Effect of Pretreatment With Detergent on Mechanical Properties of Photocured Coir (*Cocos nucifera*) Fiber With Ethyleneglycol Dimethacrylate

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ABSTRACT: Coir fibers were treated with ethylene glycol dimethacrylate (EGDMA) mixed with methanol (MeOH) under UV radiation. A series of solutions of different concentrations of EGDMA in methanol along with a photoinitiator, Irgacure-500 (mixture of 1-hydroxylcyclohexylphenylketone and benzophenonc), were prepared. Monomer concentration, soaking time, and radiation intensity were optimized in terms of polymer loading (PL) and mechanical properties. EGDMA (50%), 5 min soaking time at the 4th pass of radiation, produced higher PL and tensile strength (TS), and the values of PL is 17% and TS is 1.3 times of the nontreated one. Then, coir fiber was pretreated with detergent and then treated with the optimized monomer formulation, which exhibited a higher PL of 69% and produced TS of the coir fiber of 4.4 times of the nontreated one. Coir fiber pretreated with detergent along with UV radiation showed the highest TS, which is 18.2 times of nontreated one. Water uptake, degradable properties, and simulated weathering of treated and virgin fibers were also monitored, which showed that EGDMA treatment under UV radiation improved the degradable property. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 1630–1636, 2006

Key words: coir fiber; EGDMA; UV radiation

INTRODUCTION

Both the natural and synthetic polymers are of great importance for industrial uses. The main advantage of synthetic polymers over natural polymers is that they can be made to the requirement of users, whereas the characteristic properties of natural polymers are essentially unalterable.[1] With the advent of synthetic fibers, the use of natural fibers has diminished to such an extent that many industries dealing with natural fiber products have closed. There has been a renewal interest in utilizing natural fiber, driven in part by environmental concern. However, natural fibers are generally inferior to their synthetic analogues in most respects, requiring some modification to realize performance benefits.[2] The three main groups of lingocellulosic fibers are bast fibers, leaf fibers, and seed-hair fibers. Coir is one of the most common and well-known natural fibers on the earth. Coir is the seed-hair fiber obtained from the outer shell, or husk, of the coconut, the fruit of *Cocos nucifera*, a tropical plant of the Arecaceae (Palmae) family.[3] The cost of coir fiber is comparatively low compare to others. Coir is mainly composed of cellulose (43%), lignin (45%), hemicellulose (0.25%), pectin, and related substances (3%). Cellulose is a hydrophilic glucan polymer consisting of a linear chain of 1,4-β-bonded anhydroglucose units that contains alcoholic hydroxyl groups.[4] The entire natural fiber is hydrophilic in nature.[5] High moisture absorption by the natural polymer materials is one of the reasons for their short life.[6] Various treatments have been used to improve the properties of natural fibers.[7,8] Graft copolymerization is a novel technique for the development and improvement of cellulose and cellulosic material.[9,10] It was reported in the previous study that treatment of natural fibers with various monomers of different functionalities under UV irradiation improves their physical properties.[11–15] Ethylene glycol dimethacrylate (EGDMA) is one of the difunctional monomers that could be cured under UV radiation.

The present article reports on the improvement of the physical and mechanical properties of coir fiber by immersing it in formulated solutions made from EGDMA and then curing under UV radiation. The mechanical properties of treated coir fibers were further improved by pretreating the coir with detergent and UV radiation before grafting with the optimized monomer solutions.

EXPERIMENTAL

Materials

Coconut and detergent, surf excel (Lever Brothers Bangladesh Ltd.), were supplied locally. EGDMA and methanol was procured from Merck (Germany). Acetone was purchased from BDH (UK). The photoinitiator Irgacure-500 was obtained from Ciba-Geigy (Switzerland).

