

Influences of Various Surface Pretreatments on the Mechanical and Degradable Properties of Photografted Oil Palm Fibers

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ABSTRACT: Natural-fiber-reinforced polymers can exhibit very different mechanical performances and environmental aging resistance depending on their interphase properties. A lack of good interfacial adhesion and poor resistance to moisture absorption make oil palm fibers less attractive as reinforcing agents. To improve the interfacial properties, oil palm fibers were grafted with different percentages of allyl methacrylate (AMA) in methanol. Darocur 2959 was added as a photoinitiator and initiated photochemical polymerization. The fiber surfaces were pretreated by an alkali, KMnO_4 , and dewaxed; this was followed by grafting with AMA to determine the effect of various surface treatments on the physicomechanical properties of grafted fibers. The alkali treatment increased the surface roughness with

better impregnation of the polymer, which increased the tensile properties by about 20%. A minute amount of an additive (urea) and a coupling agent (silane) were added to the optimum formulations. These increased the physical properties of the grafted oil palm fibers because of increased crosslinking. The treated and untreated fiber samples were also subjected to various weather conditions, such as simulated weather, soil, and water aging, to determine the degradation properties, and lower losses were observed for the treated samples than the untreated fibers. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 105: 3077–3086, 2007

Key words: biofibers; coatings; graft copolymers; irradiation; photopolymerization

INTRODUCTION

In recent years, increasing interest has been focused on thermoplastic composites reinforced with lignocellulosic- and cellulosic-based materials.^{1–3} Lignocellulosics are favored as a new generation of reinforcing materials in thermoplastics because they are renewable natural resources. Moreover, increasing concern about the environment has promoted recyclable raw materials and products, emphasizing the demand for lignocellulosic–thermoplastic composites. Lignocellulosic fibers such as jute, oil palm, coir, and pineapple have been reportedly used as reinforcements in polymer matrices.⁴ The oil palm has a bunch of fibers in which the palm fruits are embedded, consisting of about 65% holocellulose and 25% lignin in the form of a fiber strand. The cell length of an oil palm empty fruit bunch is 1.3 cm

with a diameter of 19.1 μm ; it has a tensile strength (TS) of 130 MPa, a modulus of 3.58 GPa, and an elongation at break (Eb) of about 9.7%.⁵ The main cellulose constituent of oil palm fibers is a hydrophilic glucan polymer consisting of a linear chain of 1,4- β -bonded anhydroglucose units, and it also contains alcoholic hydroxyl groups.⁶ These hydroxyl groups form intermolecular hydrogen bonds inside the macromolecule itself and in other cellulose macromolecules. Therefore, oil palm fibers are hydrophilic in nature, particularly at elevated temperatures. Consequently, this certainly affects the mechanical performance of oil palm polymer composites, especially under long exposure to environmental conditions with elevated temperatures and atmospheric humidity. To overcome these problems, it is necessary to modify the fiber surface through the employment of physical and chemical methods to reduce the hydrophilic nature of the fiber and, therefore, the fiber–matrix bonding. In general, surface modifications of practical interest are readily achieved with suitable chemical treatments of the fibers. Various surface pretreatments, such as a permanganate treatment, a fiber peroxide treatment, and a resin peroxide treatment, of oil palm fibers were conducted by Agarwal et al.,⁷ and improved thermal properties were observed for oil palm fiber/polymer

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composites. Mishra et al.⁸ showed that the interface quality for pineapple leaf fiber/polyester composites could be enhanced through different surface modifications, such as dewaxing, alkali treatments, cyanoethylation, and the grafting of acrylonitrile monomer onto dewaxed fibers.⁹

Several processes, such as chemical treatments,^{10–13} photochemical treatments,^{14–19} and plasma treatments,²⁰ have been developed to modify polymers and fiber surfaces. Improved physicomaterial properties of natural fibers with different impregnating solutions under γ radiation^{14,15} and UV radiation^{16–19,21} have been reported, and similar processes can be applied to oil palm fibers to improve their physicomaterial properties.

It has been previously observed that both the polymer loading (PL) and TS of wood–plastic composites are enhanced with additives containing a carbamide group.^{22–24} Urea is a very cheap chemical and contains this group, so it has been selected as an additive for oil palm fibers. Coupling agents such as silane usually improve the degree of crosslinking in the interface region and offer a perfect bonding result. Silane coupling agents are effective in modifying the natural fiber/matrix interface. Various silanes are effective in improving the interface properties of wood–polypropylene composites,²⁵ mineral-filled elastomers,²⁶ fiber-reinforced epoxies, and phenolic composites.²⁷ Alkoxy silanes are able to form bonds with hydroxyl groups. The coupling agent silane has been tested in fiber treatments to improve the interface properties.

This work is an attempt to modify oil palm fibers with cheap chemical reagents and simple techniques for use as a good reinforcement in developing lightweight and low-cost composites. The aim of this work is to rationalize the effects of various surface pretreatments on the physicomaterial and degradable properties of polymer-grafted oil palm fibers.

EXPERIMENTAL

Materials

Oil palm fibers were collected from Malaysia. All chemicals, such as sodium hydroxide, ethanol, benzene, KMnO_4 , and acetone, were analytical-grade and were used without further purification. The monomer allyl methacrylate [AMA; i.e., $\text{H}_2\text{C}=\text{C}(\text{CH}_3)\text{CO}_2\text{CH}_2\text{CH}=\text{CH}_2$] was procured from Fluka Chimica (Buchs, Switzerland). The photoinitiator, Darocur 2959, was procured from Ciba–Geigy (Klybeckstrasse, Switzerland).

