Adv. Composite Mater., Vol. 16, No. 4, pp. 299–314 (2007)

Also available online - www.brill.nl/acm

Property improvement of natural fiber-reinforced green composites by water treatment

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This paper was written based on the *Proceedings of the 4th International Workshop on Green Composites (IWGC-4)* held on September 14–15, 2006 in Tokyo, Japan.

Received 30 January 2007; accepted 31 July 2007

Abstract—In the present study, natural fibers (jute, kenaf and henequen) reinforced thermoplastic (poly(lactic acid) and polypropylene) and thermosetting (unsaturated polyester) matrix composites were well fabricated by a compression molding technique using all chopped natural fibers of about 10 mm long, respectively. Prior to green composite fabrication, natural fiber bundles were surface-treated with tap water by static soaking and dynamic ultrasonication methods, respectively. The interfacial shear strength, flexural properties, and dynamic mechanical properties of each green composite system were investigated by means of single fiber microbonding test, 3-point flexural test, and dynamic mechanical analysis, respectively. The result indicated that the properties of the polymeric resins were significantly improved by incorporating the natural fibers into the resin matrix and also the properties of untreated green composites were further improved by the water treatment done to the natural fibers used. Also, the property improvement of natural fiber-reinforced green composites strongly depended on the treatment method. The interfacial and mechanical results agreed with each other.

Keywords: Green composites; natural fibers; water treatment; interfacial shear strength; flexural properties; dynamic mechanical properties.

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Edited by the JCSM

1. INTRODUCTION

Recently, eco-friendly green composites or natural fiber-reinforced plastics (NFRP), which may replace conventional glass fiber-reinforced plastics (GFRP), have been increasingly considered as promising materials due to their many advantages, like using a natural resource, environmental friendliness, sustainability, lightness, carbon dioxide reduction in nature, cost-effectiveness, etc. over the traditional GFRP [1, 2]. There however are some drawbacks, like poor adhesion between raw natural fibers and polymer resins, fiber variability, finite fiber length, irregular fiber shape and limited availability.

Among a variety of plant-based natural fibers, jute and kenaf, which are bast fibers, and henequen, which is a leaf fiber, were frequently used as reinforcement for NFRP [3–5]. Jute (*Corchorus capsularis L*) and kenaf (*Hibiscus cannabinus*) are important tropical crops and agro-fibers cultivating in Bangladesh, India and some other countries. Henequen (*Agave fourcroydes*) native to Yucatan, Mexico, which is a similar family to sisal, has been used extensively to make twines, ropes, carpets and cordages for a long period of time. A small number of papers have been reported on green composite materials using henequen [6, 7]. Such natural fibers are inexpensive, non-abrasive, and eco-friendly and, in general, they have excellent insulating characteristics against heat and noise due to their tubular or cellular structure. In addition, they have acceptable specific flexural and tensile properties as potential candidate for composite reinforcement. Considering the low specific gravity, the specific modulus of natural fibers is comparable with that of E-glass fibers. Also, one can avoid processing difficulties due to the fiber degradation during composite processing [8] if the processing temperature below about 230°C is used.

Polypropylene (PP) is one of the most widely used thermoplastic resins, especially in automobile part applications. It can also be extruded and injection-processed for molding composite parts with chopped natural fibers. Therefore PP has been most frequently utilized as matrix resin for developing NFRP or green composites. Poly(lactic acid) (PLA) is a promising and versatile polymer made from renewable agricultural sources like corn starch [9]. PLA is thermoplastic and completely biodegradable but stiff and brittle. Also, it has comparable mechanical properties with the enhanced thermal stability to polypropylene. PLA can be processed in a similar way to polypropylene. Unsaturated polyester (UP) resin is the most popular thermoset used in glass fiber composite industries. With this liquid-state resin, manual lay-up, spray lay-up, prepregging and autoclave methods can be utilized with good wettability even in a natural fiber-reinforced green composite system.

The interfacial adhesion between hydrophilic natural fibers and hydrophobic polymer surfaces has always been an important issue in most green composite systems. A strong interfacial bond is essential not only for improving mechanical and thermal properties but also for enhancing performances and lifetime of green composites. Hence, a large number of chemical and physical surface modification methods on a variety of natural fibers have been utilized primarily to increase the interfacial adhesion between natural fibers and a polymer matrix and as a result

to improve the composite properties [10–13]. However, chemical modification methods [10, 11], such as alkalization, acetylation and silane coupling, which have been extensively studied, are not environmentally benign and easily processible due to the organic substances used during the modification. Physical modification methods [12, 13], such as plasma treatment and electron beam irradiation, are relatively expensive and they are not easily facilitated industrially although they are dry and clean processes.

In this work, environmentally benign surface treatment methods using normal tap water, which may be industrially favorable, were utilized to improve the mechanical properties through an increase of the interfacial adhesion between natural fibers and a polymer matrix. Two different approaches for the surface treatment of natural fibers were performed. One is a static method by soaking and the other is a dynamic method by ultrasonication. The latter is to explore the effect of ultrasonic vibration exposed to the fiber surfaces during the treatment.