Methods

The husks of the coconut were broken mechanically and fibers were extracted from the husks by hand, the samples were soaked in acetone for half an hour to remove of extraneous matter from the fiber's surface. The washed samples were dried at 105°C for 2 h to remove moisture and then cooled in desiccators. The samples were then weighed by means of an electronic balance. The samples were stored in desiccator. A series of solutions were prepared with different proportions (10, 20, 30, 50, and 70%) of monomer and EGDMA in methanol with 2% photo initiator; the dried samples were soaked in the different solutions for various times (3, 5, 10, and 30 min). The soaked samples were irradiated with a minicure-200 UV lamp (1st-techink, Germany). The lamp (254–313 nm) has 2 kW light intensity, the conveyor speed was 4 m/min, and the minicure-200 has efficiency within 11%. Several passes were required to ensure the complete curing of monomer onto the coir fiber. Before treatment with monomer, a separate batch of virgin fibers was also washed with 5% detergent for 2, 3, and 4 h. After 24 h, the cured samples were washed with acetone for 5 min for the removal of unreacted monomer from the coir fiber's surface. Then the samples were dried in an oven at 105°C for 2 h and cooled in a desiccator. Polymer loading (PL) was calculated as %PL = $(W_t - W_0)/W_0$ where W_t and W_0 are the weight of the treated dry sample and the virgin sample before any treatment respectively. The treated samples were then used to determine the tensile properties (tensile strength, TS, and elongation at break, E_b) using an INSTRON machine (model 1011, UK). The gauge length was 20 mm and the crosshead speed was 20 mm/min. The change in TS and elongation at break (E_b) were expressed by a TS factor (T_f) and elongation at break factor (E_f), where $T_f = TS_t/TS_0$ and $E_f = E_{bt}/E_{b0}$, TS_t and TS_0 are the TS of the treated and virgin fiber respectively.

Water uptake

Water uptake was measured by soaking the fibers for about 30 min in water contained in a static bath at 25°C. Weights of the samples were recorded after different contact times. Samples were wiped carefully with tissue paper to remove surface water before weighing. The weight gain for various samples was calculated as $W_g = [(W_a W_o)/W_o] \times 100$, where W_g is the percentage of weight gained by the sample due to water absorption and W_a and W_o are the weight of the sample after and before water treatment respectively.

Simulating weathering effect

A simulated weathering tester (model Q.U.V.) from Q-PANEL CO., USA, was used to weather the treated and virgin fibers. Both virgin and treated samples were exposed to serve weathering testing over a period of about 300 h of simulated sunshine and condensation of alternating cycles.

Degradation study

Both treated and virgin fibers were buried in unsterile soil (15% water) for a period of 105 days to study the effect of such environmental conditions on the degradability of the samples. The samples were removed from the soil at every 15 day intervals, washed with running water, and finally with acetone for the removal of soil. These samples were dried in oven at 105°C for 2 h. The loss of weight and tensile properties were measured.