Experimental procedure and data analysis

Oil palm fibers were cut into small pieces (10–15 cm long) and washed in a 2% Surf (commercially

available washing powder, Unilever, United Kingdom) detergent solution at 70°C for 1 h. The fibers were washed with tap water and then distilled water and finally dried in an oven at 105°C.

Photografting of the fibers was performed through the soaking of the fibers for 20 min in monomer–methanol solutions of different concentrations and through irradiation under UV light (model IST, Technik, Germany) at 254–313-nm wavelengths of radiation and 2 kW of power with a 50-A current. The UV-radiation source contained a conveyor belt that moved around the mercury lamp, and after it moved once to the lamp, one pass was considered complete. The optimum curing conditions with respect to different physical properties were measured. The amount of the polymer loaded by the treated sample was measured with the following formula:

$$\text{PL (\%)} = [(W_t - W_0)/W_0] \times 100 \quad (1)$$

where W_t is the weight of the treated dry fiber and W_0 is the weight of the untreated virgin fiber. The UV-cured fibers were put in a Soxhlet extractor and refluxed in acetone for 48 h. The fibers were then washed with acetone several times and dried in an oven. The amount of the grafted polymer was determined with the following formulas:

$$\text{Grafting (\%)} = [(W_1 - W_2)/W_1] \times 100 \quad (2)$$

where W_1 and W_2 are the weights of the AMA-grafted oil palm fibers before and after extraction in acetone. Tensile properties such as TS and Eb of both grafted and untreated virgin fibers were measured with an Instron (Liverpool, UK) model 1011 TS machine run at a crosshead speed of 2 mm/min and a gauge length of 30 mm. The changes in TS and Eb were expressed by the TS factor or tenacity factor (T_f) and elongation factor (E_f):

$$T_f = \text{TS}_t/\text{TS}_0 \quad (3)$$

$$E_f = \text{Eb}_t/\text{Eb}_0 \quad (4)$$

where TS_t and TS_0 are the tensile strengths of the grafted and untreated virgin samples and Eb_t and Eb_0 are the elongation percentages of the treated and untreated empty fruit bunches of oil palm fibers. The water absorption properties of the treated and untreated fibers were determined by the placement of the samples in a static water bath at room temperature, and the water uptakes for different swelling times were measured with this formula:

$$W_g = [(W_a - W_0)/W_0] \times 100 \quad (5)$$

where W_g is the percentage of the water uptake and W_a and W_0 are the weights of the samples before and after water swelling.

Fiber surface pretreatment

The surface modifications consisted of mercerization, dewaxing, and a treatment with KMnO_4 . All the pretreatment processes were performed individually, and they did not follow one another. The first surface pretreatment process was mercerization with NaOH . Fibers were soaked in 5% NaOH (w/v based on the solution) for about half an hour to activate the OH groups of the cellulose and lignin in the fibers. The fibers were then washed many times in distilled water and finally dried to obtain alkali-treated fibers. The alkali-treated fibers were then grafted with an optimized AMA solution.

The fiber surfaces were dewaxed with benzene and ethanol in a 1 : 2 ratio for 72 h at 50°C , and this was followed by washing with tap water and distilled water. The fibers were then dried in an oven and treated with an optimized AMA solution.

The detergent-washed fibers were also subjected to a pretreatment with a 0.05–1% KMnO_4 solution. In this respect, the fibers were soaked in a KMnO_4 solution for about 5–10 min, and this was followed by thorough washing with tap water and rinsing with distilled water until the permanganate color was removed.

Addition of the additive and coupling agent

Urea and silane [3-(trimethoxysilyl) propylmethacrylate] were added to the optimized formulating solutions as the additive and coupling agent. They may have further increased the mechanical properties of the empty fruit bunches of oil palm fibers.

Degradable properties in soil and water environments

The fibers samples were weighed individually, some were buried in soil (having a 20% moisture content), and some were immersed in water for about 45 days. The weight losses and tensile properties due to aging were periodically monitored.

Simulated weathering test

The loss of the tensile properties by the impact of accelerated weathering testing was determined with an alternating cycle of sunshine (over 4 h at $65 \pm 2^\circ\text{C}$) and dew and condensation (for 2 h at $45 \pm 2^\circ\text{C}$) with the help of an accelerated weathering tester (model Q.U.V., Q-Panel Co., Westlake, OH) for a period of 100 h.

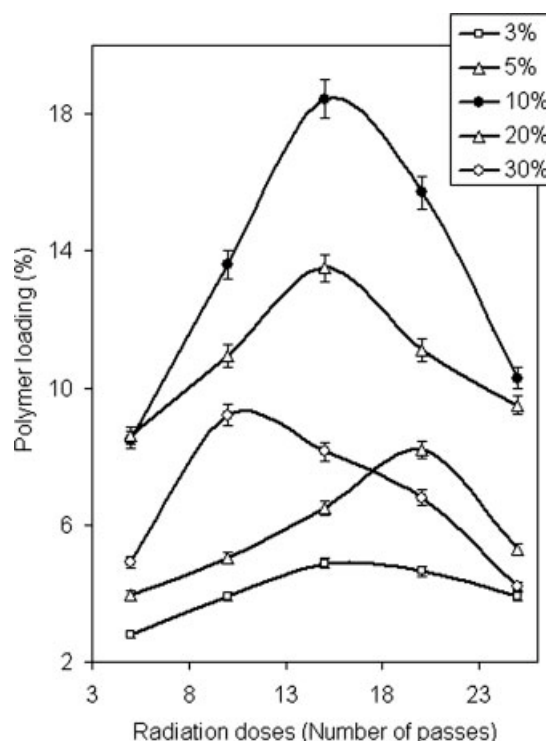


Figure 1 PL (%) of oil palm fibers at different radiation intensities as a function of the AMA concentration.