2. EXPERIMENTAL

2.1. Materials

Straight jute and kenaf fiber bundles of 80–90 cm long were purchased from Bangladesh Jute Institute, Bangladesh. Straight henequen fiber bundles of 70–80 cm long were supplied from Mexico. The natural fiber bundles were chopped to 10 mm long in average for fabricating randomly oriented natural fiber-reinforced green composites.

Poly(L-lactic acid) (PLA) pellets (Cargill-Dow Co., USA) with an average molecular weight of about 150 000 was used. PLA fibers having an average diameter of 200 μm were prepared by melt-spinning the PLA pellets using a capillary rheometer (Kayeness Inc., D 80528). The spinning rate was 0.5 mm/s at 200°C. The PLA fibers were chopped to 5 mm for jute/PLA green composites. PLA fibers, which were produced by Huvis Co., Korea using PLA from Cargill-Dow Co., were also used for fabricating kenaf/PLA green composites. The jute and kenaf fibers and PLA were fully dried at 70°C for 3 h in a vacuum oven prior to use. PP fibers (P-4203, Kolon Glotech Co., Korea) and UP resin (UP-GR235, ortho-type, Sewon Chem. Co., Korea) were used as thermoplastic and thermosetting matrix resins for henequen-reinforced green composites, respectively. During green composite processing, the PLA and PP fibers can be transformed to the matrix by melting, flowing, and then consolidating in the mold. The UP resin has the styrene content of 35 wt%. Methyl ethyl ketone peroxide (MEKP) of 1.0 wt% was used as catalyst.

2.2. Fiber surface treatment

'As-supplied' (raw) natural fiber bundles were treated with tap water by soaking and ultrasonication at ambient temperature for 2 h, respectively. The ultrasonic

frequency and power were 40 kHz and 400 Watt, respectively. Here, for both fiber surface treatments, hereinafter soaking is referred to as 'static treatment', whereas ultrasonication is referred as 'dynamic treatment' due to the ultrasonic frequency used.

2.3. Natural fiber/resin microdroplet formation

A single filament of thermoplastic PLA or PP was placed perpendicularly across with a single natural fiber (jute, kenaf or henequen) on a hot plate at about 210°C. The natural fiber was lifted up as soon as each PLA or PP filament was melted again. On this occasion, a tiny amount of the thermoplastic resin was covered over the single jute fiber surface and the resin about the fiber axis was rotated with re-melting by holding both ends in order to make the resin microdroplet uniform and then it was cooled down to ambient temperature to consolidate. Finally the PLA microdroplet was uniformly formed around the single jute or kenaf fiber embedded. On the other hand, a thermosetting (UP) resin microdroplet could be simply formed by touching a tiny amount of the liquid resin on a single henequen fiber and then fully curing in an oven according to scheduled curing profile.

The procedure for forming a resin microdroplet on a single natural fiber modified by both static and dynamic treatments was repeated to prepare a sufficient number of natural fiber/polymer green composite model specimens for single fiber microbonding tests. About 30 specimens of each natural fiber/polymer composite were prepared for the microbonding tests. The formed microdroplets were observed with an optical microscope (Nikon, Eclipse E200) before and after the test.

2.4. Green composite fabrication

Prior to composite fabrication, the natural fibers were completely dried in an oven. The fiber contents of jute/PLA, kenaf/PLA and henequen/PP green composites were 40 vol% and those in henequen/UP were 30 vol%. The chopped fiber length was about 10 mm on average. The chopped natural fibers and PLA or PP fibers were uniformly mixed and each molding compound was placed in a stainless steel matched-die mold. All green composites used were made by a compression molding technique. For PLA processing, the molding temperature and pressure were 195°C and 6.9 MPa, respectively. The holding time at 195°C was 10 min. In order to make henequen/PP green composites, the molding compound of chopped henequen fibers and PP fibers was placed in a stainless steel mold and then heated to 175°C for 30 min, holding at 6.9 MPa for 10 min using a hot-press. During processing, the PP fibers were melted enough to flow into the henequen fibers and then to encapsulate them. A pressure of 6.9 MPa was retained until the mold was naturally cooled down to ambient temperature.

For henequen/UP green composite processing, the uncured UP resin was uniformly poured into a mold with chopped henequen fibers and then cured primarily at 50°C for 10 min and further cured at 70°C for 1 h. The dimensions of the green