RESULTS AND DISCUSSION

Effect of EGDMA treatment on PL

Studies of non treated coir fiber revealed its comparatively lower mechanical properties like TS and E_b. A Suitable monomer could play an important role to improve the properties of the coir fiber. A series of formulation were prepared in methanol in the presence of a photoinitiator, lrgacure-500. Effect of the concentration of EGDMA and variation of soaking time in the formulation were studied. PL values of the treated coir fiber were recorded at different monomer concentrations (10, 20, 30, 50, and 70%), which were irradiated under UV radiation with a different number of passes (2, 4, 6, 8, and 10 passes). These results are presented in Figure 1. Effect of soaking time on PL in an EGDMA solution is presented in Figure 4. PL values were recorded for the 50% EGDMA solution for different soaking time (3, 5, 10, and 30 min) after UV irradation. PL values were increased sharply at the initial irradiation time, after which the PL values decrease with further increase of radiation time and monomer concentration. The highest PL value (17%) was obtained at 4th pass of radiation, 50% monomer concentration and 5 min soaking time. The decrease of PL values after the attainment of the maximum PL values could be caused by radiation degradation at higher UV doses.[16] The decrease of the PL value at
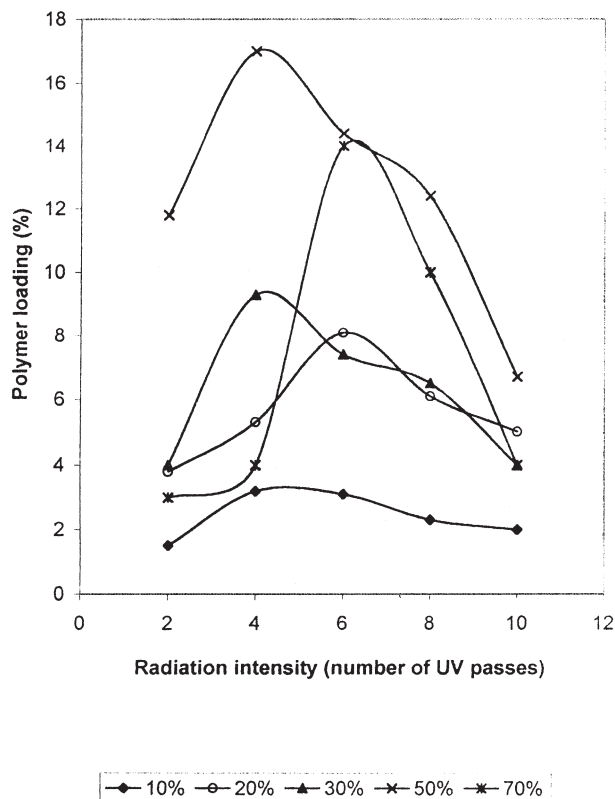
Figure 1 PL (%) of treated coir fiber against UV radiation intensities with respect to EGDMA concentration for 5 min soaking time.

higher monomer concentration could be associated with the fact that the homopolymerization reaction between monomer and monomer-radicals is dominant compared with that of the monomer and coir fiber and is due to the low MeOH concentration, leading to insufficient swelling of the fiber, resulting in lower level of impregnation and the swelling of the cellulose backbone with MeOH is insufficient.[17]

Effect of EGDMA treatment on mechanical properties

Effect of the concentration of EGDMA and variation of soaking time in the formulation on TS and elongation at break were studied. In each case, coir fiber was treated after soaking in the monomer solution. T_f and E_f were used to express the change of tensile properties. Figure 3 shows the effect of EGDMA treatment of coir fiber under UV irradiation at 5 min soaking time on TS and Figure 5 shows the effect of soaking time in 50% EGDMA solution. E_f for both treatment are presented in Figures 4 and 6. It is observed that T_f and E_f values increased with the monomer concentration and doses of radiation. The maximum T_f (1.3) and E_f (1.5) were obtained at the 4th and 6th pass of radiation respectively, and after attainment of the maximum

values, T_f and E_f are decreased. This could have been caused by the damage to the samples by the higher radiation dose. The decrease in TS with the increase in EGDMA after the 50% EGDMA concentration may take place for various reasons, such as at higher EGDMA concentration, the fibers became brittle and the rate of radical–radical combination and recombination reactions between them (radical) were also different from that of the optimum conditions.[18] T_f increase with soaking time because increased swelling led to increased diffusion of monomer into the sites of reaction and a greater amount of grafting, hence T_f values are increased.