RESULTS AND DISCUSSION

The data presented in this report are average values of six replicates, and the associated error is $\pm 3\%$.

Treatment with AMA

The samples were soaked for 20 min in different concentrations of AMA (3–30%) in methanol before they were cured by UV radiation of different intensities. The cured samples were kept for 24 h under normal weathering conditions to complete the polymerization reaction. The PL and tensile properties were measured as functions of the monomer concentration and at different radiation doses. The optimum monomer concentration and radiation doses were determined from the study of the mechanical properties of the cured oil palm fibers.

PL

The PL values of the AMA-treated fibers at different radiation intensities are given in Figure 1 as a function of the monomer concentration. The values are relatively low at low monomer concentrations and increase with increasing monomer concentration up to 10%. The samples treated with more than 10% AMA showed decreased PL values. We also observed that PL increased with increasing UV intensities up

to a certain value, and after the attainment of the maximum values, it started to decrease with increasing radiation doses. The highest PL (18.44%) was observed for the fiber treated with 10% AMA and 15 passes of UV radiation. The vinyl monomer AMA promotes a rapid propagation reaction with the help of a photoinitiator, leading to a network polymer structure through a graft copolymerization reaction via the double bond. As the AMA concentration increases, the amount of residual unsaturation is also increased, with the result of a faster rate of formation of three-dimensional network structures causing restricted mobility. After a maximum value is achieved, the reduction of PL at a higher monomer concentration may be caused by two factors. At a higher AMA concentration, radical–radical recombination processes may dominate, creating more homopolymers rather than monomer, as well as a cellulose backbone reaction. Another reason could be that the swelling of the cellulose backbone is insufficient because of a lower amount of the solvent; methanol. As a result, the monomer molecules are incapable of penetrating the cellulose molecules in the presence of a lower solvent concentration, which may cause a smaller number of reacting sites at the cellulose backbone and thus continue to reduce the active sites. The crosslinking rate, especially during the early stages of radiation, is proportional to the monomer concentration. The monomer increases the radical–radical reaction termination and hence decreases the extent of the scission reaction and oxidation.^{16,17}

The amount of the grafted polymer was obtained from the extraction of the AMA-grafted fiber in acetone. In this process, the homopolymers were removed, and only the grafted molecules remained on the fiber. The results of grafting are graphically represented in Figure 2 versus the doses of UV radiation with respect to the monomer concentration. About 30% grafted onto the cellulose from the total amount of PL. The rest remained on the surface because of adhesive and van der Waals forces. The highest grafting value (5.65%) was obtained by the fiber samples treated with 10% AMA and irradiated with 15 passes of UV.

Tensile properties

Chemical processing directly influences the cellulosic fine structure of plant fibers. Consequently, chemical treatments (grafting with monomers) have a lasting effect on the mechanical behavior of oil palm fibers, especially on the fiber strength and stiffness. It is very important to know the strength of the fibers before they are combined into the thermoplastic matrix to understand better how the grafted fibers

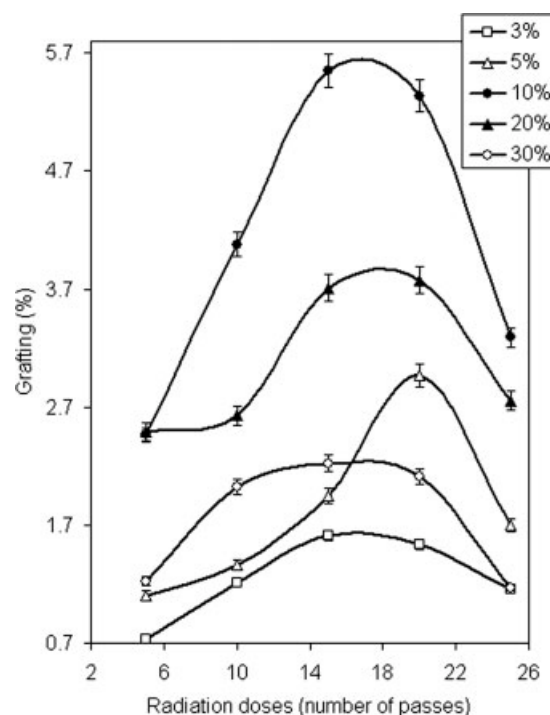


Figure 2 Grafting (%) of oil palm fibers after extraction in hot acetone at different UV doses as a function of the AMA concentration.

behave. The data show a higher strength for the treated fibers compared with the untreated fibers because of the removal of surface imperfections after the treatment. The increased uniformity of the fibers increased TS of the fibers through the presence of weak lateral fiber bonds.¹⁸ The variations in TS with the yield of the composites after different modifications were attributed to the changes in the chemical structure and bondability of the fibers. Many of the modifications decreased the strength properties because of the breakage of the bond structure and the disintegration of the noncellulosic materials.¹⁹ The reinforcing ability of the fibers depended not just on the mechanical strength of the fibers but also on many other features, such as the polarity of the fibers, surface characteristics, and the presence of reactive centers. These factors control interfacial interactions. The improved stiffness of the fibers was attributed to the crystalline region (cellulosic) of the fibers. The fibers also showed enhanced elongation properties. Mishra et al.²⁰ investigated the tensile properties of untreated, chemically modified, and AN-grafted sisal fibers. Chemically modified fibers showed an appreciable decrease in the tensile properties. This decrease was attributed to the substantial delignification and degradation of cellulosic chains during the chemical treatment. The extension at break of these fibers did not change much.