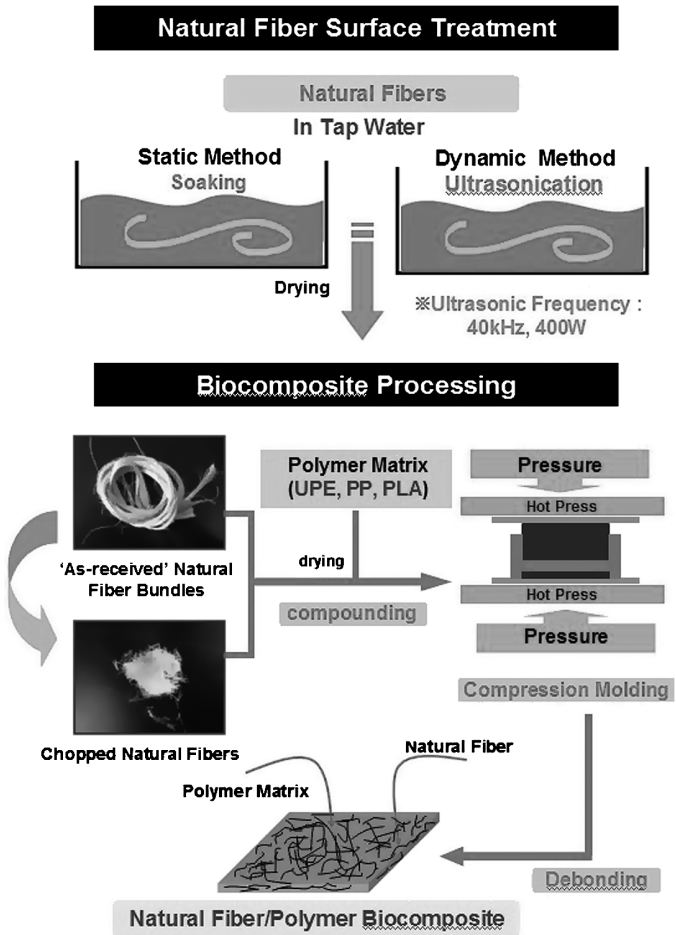

Figure 1. Schematic illustration of the experimental procedure of green composite fabrication.

composites with randomly oriented natural fibers were varied according to each analytical requirement. PLA, PP and UP plaques without natural fibers were also made for comparison. Figure 1 displays the experimental procedure of green composite fabrication used in the present work.

2.5. Analysis

Prior to resin microdroplet formation, natural fibers with a relatively uniform fiber diameter were selected for the single fiber microbonding test because cellulose-based natural fibers including henequen may, in general, have different fiber diameters in micrometer scale, depending on the fiber locations due to the irregular fiber surfaces. A universal testing machine (Instron 4467) was used for a single fiber microbonding test. The load cell was 100 N and the crosshead speed was 2 mm/min. The micro-vise grip distance was 20 mm. The testing of each sample was carried

out with about 30 specimens. The average value of the interfacial shear strength for each composite material was measured from all the test values obtained using the following equation.

$$\tau = F/(\pi \cdot D_f \cdot L_e).$$

Here, τ is the interfacial shear strength (IFSS). F is the force required for debonding the resin microdroplet from the single natural fiber filament while tensile loads are applied. D_f is the diameter of the measuring fiber; and L_e is the fiber length embedded in the resin microdroplet.

Three-point flexural tests were performed according to ASTM D790M-86 using a UTM. A load cell of 30 kN was used. The crosshead speed was 0.85 mm/min and the span-to-depth ratio was 16:1. The average values of the flexural strength and modulus of each composite were obtained from about 10 specimens.

Dynamic mechanical analysis (DMA Q800, TA Instruments) was done purging nitrogen gas with a heating rate of 5°C/min at a fixed frequency of 1 Hz and with the oscillation amplitude of 0.2 mm. A single cantilever bending mode was used. The specimen dimensions were $35 \times 12 \times 2$ mm^3.

3. RESULTS AND DISCUSSION

Figure 2 shows that the interfacial shear strength (IFSS) of untreated natural fiber/polymer green composites was significantly improved by both static and dynamic surface treatments with water, especially in jute/PLA composite (a). The water-treated jute/PLA green composite had an IFSS value much greater (>100% improvement) than the untreated counterpart. The water-treated henequen/UP and henequen/PP composite showed an IFSS value about 37% and 19% (static treatment) and about 58% and 48% (dynamic treatment) greater than the untreated case, respectively. Kenaf/PLA green composite (b) exhibited the lowest improvement (less than 10%) of the IFSS after the treatment. For the corresponding composite system, the improvement of the IFSS was greater with the dynamic treatment than with the static one.

The variation of the IFSS in the untreated and treated composites may be explained based on the topological change between the fibers untreated and treated, as reported earlier [5]. The impurities existing on the untreated fiber were removed by both the static and dynamic treatments. The earlier work demonstrated that the water treatment removed not only the surface impurities but also some waxy components present on the fiber surfaces. As a result, the treated fibers had rougher surfaces with more crevices than the untreated. It was also found that the natural fiber surfaces treated dynamically were rougher, more undulated and had more cellulose fibrils, compared to the statically treated one. This can explain the fact that the IFSS of the composite with the natural fibers treated dynamically by ultrasonication was greater than that treated statically by soaking. A possible reason for the surfaces modified by the dynamic treatment with tap water is that chlorine