Effect of pretreatment of coir fiber by detergent and UV radiation

After soaking in acetone, the virgin fibers were washed with 5% detergent for 2, 3, and 4 h in a breaker. Then, they were washed with running water and dried in an oven for 2 h (loss of weight was 5%). Some detergent-treated samples were treated with monomer and passed under 2, 4, 6, 8, and 10 UV passes. Some detergent-treated samples were also treated under UV radiation (10th UV passes) and soaked in optimized monomer concentration and their PL and mechanical properties are shown in Table I. The irradiated samples were washed with acetone
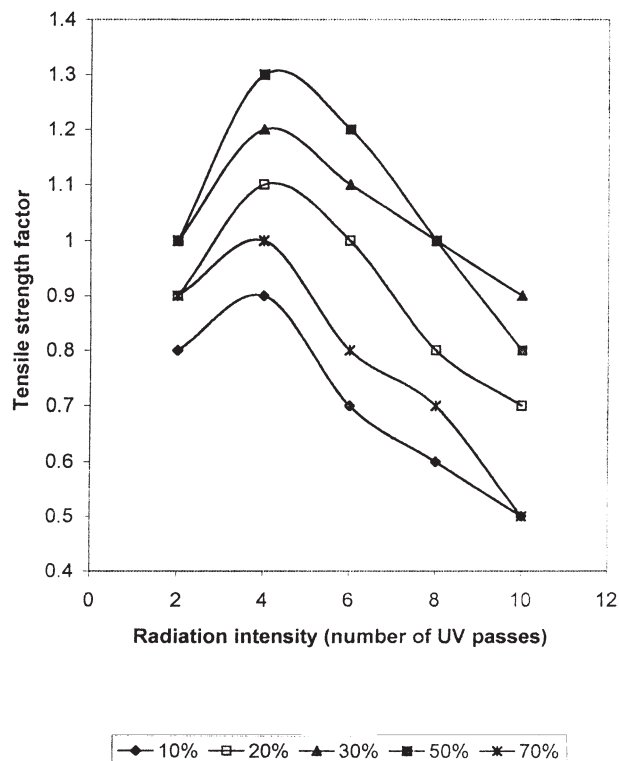
Figure 2 T_f of treated coir fiber against UV radiation intensities with respect to EGDMA concentration for 5 min soaking time.
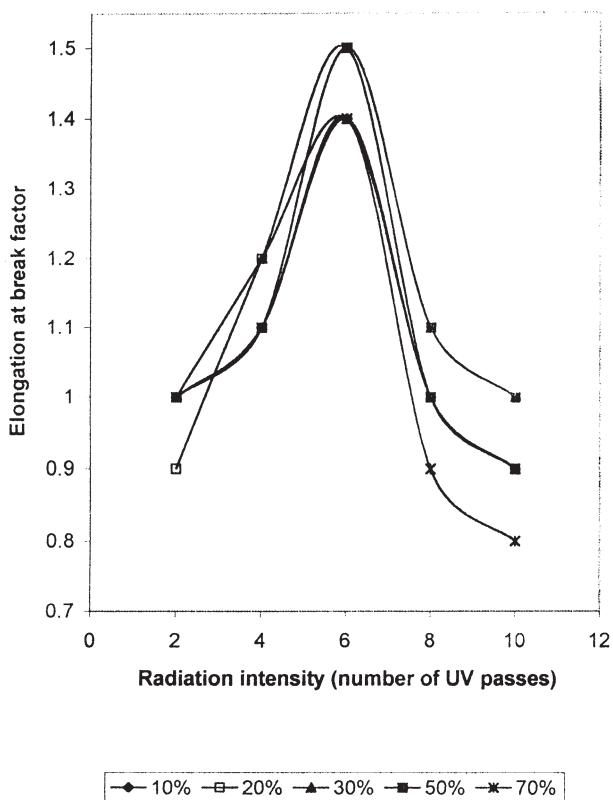
Figure 3 E_f of treated coir fiber against UV radiation intensities with respect to EGDMA concentration for 5 min soaking time.

Figure 4 PL (%) of 50% EGDMA onto coir fiber against UV radiation intensities with respect to different soaking time.

after 24 h for the removal of unreacted monomer from the fibers surface. The T_f and E_f of treated fibers were also measured.