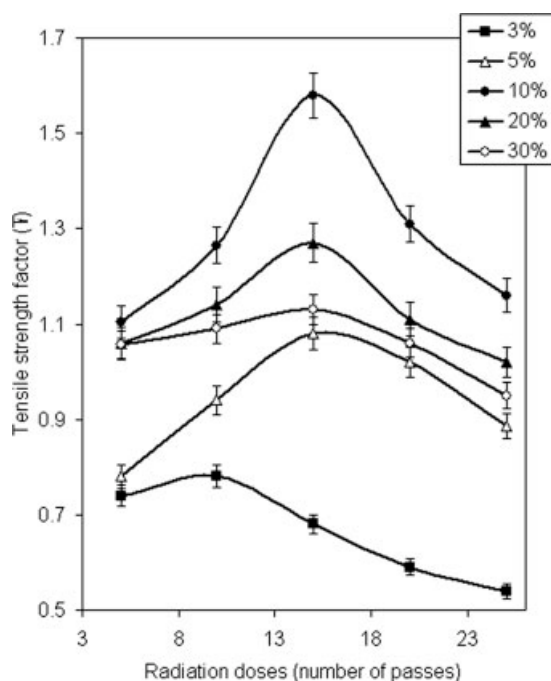


Figure 3 TS, represented by T_f , of empty oil palm fibers at different radiation intensities as a function of the AMA concentration.

The tensile properties represented by T_f and E_f at different UV intensities are graphically presented in Figures 3 and 4 as functions of the AMA concentration. T_f and E_f are the ratios of TS and Eb of grafted and nongrafted virgin fibers. We observed that both T_f and E_f increased with the UV-radiation doses, and after the attainment of the maximum values, the tensile properties decreased with the UV intensity. This decrease in T_f and E_f at a higher radiation dose could be caused by the degradation of the polymer at higher radiation doses.²¹ The highest TS ($T_f = 1.58$) was found for the sample treated with 10% AMA at the 15th pass of UV, and it was followed by the sample grafted with 20% monomer and the same radiation doses. The lowest TS ($T_f = 0.68$), which was lower than that of the virgin fiber, was found for the empty fruit bunches of oil palm fibers treated with 3% monomer and 15 passes of radiation. The enhancement of the elongation up to the breaking point of the empty fruit bunches of oil palm fiber samples was expressed as E_f and is depicted in Figure 4 versus the doses of UV radiation with respect to different AMA concentrations. As usual, the highest elongation ($E_f = 1.37$) was observed for the sample treated with 10% monomer after 15 passes of UV radiation. In this case, the highest value after 15 passes was lower than that of the virgin fiber ($E_f = 0.92$).

From the previous discussion, it can be assumed that the fiber samples treated with a 10% AMA

solution yielded higher PL and tensile properties after 15 passes of UV radiation. This curing condition was taken as the optimum grafting condition for empty fruit bunches of oil palm fibers.

Effect of the surface pretreatment

Lignocellulosic fibers offer attractive properties, but they are used only to a limited extent in industry.¹³ Natural fibers are strongly hydrophilic materials, and moisture absorption leads to a significant deterioration of their mechanical properties. Furthermore, most polymers are hydrophobic, and because of this divergent behavior, the interface in natural-fiber composites is rather poor. Any alteration of the characteristics of the cell wall, either chemical or morphological, has an effect on the mechanical properties of the fibers. Through the limitation of the substitution reaction at the fiber surface, good mechanical properties are reserved, and a degree of biodegradability is maintained.

Mercerization (alkali treatment)

An alkali treatment of cellulosic fibers, also called mercerization, is the usual method used to produce high-quality fibers. An alkali treatment improves the fiber-matrix adhesion because of the removal of natural and artificial impurities.²⁸ An alkali treatment increases surface roughness, resulting in better

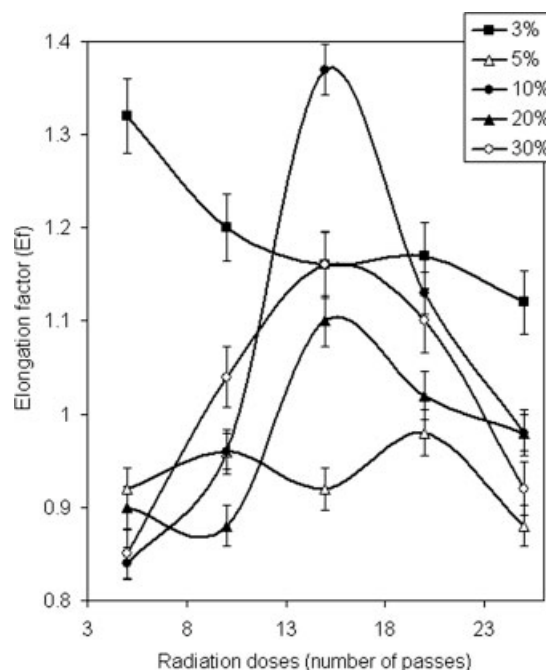
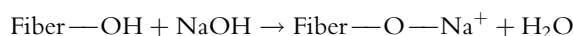


Figure 4 Eb, represented by E_f , of oil palm fibers at different radiation intensities as a function of the AMA concentration.

mechanical interlocking and more cellulose exposed on the fiber surface. This increases the number of possible reaction sites and allows better fiber wetting. The following reaction takes place as a result of an alkali treatment:



