA treated fiber, and the fiber grafted with 0.2 silane along with 3% EA were subjected to accelerated weathering tester over a period of 300 h to study the degradation properties. The weight loss and the reduction of tensile properties particularly TS and Eb of the samples were periodically monitored and are shown in Figure 7. TS loss for untreated sample was found to be 13.5%, while EA-grafted sample showed 6.8% and the sample treated with silane additives yielded minimum loss of tensile strength. We have also observed that the minimum loss of all of these three properties due to severe weathering treatment was from the sample that contains silane.

### Soil degradation

Untreated Henequen fiber, 3% EA-treated fiber, and the fiber grafted with 0.2 silane along with 3% EA were buried in soil (~ 25% moisture) for a period of 10 weeks to study the effect of such environmental condition on the degradable properties. We have observed that untreated fiber showed maximum loss of all properties (23% wt. loss, 57% TS and 68% Eb loss), while the fiber treated with silane additives yielded minimum loss (12.5% wt. loss, 28% TS and



**Figure 6** Water uptake of treated and untreated fibers against soaking time.



**Figure 7** Loss of physical properties due to degradation by simulated weathering treatment over a period of 300 h (a1 = % loss of weight of untreated fiber, b1 = % loss of weight of EA treated fiber, c1 = % loss of weight of additives treated fiber, a2 = % loss of tensile strength of untreated fiber, b2 = % loss of tensile strength of EA treated fiber, c2 = % loss of tensile strength of additives treated fiber, a3 = % loss of elongation of un treated fiber, b3 = % loss of elongation of EA treated fiber, c3 = % loss of elongation of EA treated fiber).

29% Eb loss) and the samples grafted with only 3% EA showed little more loss of properties than that showed by additives containing samples.

## CONCLUSIONS

Henequen fibers were modified with 2-hydroxyethylacrylate (HEA) and ethylacrylate (EA) by photocuring method using UV radiation. Henequen fibers treated with EA + MeOH solution showed enhanced physical properties than that treated with HEA + MeOH solution. Furthermore addition of aloxysilane; 3-(trimethoxysilyl)propylmethacrylate with 3% EA solution enhanced tensile properties and PL while reduces degradable properties of the henequen

fibers. These processes are independent of temperature and catalyst, and required a very small amount of monomer (3%). Under these processes the degree of polymerization can be controlled and formation of homopolymer is very low. These are very clean, safe, and fast, as well as reliable and space saving processes. Different surface pretreatments can also increase the mechanical properties without compromising the degradable character of the natural fibers. This is the significant improvement of henequen fiber obtained by using an easy, fast, and pollution free photo-curing method. These fibers could be used in diverse fields.

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