Effect of pretreatment of coir fiber by detergent on PL

PL values of pretreated coir fiber (with detergent) against a number of UV passes with respect to different washing times (0, 2, 3, and 4 h) are shown in Figure 7. The maximum PL value (69%) was obtained at 3 h washing time at the 4th pass of radiation. Detergent-treated fibers are also treated with 10 UV passes and this gave a PL (7%) at 6th pass of radiation. Because of the time lapse, the long-lived radical sites are a major factor in the initiation of graft polymerization. This technique leads to high graft yields and longer branches. Detergent molecules can aggregate in water into spherical clusters called micelles. The hydrocarbon parts of the molecules gather together on the inside of the micelle and the polar groups are on the outside 000 compounds are often dissolved into center of the micelle attracted by the 000 groups. This process is known as solubilization.[19] Extent of PL is not commensurable with increase of T_f and E_f, because in
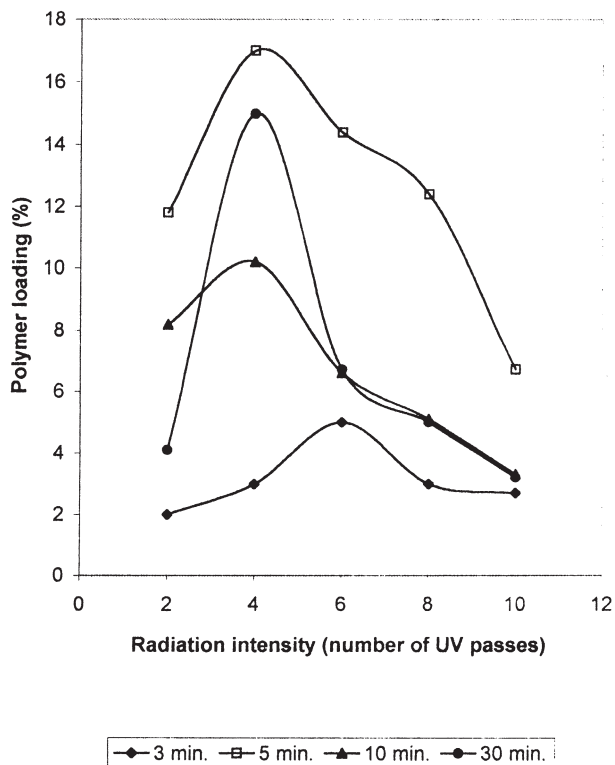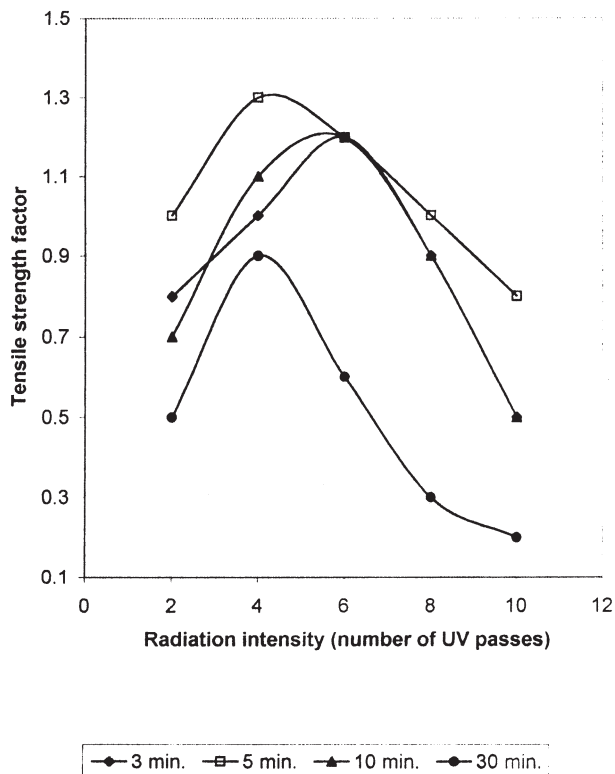