Consequently, mercerization or a more general alkali treatment has a lasting effect on the mechanical behavior of coir fibers, especially on the fiber strength and stiffness.⁵ Several other studies have been conducted on alkali treatments.^{18,29,30} They have reported that mercerization leads to an increase in the amount of amorphous cellulose at the expense of crystalline cellulose and the removal of hydrogen bonding in the network structure. The oil palm fiber treated with an alkali solution at 25°C was thoroughly washed and dried. After drying, the fiber was grafted with the optimized monomer-methanol solution and cured with different passes of UV radiation. The Fourier transform infrared (FTIR) spectra for untreated fibers showed an intense and broad band ranging from 3100 to 3800 cm⁻¹ due to the hydrogen-bonded —OH vibration of the cellulose structure of oil palm fibers. The peak at 2917 cm⁻¹ was assigned to the C—H stretching vibration from the —CH₂ group of cellulose and hemicellulose. The characteristic peak at 1736 cm⁻¹ was attributed to C=O stretching of carbonyl groups (>C=O) in hemicellulose. The peaks centered on 1436 and 1254 cm⁻¹ indicated the existence of lignin and hemicellulose structures, respectively.^{31,32} IR spectra of alkali (NaOH)-treated oil palm fibers showed that a broad peak at 3400 cm⁻¹ increased. This may have been due to the formation of more —OH groups as a result of the breaking of crosslinks of lignin and hemicellulose with cellulose after an alkali treatment.³³ The most remarkable change in the FTIR spectrum of NaOH-treated oil palm fibers was the absence of characteristic peaks at 1736 and 1254 cm⁻¹. This was accompanied by a reduction in the peak intensity at 1436 cm⁻¹. This was due to the complete decomposition of hemicellulose and partial leaching of lignin by NaOH. These results indicate that the ester type in hemicellulose is more easily removed by an alkali solution. Mwaikambo and Ansell³⁰ also reported that an alkali treatment is prone to attack the hemicellulose in sisal and jute rather than lignin. It appears that during an alkali treatment, substantial portions of uranic acid, a constituent of hemicellulose, and xylene are removed, and this results in the disappearance of this peak. The crystallinity increases as a result of the alkali treatment, and this results in a reduction in the sorption capacity of the fibers. The moisture content decreases as a result of the treatment by NaOH. Therefore, that part of the

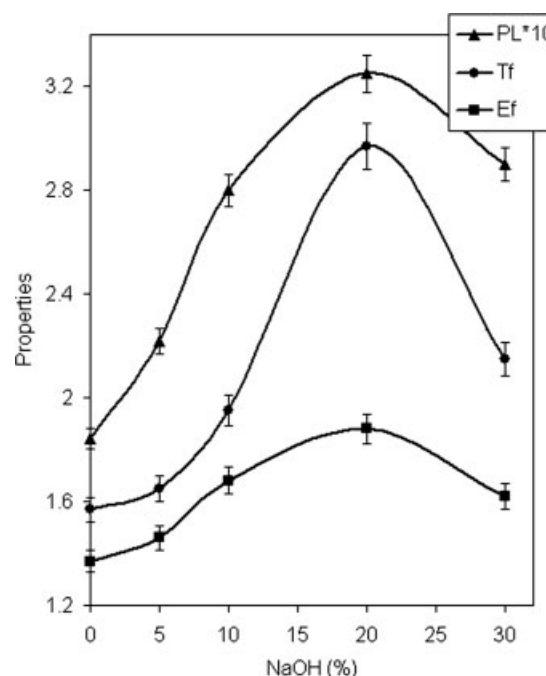


Figure 5 PL and tensile properties of alkali-treated, AMA-grafted fibers versus different alkali (NaOH) concentrations.

orientation polarization due to the presence of polar water molecules decreases. The alkali treatment leads to fiber fibrillation, that is, the breakdown of the composite fiber bundle into smaller fibers, thus increasing the effective surface area available for contact with the matrix.

The results for the PL and tensile properties are graphically presented in Figure 5. We observed that PL and TS increased to a great extent. The highest PL (32.5%) and tensile properties ($T_f = 2.97$, $E_f = 1.88$) were obtained for the sample pretreated with a 20% alkaline solution. The results of these parameters for different alkali concentrations are shown in Figure 5. We found that the alkali treatment increased PL by about 72% and TS by about 88% in comparison with those of samples with no alkali treatment.

Treatment with KMnO₄

A permanganate treatment was carried out to improve the bonding at the fiber-polymer interface. Permanganate solution concentrations of 0.05–1.0% were used. The fibers were then washed in distilled water and finally dried. The fibers became soft and their color changed upon the permanganate treatment. This treatment led to the formation of cellulose radicals through MnO³⁻-ion formation. The radicals enhanced the chemical interlocking at the interface.

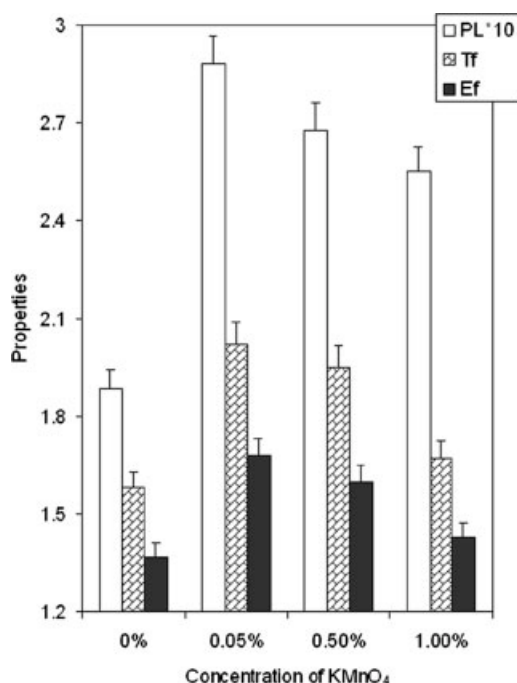


Figure 6 Effect of KMnO_4 on the mechanical properties of oil palm fibers as a function of the KMnO_4 concentration.