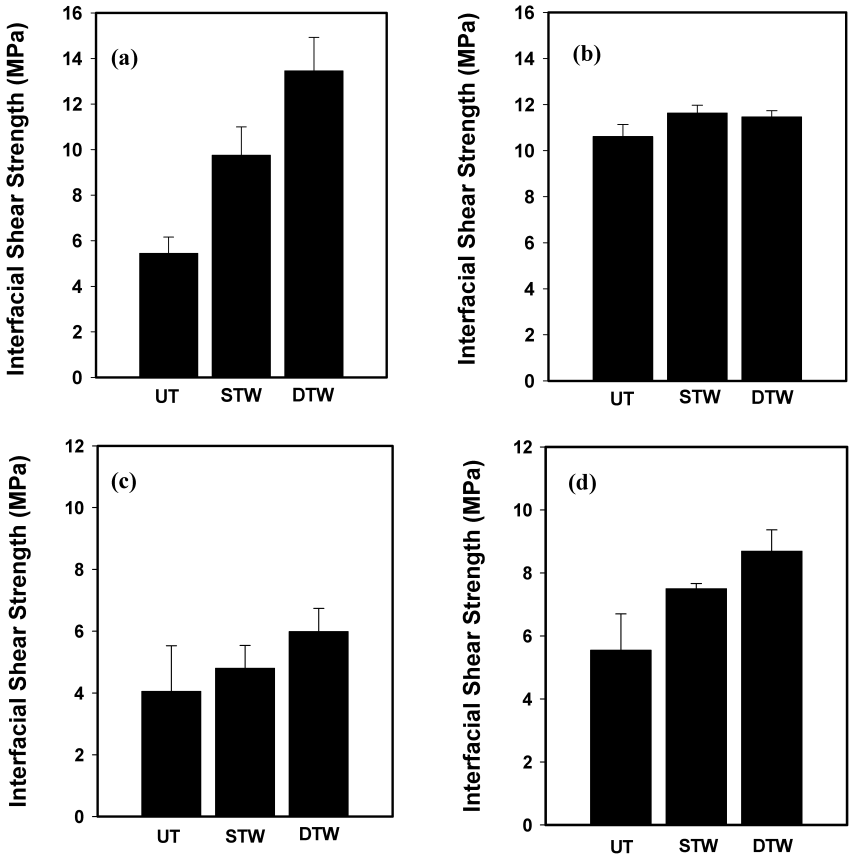

Figure 2. A comparison of the interfacial shear strengths of four green composites with natural fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water. (a) Jute/PLA, (b) Kenaf/PLA, (c) Henequen/PP, (d) Henequen/UP.

and metal ions present in tap water may somewhat contribute to the topological change of the natural fibers exposed to ultrasonic vibration with a high frequency. The greatest improvement of the IFSS value in the jute composite may be because the raw jute fibers used here have the lowest quality and cost among the three natural fibers utilized in this work. It has been expected that 'as-supplied' jute fiber bundles have the greatest surface impurities and foreign artifacts, which are not desirable for the interfacial adhesion between the fibers and the resin as long as they remain on the surfaces without removal.

In many applications, the flexural test is more relevant with advantages for material design or specification purposes than a tensile test [14]. In Figs 3 and 4, PLA itself has a low flexural strength. Therefore its flexural strength was largely influenced by incorporating jute and kenaf fibers and further increased by the surface treatment. Figure 3 depicts the flexural strength and modulus measured for jute/PLA green composites reinforced with the jute fibers untreated and treated

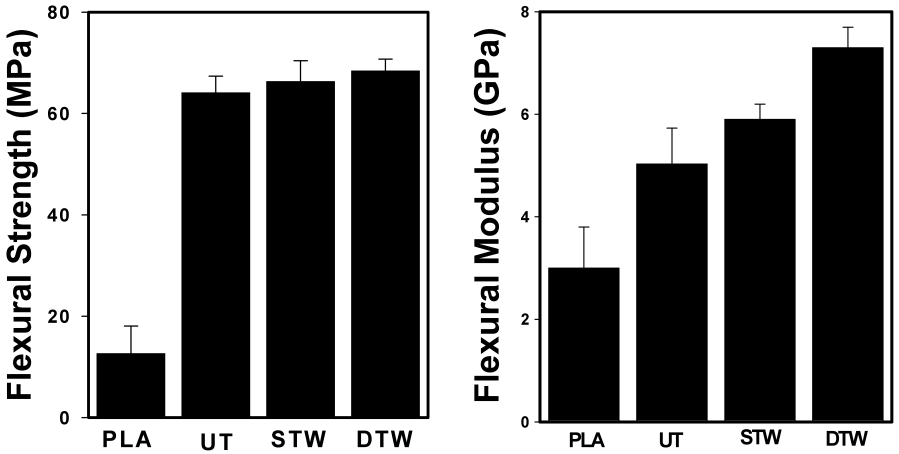

Figure 3. Flexural strength and modulus of PLA control and jute/PLA green composites using jute fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

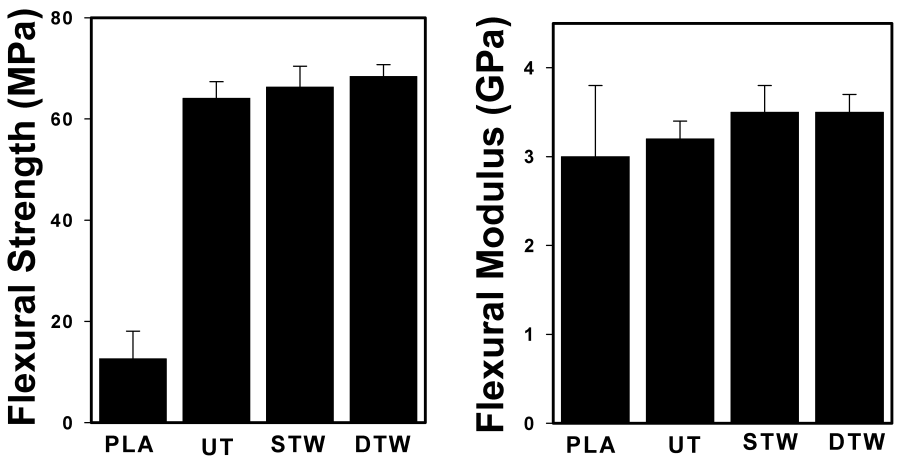

Figure 4. Flexural strength and modulus of PLA control and kenaf/PLA green composites using kenaf fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