Figure 5 T_f of 50% EGDMA onto coir fiber against UV radiation intensities with respect to different soaking time.
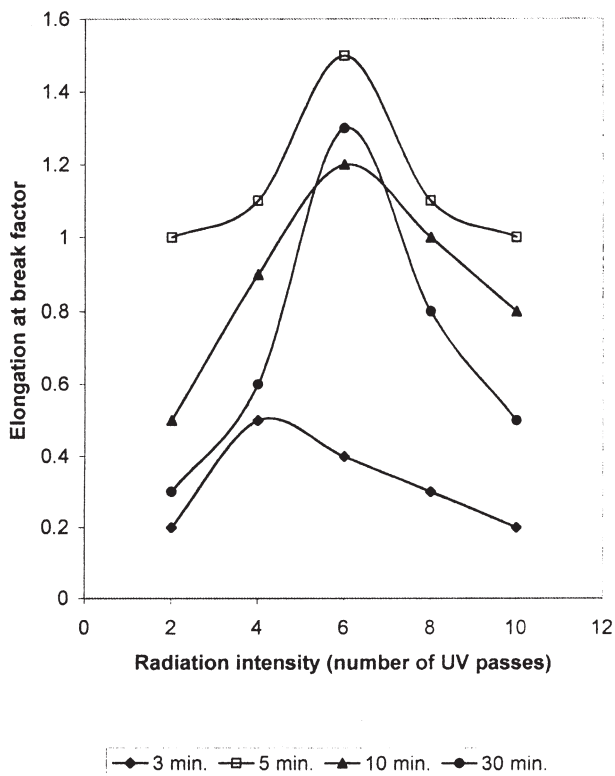
Figure 6 E_f of 50% EGDMA onto coir fiber against UV radiation intensities with respect to different soaking time.

some cases, PL may occur without forming any linkages between cellulose and monomer.[20]

Effect of pretreatment of coir fiber by detergent on mechanical properties

Coir fibers were pretreated with detergent and treated with optimized monomer concentration. It is observed from the Figures 8 and 9 that maximum T_f (4.4) and E_f^2 were obtained at 3 h washing time. Detergent molecules are made of a long oil-soluble chain and a polar salt-like head soluble in water.[21] When fibers were soaked in detergent, the oil-soluble tail in detergent
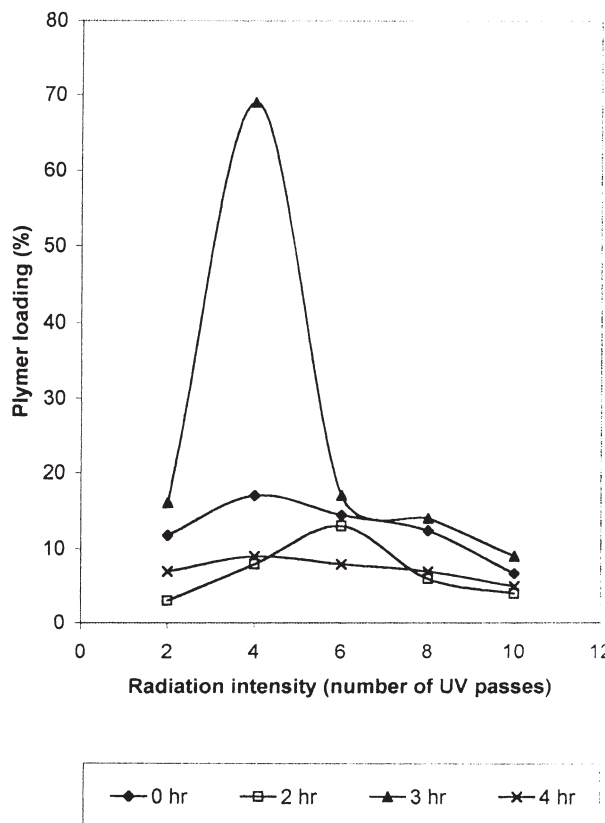

Figure 7 Effect of detergent pretreatment and monomer treatment on PL of coir fiber against UV radiation intensities with respect to washing time.

molecules dissolves in the oily layer of the fibers, which can be easily washed away by a stream of water. Coir fibers were pretreated with UV radiation for further surface modification. It was reported that UV irradiation improves the mechanical properties.[15,22]