The results for the PL and tensile properties of fibers pretreated with KMnO_4 and then grafted with AMA are shown in Figure 6 for different KMnO_4 concentrations (0.05–1.0%). Among the different concentrations, 0.05% KMnO_4 produced the best results ($\text{PL} = 28.8\%$, $T_f = 2.02$). In the presence of KMnO_4 , the cellulose molecules of oil palm fibers undergo the following reactions, and the cellulose free radical is quite stable; it supports the higher rate of reaction with the monomer molecules and induces the grafting of AMA onto cellulose molecules of oil palm fibers. Comparing the results of alkali-treated and KMnO_4 -treated fibers, we have observed that KMnO_4 has less effect on fiber properties.

Dewaxing (treatment with benzene and ethanol)

The surface modification consisted of dewaxing; the detergent-washed fibers were soaked in a 1 : 2 mixture of ethanol and benzene for 72 h at 50°C , and this was followed by washing with distilled water. This treatment removed fats and waxes from the oil palm fiber surfaces. The dried fibers were grafted with AMA and cured under UV radiation. The PL and tensile properties were measured and compared with those of the monomer-grafted fibers. Benzene is a nonpolar solvent, and it attracts nonpolar parts, whereas ethanol is a polar solvent that attracts the polar substance of the oil palm fibers. As a result, impurities were removed from the fibers. As the boiling point of ethanol is lower than that of benzene, the polar substances were removed first.

The PL after dewaxing was 31.09%, with a T_f value of 2.25, and the elongation value was almost unchanged from those of the AMA-grafted fibers. Among the three surface pretreatment processes used in this work, the alkali treatment was found most useful. Dewaxing and KMnO_4 had less effect on the properties of the optimized monomer-grafted oil palm fibers.

Effect of the additive (urea) on the properties of oil palm fibers

Urea of different concentrations (1–5%) was added to an optimized AMA solution. Enhanced properties ($\text{PL} = 25\%$, $T_f = 1.82$) were achieved with 1% urea. The results for PL, T_f , and E_f are presented in Figure 7. Urea possesses $>\text{C}=\text{O}$ groups adjacent to a nitrogen atom having a lone pair of electrons, which are activated to form a bridge between the monomer and the cellulose through the additive. Urea is an inclusion compound whose properties could assist in partitioning by complexing with the monomer. This could lead to an increase in the monomer concentration at a grafting site and thus enhance the reactivity at that site, ultimately increasing the physico-mechanical properties of the grafted fibers.²⁴

Effect of the coupling agent (silane) on the properties of oil palm fibers

Silanes undergo hydrolysis, condensation, and bond formation. Silanols can form polysiloxane structures

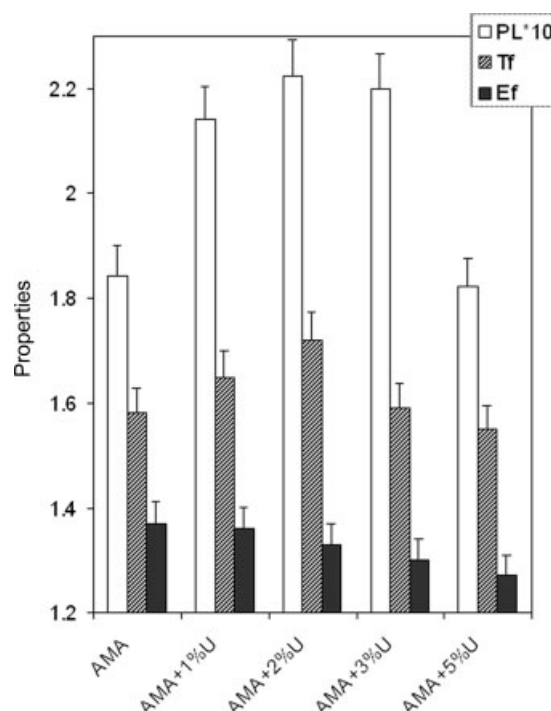


Figure 7 Effect of the addition of urea on the physico-mechanical properties of AMA-grafted oil palm fibers as a function of the urea concentration.

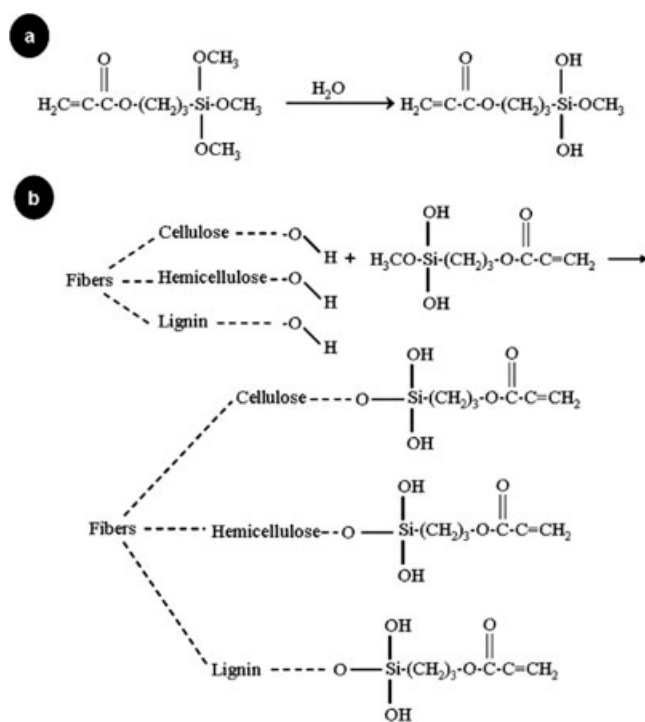


Figure 8 Reactions of oil palm fibers with silane: (a) hydrolysis of silane and (b) hypothetical reactions of fibers and silane.