dynamically using tap water. The result is also compared with PLA control. The result demonstrated that the flexural strength and modulus of PLA were dramatically improved by incorporating chopped jute fibers. The flexural strength of PLA was markedly improved about 6 times in the untreated jute/PLA green composite by the static treatment with water. The flexural strength of the untreated green composite was greatly increased by about 15% with the statically treated fibers and further increased by using dynamically treated fibers. This implies that the ultrasonic vibration with a high frequency exposed to the fibers during the treatment may influence more greatly the fiber topography.

There was a significant improvement (about 73%) in the flexural modulus of PLA by incorporating natural fibers into the PLA resin. The greatest improvement (about

150%) of the PLA modulus was found in the green composite reinforced with the jute fibers statically treated in water. Compared with the untreated composite, the modulus was increased by 48% in the statically treated jute/PLA composite and by 16% in the dynamically treated specimen.

Figure 4 demonstrates that the flexural properties of PLA were also greatly improved by incorporating chopped kenaf fibers into the PLA resin and further improved by the surface treatments given. However, the relative values of the flexural strength and modulus of kenaf/PLA green composites were measured to be lower than those of jute/PLA composites. This may be due to the difference of the PLA fiber types applied to the two composites. That is, as mentioned earlier, the straight PLA fibers used in the jute composites were from melt spinning through a capillary rheometer, whereas the PLA fibers used in the kenaf composites were in the woolly entangled fiber form. Accordingly, the former could have better fiber alignment and more uniform distribution of the chopped PLA fibers with the chopped jute fibers than the latter although the same natural fiber/PLA ratios were used and both PLA fibers were completely transformed into the PLA matrix by melting, flowing and subsequently consolidating during the composite processing. However, the effect of water treatment on the flexural properties was not as great as found in the jute composites, as similarly found in the interfacial shear strength result. This is because the kenaf fibers used here have better surface quality than the jute fibers. In other words, the jute fiber surfaces are readily subject to be changed with the static or dynamic treatment.

The increase of the mechanical properties of the green composite by incorporating the treated jute fibers may be caused by some topological changes in the treated natural fibers. The jute fiber surfaces became rougher with more undulation and surface area after the treatment, as found earlier [5]. Such the topological change may lead to mechanical interlocking between the jute fibers and the PLA resin. It may contribute to increasing the rigidity and integrity of the composite and as a result the flexural properties were increased.

Consequently it is concluded that the interfacial and flexural properties of PLA can be significantly improved by introducing bast plant-based natural fibers to the PLA matrix and also further increased by the water treatment. It was also noted that the increase of the interfacial strength in the natural fiber/PLA green composites by the water treatment played a significant role in improving their flexural properties.

In henequen/PP and henequen/UP composites of Figs 5 and 6, the flexural properties were not significantly increased by incorporating chopped henequen fibers into PP and UP resins. Rather, the flexural strengths of PP and UP were decreased, especially in the thermosetting UP resin. This may be because the presence of hollow-type henequen fibers with multiple cells in the PP matrix decreased the strength. The cells played a role as pores or microstructural defects therein, which are directly related to the strength. It has been found that henequen has a greater number of micrometer-sized cells than jute or kenaf, as observed earlier [13]. As a matter of fact, henequen fibers have different shape, fiber morphology

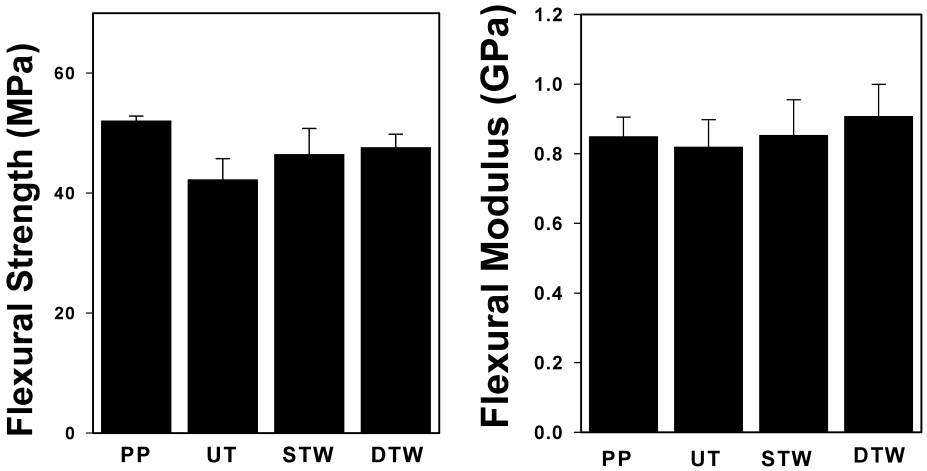

Figure 5. Flexural strength and modulus of PP control and henequen/PP green composites using henequen fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