Detergent-washed and UV-pretreated coir fibers were cured with the optimized solution of EGDMA under UV radiation. It is observed from the Table I that the highest T_f and E_f were recorded as 18.2 and 1.4 respectively for 50% EGDMA solution at 6 UV passes. In the case of increasing tensile properties with increasing radiation doses, intercrosslinking between the neighboring cellulose molecules that occurs under UV radiation and tensile properties decrease at higher UV doses. During photodegradation, there will be loss in strength due to primary bond breakage in the cellulose constituent, a reduced degree of polymerization and an increase of number of carboxyl and carbonyl group and therefore be related to photochemical changes taking place in the middle lamella, which reduce the ultimate cell.[20] The time lapse between irradiation and exposure is one of the important features that minimizes homopolymer formation.[19] So, detergent can remove pectin and related substance from the coir fiber and increases T_f.
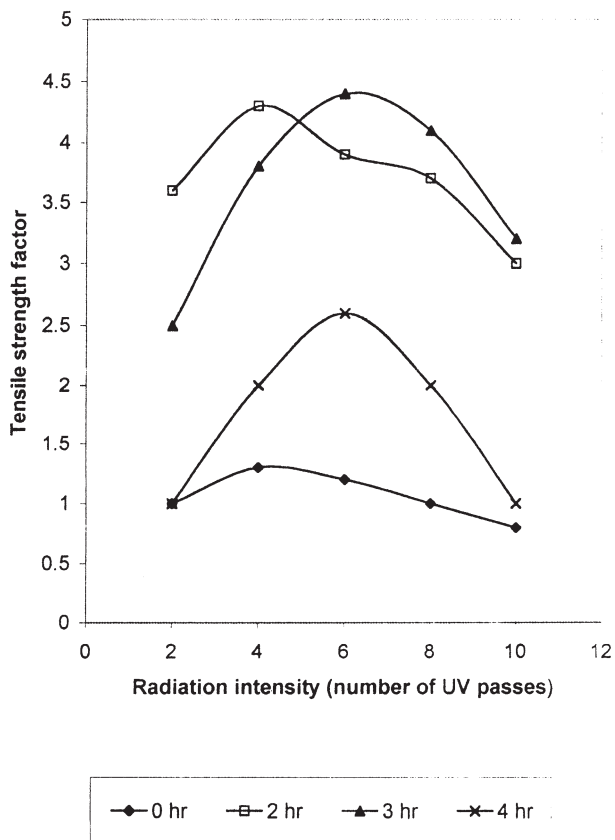
TABLE I
Effect of UV (10th pass) Radiation, Detergent (5%), and Monomer Treatment on Polymer Loading and Tensile Properties of Coir Fiber

Number of UV passes	Properties		
	Polymer loading (%)	Tensile strength factor (T_f)	Elongation at break factor (E_f)
2	2	8.7	1
4	5	10.8	1.1
6	7	18.2	1.4
8	3	7.4	1.2
10	2	5.7	1

TABLE II
Degradable Properties of Treated and Virgin Coir Fiber

Properties	Simulating weathering			Degradation in soil		
	V	V + E	V + D + E	V	V + E	V + D + E
Loss of weight (%)	40	30	25	50	38	30
Loss of TS (%)	55	41	30	80	50	35
Loss of E_b (%)	38	29	25	70	45	29

V, Virgin; V + E, Virgin + treated with 50% EGDMA; V + D + E, Virgin + detergent pretreated + treated with 50% EGDMA.

coir fibers due to weathering is shown in Table II. The loss of TS of the virgin sample is about 55%, while that of treated samples is from 30 to 41%, which shows the effect of the treatment. Investigation proved that reaction between the monomer and the cellulose changes the properties significantly.

Degradable properties

Degradation of samples in unsterile soil (15% water) for 105 days are shown in Table II. The virgin samples lost its 80% of TS in mud, whereas loss of TS of EGDMA- and detergent–EGDMA-treated fibers are 50 and 35% respectively. The rate of decomposition of the treated samples was lower compared to that of untreated samples. Investigation showed that reaction between the monomer and the cellulose changes the degradation properties in weather and mud significantly.

CONCLUSIONS

EGDMA treatment under UV radiation changes the properties of physical and degradable properties significantly. PL of treated fibers could be increased about 69% when pretreated with detergent. It was shown from investigation that the TS of coir fiber could be increased by 18.2 times of its original TS, when coir were pretreated with detergent and UV radiation and then treated with 50% EGDMA solution under UV radiation. Water uptake of the coir fiber is reduced considerably after the treatment. Losses of the physical properties after the treatment were reduced significantly.

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