by a reaction with hydroxyl groups of the fibers. The reaction schemes are given in Figure 8. The PL and tensile properties of oil palm fibers treated with different percentages (0.5–3%) of silane in formulating solutions are shown in Figure 9. The results are compared with those for 10% AMA treated fibers without any additives. Silane increased both the PL and tensile properties to a large extent in comparison with those of the AMA-treated fiber. We also found that 2% silane in the formulation yielded the highest PL and enhanced TS. This increase in the properties can be explained as follows: in the presence of moisture, hydrolyzable alkoxy groups lead to the formation of silanols. González et al.³³ investigated the effect of a silane coupling agent on the interface performance of henquen-fiber-reinforced high-density polyethylene composites. The fiber surface silanization resulted in better interfacial load transfer efficiency but did not improve the wetting of the fiber. Hydrogen- and covalent-bonding mechanisms could be found in the natural fiber/silane system. It was assumed that the hydrocarbon chains provided by the silane application influenced the wettability of the fibers, thus improving the chemical affinity to polyethylene. A silane treatment of cellulosic fibers can increase the interfacial strength and therefore the mechanical properties.^{34,35}

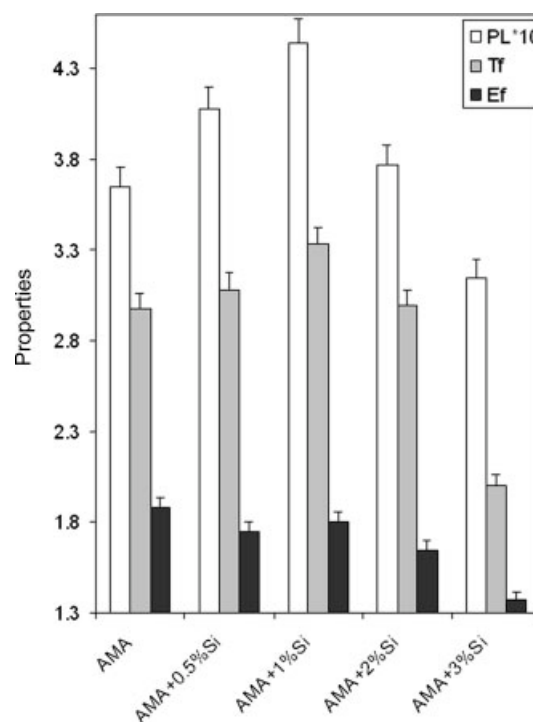


Figure 9 Effect of the addition of silane on the physico-mechanical properties of AMA-grafted oil palm fibers as a function of the urea concentration.

Water absorption properties

Major restrictions on the successful use of natural fibers in durable composite applications are their

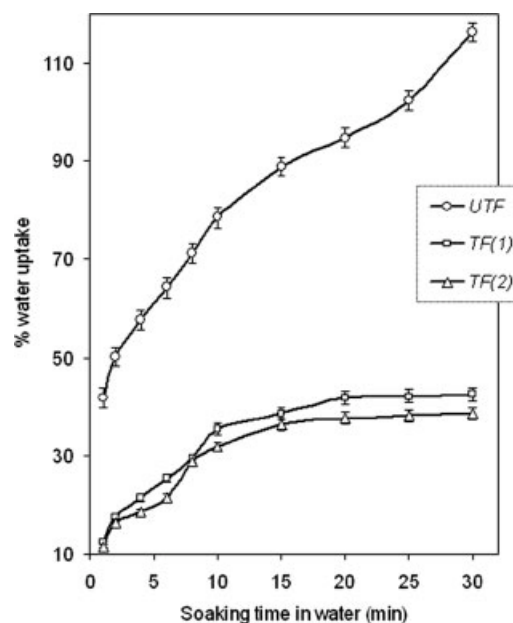


Figure 10 Water uptake (%) of treated and untreated empty fruit bunches of oil palm fibers versus the soaking time in water at 25°C [UTF = untreated fibers; TF(1) = 10% AMA grafted, alkali-pretreated fibers; TF(2) = 10% AMA grafted, KMnO₄-pretreated fibers].

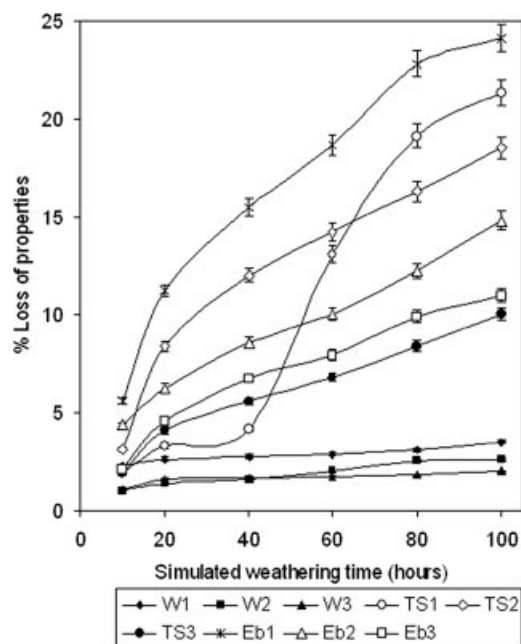


Figure 11 Loss of weight and tensile properties of empty fruit bunches of oil palm fibers due to weather simulation testing at different weathering times (W1 = weight loss of untreated fibers; W2 = weight loss of alkali-pretreated and 10% AMA/2% urea grafted fibers; W3 = weight loss of alkali-pretreated and 10% AMA/2% silane grafted fibers; T1 = loss of TS by untreated fibers; T2 = loss of TS by alkali-pretreated and 10% AMA/2% urea grafted fibers; T3 = loss of TS of fibers with 10% AMA/2% silane; Eb1 = loss of elongation by untreated fibers; T2 = loss of elongation by 10% AMA/2% urea treated fibers; T3 = loss of elongation of fibers with 10% AMA/2% silane).