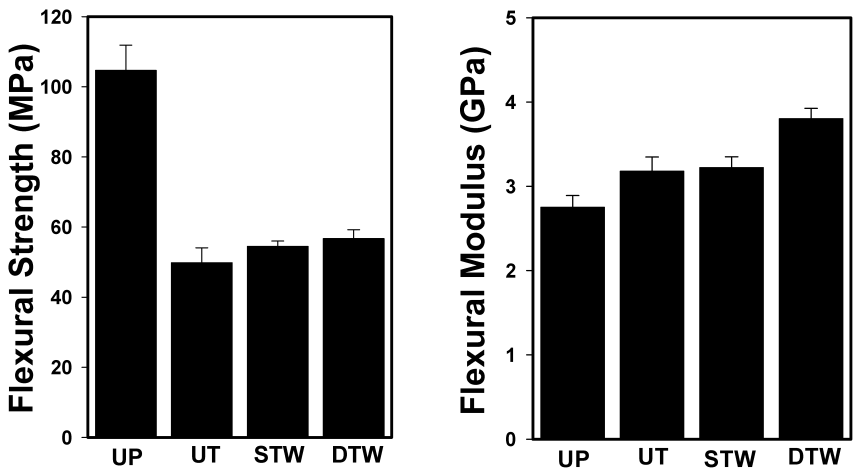

Figure 6. Flexural strength and modulus of UP control and henequen/UP green composites using henequen fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

and cell structure from jute and kenaf fibers. Each cell of henequen is much less dense than that of jute and kenaf fibers. Also henequen has a greater fiber diameter and a larger number of cells in a single fiber.

The flexural strength of henequen/PP composites was more or less recovered by both static and dynamic treatments. The flexural strength of untreated henequen/PP green composite was improved with both static and dynamic treatments. The flexural strength of the untreated henequen/PP and henequen/UP composite was improved by about 10% and 5% with the static treatment, respectively. And it was further increased slightly by 3% and 2% with the dynamic treatment, respectively.

With this treatment, the decreased flexural strength of PP was completely recovered, showing the strengths 9% (static) and 12% (dynamic) greater than the PP control.

Figures 5 and 6 also depict the effect of the static and dynamic treatments applied to henequen fiber surfaces on the flexural modulus of untreated henequen/PP and henequen/UP composite. The flexural modulus of surface-treated henequen composites was greater than that of the matrix resin and the untreated counterparts. The modulus was increased slightly by the static treatment and further by the dynamic treatment. This is mainly because the incorporated henequen fibers with a high aspect ratio more influenced the modulus than the strength. The dynamically treated henequen fibers contributed a greater flexural modulus to the composite than the untreated and the statically treated fibers. This indicates that the ultrasonic vibration with a high frequency exposed to the henequen fibers during the treatment may contribute to the fiber surface change, as found in jute and kenaf composites.

As a result, it may be said that both kenaf and henequen fibers are not as sensitive to the water treatment done, compared with the jute fibers. One plausible reason for that is that 'as-supplied' henequen and kenaf fiber bundles were of better fiber quality than 'as-supplied' jute fiber bundles, so that the fiber surfaces may not be subject to change by the treatment given as greatly as in the jute fibers of poor fiber quality and cleanness.

Figure 7 shows the variations of the storage modulus (top) and $\tan\delta$ (bottom) of PLA control and jute/PLA green composites untreated and treated statically and dynamically with water, respectively. Figure 8 compares the storage moduli found at three different temperatures, 40°C, 60°C and 85°C. Each temperature was selected to examine the modulus change at the temperatures far below the glass transition temperature (T_g), below the T_g and near the T_g, respectively. As can be seen, the storage modulus of PLA was greatly improved by reinforcing with the chopped jute fibers, indicating the higher dynamic stiffness. At 40°C, the storage modulus of PLA was 2.4 GPa, whereas the modulus of jute/PLA composites was in the range of 4.8 GPa–5.6 GPa, depending on the surface treatment utilized. The modulus of the untreated composite was significantly increased by the water treatment and the greatest improvement (about 133%) was achieved by the dynamic treatment. Although the storage modulus of the PLA and the green composites was decreased largely in the glass transition region, the improvement exhibited a similar tendency as shown at 40°C and 60°C.

The intensity of the $\tan\delta$ peak, which is related to the damping property of a material, for PLA was greatly decreased by reinforcing with jute fibers. It was also found that there were two distinguishable $\tan\delta$ peaks in the specimens measured. The first peak was related with the T_g of the material. The T_g of PLA was measured to be 88°C. This value is relatively higher than conventional PLA. Here, the fiber form of PLA probably exhibits somewhat higher T_g. In addition, the T_g determined using the DMA technique is normally higher than that determined using DSC or other thermal methods, depending on the polymer itself and measurement

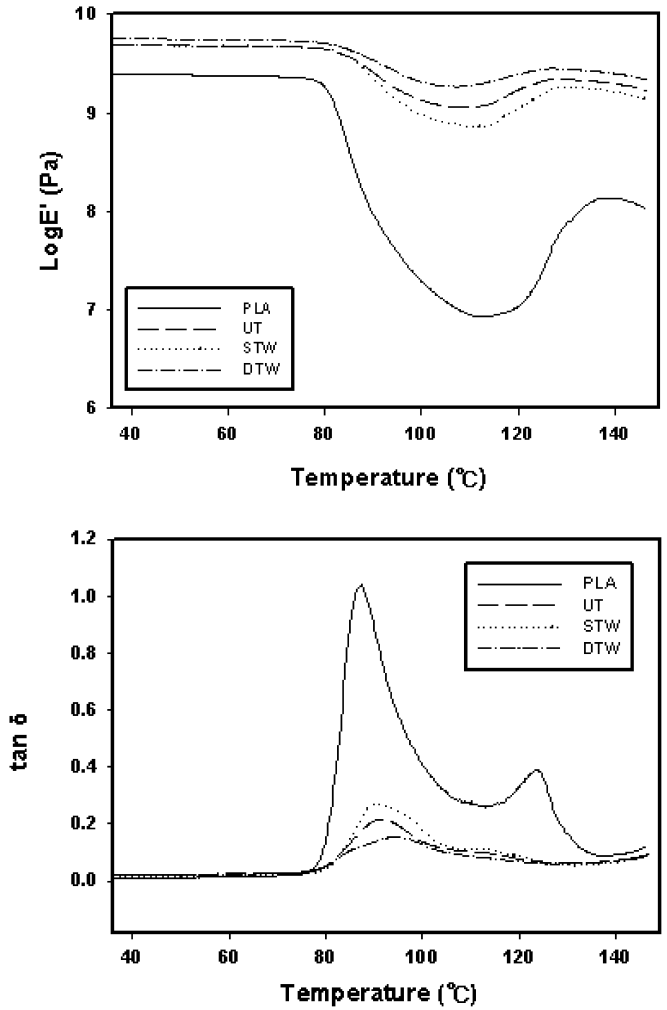

Figure 7. Storage modulus and tan δ of PLA control and jute/PLA green composites using jute fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

conditions. DMA is more sensitive to molecular motion of the polymer tested than other thermal methods like DSC or TMA, resulting in a higher value of T_g [15].

The jute/PLA green composites exhibited the T_g in the range of 91°C–95°C depending on the surface treatment. The T_g of PLA was increased about 3°C–4°C by the fiber reinforcing effect. The T_g of untreated jute/PLA specimen was slightly increased about 2°C–4°C with the water treatment. The second peak observed near 120°C is related with the crystallization behavior of PLA. PLA is a semi-crystalline material. The crystallization temperature of PLA was observed to be around 120°C, as found from a differential scanning calorimetric study earlier [5]. The storage modulus of the PLA and the jute/PLA composites was decreased from about 80°C and then it was increased again from about 110°C, resulting in a small second

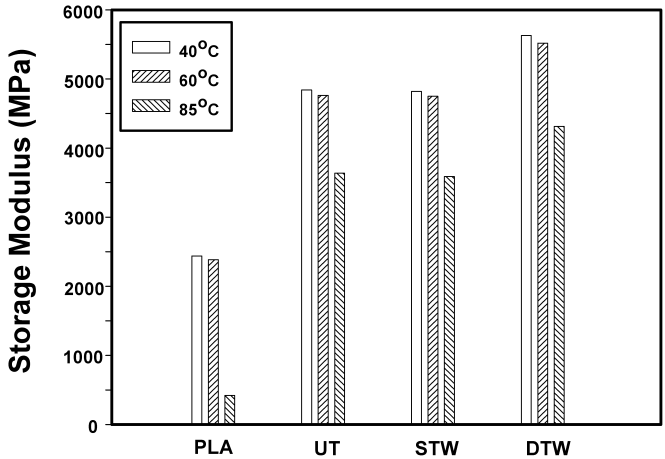

Figure 8. A comparison of the storage moduli among (A) PLA, (B) untreated, (C) statically treated, and (D) dynamically treated jute/PLA green composites examined at 40°C, 60°C and 85°C.

peak around 120°C. This was due to a typical effect of cold crystallization of PLA matrix [9]. The second peak of PLA was largely reduced by the incorporation of jute fibers into the PLA matrix. Therefore it may be thought that the crystallization behavior of PLA can be significantly restricted by the reinforcing natural fibers.

Figure 9 displays the variations of the storage modulus (top) and tan δ (bottom) of PP control and henequen/PP composites untreated and treated statically and dynamically with water, respectively. Figure 10 compares the storage moduli found at four different temperatures: -30°C (far below the T_g), -15°C (below the T_g), 40°C (above the T_g) and 80°C (far above the T_g). The storage modulus of PP was improved by incorporating with the chopped henequen fibers, reflecting the reinforcing effect. The modulus of the untreated composite was slightly increased by the treatment, indicating that there was no significant effect on the value. The storage modulus of the PP and the composites was also decreased largely above the glass transition region, exhibiting a similar tendency of the variation of the modulus at other measuring temperatures. The intensity of the tan δ peak of PP was decreased by the incorporated henequen fibers. The T_g of PP was measured to be about 0°C and the T_g of henequen/PP composites was more or less lower than that of PP. The tan δ characteristics were not distinguishable with the water treatment.