high moisture absorption and poor dimensional stability (swelling), as well as their susceptibility to rotting.⁵ The swelling of fibers can lead to micro-cracking of the composite and, therefore, to deteriorated mechanical properties. The water uptake of treated and untreated oil palm samples was monitored at 25°C in a static water bath. The results are shown in Figure 10 versus the time of soaking in water. All the samples attained the maximum water uptake within the first 15 min, and then the soaking of water stopped for treated samples, whereas the untreated sample still continued to be soaked with water slowly. The minimum amount of water (12.59%) was taken up the urea-treated fiber, whereas the untreated sample showed the highest water uptake (135.21%). The minimum water uptake shown by the sample treated with 10% AMA and 2% urea indicates that urea plays some important role in repelling water molecules. The carboamide group ($>N-CO-$) of urea becomes chemically linked to the cellulosic chain of the empty fruit bunches of oil palm fibers, thereby forming a denser integrated network structure and hence more effectively restricting the penetration of water molecules.

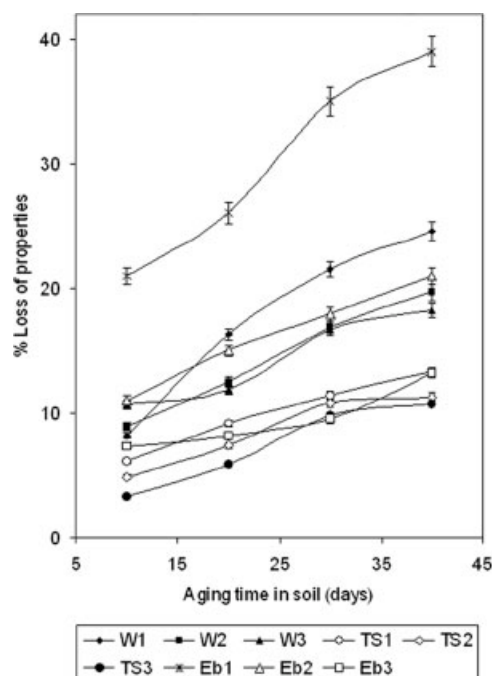


Figure 12 Loss of different properties of treated and untreated fibers in a soil environment (see Fig. 11 for a description of the terms).

The overall reduced water uptake of the AMA-treated sample in comparison with the virgin one is due to the fact that the monomer reacts with the OH group of cellulose through a graft copolymerization reaction, which reduces the hydrophilic nature of the fiber, and the polymer fills the void space of the

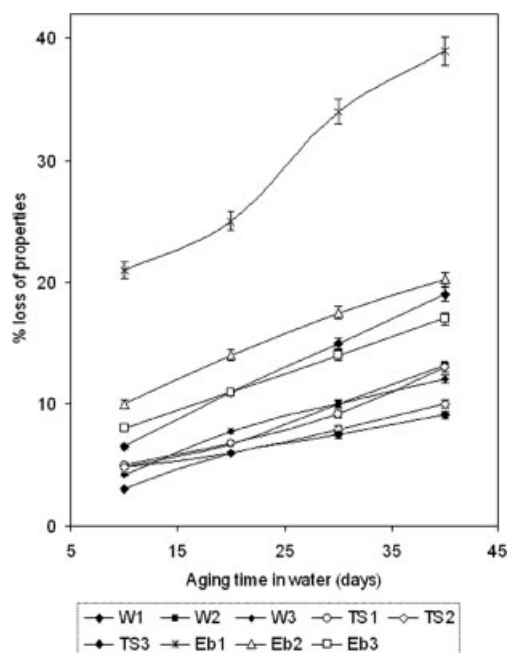


Figure 13 Loss of different properties of treated and untreated fibers in a water environment (see Fig. 11 for a description of the terms).

treated fiber. Through the replacement of some of the hydroxyl groups on the cell walls of polymers with bonded chemical groups, the hygroscopicity of the lignocellulosic material is reduced.

Degradability test

Simulated weathering test

The treated and untreated fibers were subjected to simulated weathering tests to monitor the effect on the mechanical properties of oil palm fibers. The losses of the properties of the virgin fibers are much higher than those of the grafted fibers. Virgin oil palm fibers, alkali-treated and then 10% AMA/2% urea grafted fibers, and alkali-treated and then 10% AMA/2% silane grafted fibers were subjected to simulated weathering tests, and the results are shown in Figure 11 versus the weathering time. The minimum loss of the fiber weight (2.01%) and tensile properties (TS = 10%, Eb = 11%) was observed for the alkali-treated sample grafted with the formulation containing the coupling agent silane.

Soil and water aging

The degradable properties of both treated and untreated samples were also monitored through aging in water and soil for about 2 months. The results of these losses are graphically plotted in Figures 12 and 13. The losses of the fiber weight and the tensile properties were higher in the soil environment than water. In both cases, minimum losses were obtained by the oil palm fibers containing the coupling agent silane.

CONCLUSIONS

An analysis has shown that a fiber treatment with AMA increases the physicomechanical properties and decreases the water absorption properties. The reason may be that the treatment of lignocellulosic fibers leads to the formation of a lignin-cellulose complex, which gives more stability to the fibers. A surface treatment with an alkali showed increased surface roughness and thus increased crosslinking in the polymer matrix, which ultimately increased the mechanical properties of the fibers. However, dewaxing and permanganate treatments showed little influence on the mechanical properties of the oil palm fibers.

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