4. CONCLUSIONS

The interfacial shear strength of untreated kenaf/jute, henequen/PP and henequen/UP green composites was significantly improved by water treatment, particularly with a dynamic ultrasonication method. The flexural strength of PLA was greatly improved by introducing chopped jute and kenaf fibers to the PLA matrix, whereas the strength of polypropylene and unsaturated polyester were rather decreased with

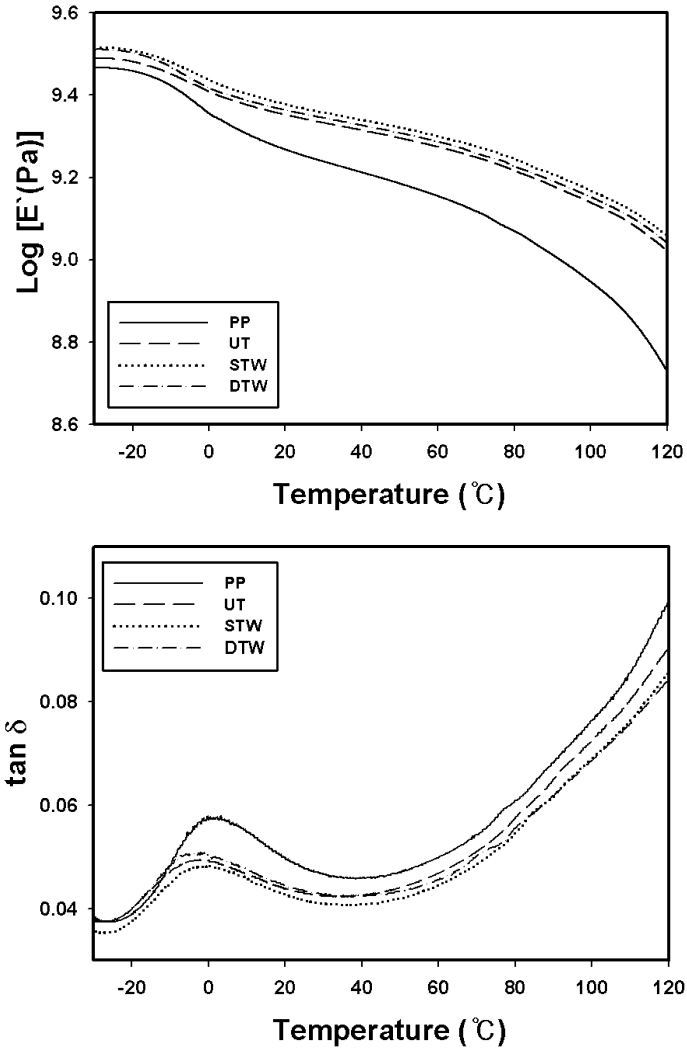

Figure 9. Storage modulus and tan δ of PP control and henequen/PP composites using henequen fibers untreated (UT) and treated statically (STW) and dynamically (DTW) with water.

the addition of chopped henequen fibers. The flexural properties of all the untreated green composites were somewhat increased by the water treatment done on to the natural fibers used, particularly with a dynamic method. The dynamic storage modulus of PLA and PP resins was increased with the incorporation of chopped jute and henequen fibers, respectively. The storage modulus of the untreated jute/PLA and henequen/PP composites was more or less increased by the water treatment done, as similarly found in the interfacial and flexural results.

The interfacial shear strength, flexural properties and dynamic mechanical properties of untreated green composites were improved by both static soaking and dynamic ultrasonication treatments applied to jute, kenaf and henequen with tap water,

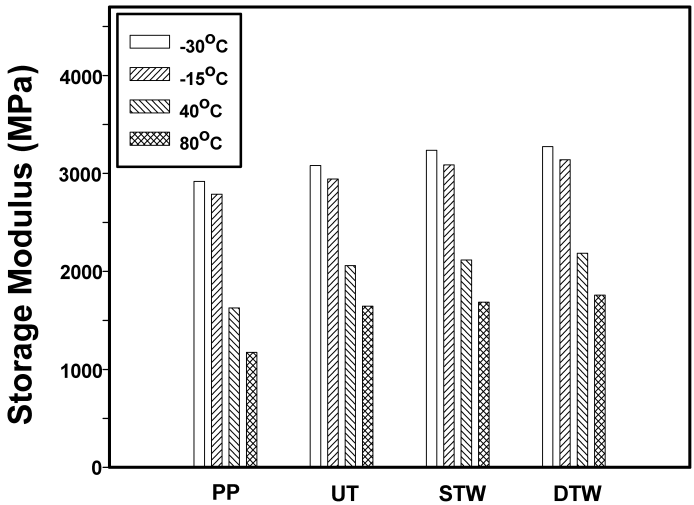

Figure 10. A comparison of the storage moduli among (A) PP, (B) untreated, (C) statically treated, and (D) dynamically treated henequen/PP composites examined at −30°C, −15°C, 40°C and 80°C.

depending on the type of natural fibers. The dynamic method was more effective to the property improvement of the green composites than the static method. It may be said that obtaining the increased interfacial shear strength between the natural fibers and the resin in an untreated green composite contributes to improving not only the flexural properties but also the dynamic mechanical thermal properties. It is also noted that the effect of the surface treatment method on the interfacial and mechanical properties of a green composite system depends on the type of natural fibers used. The present study suggests that the water treatment of natural fibers may be practically favorable with many advantages for improving the properties of green composites.

Acknowledgements

This research was supported by the Program for the Training of Graduate Students in Regional Innovation which was conducted by the Ministry of Commerce Industry and Energy of the Korean Government.

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