Effect of Natural Fiber Surface Treatments on the Interfacial and Mechanical Properties of Henequen/Polypropylene Biocomposites

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Abstract: The surfaces of henequen fibers, which can be obtained from the leaves of agave plants, were treated with two different media, tap water and sodium hydroxide, that underwent both soaking and ultrasonic methods for the fiber surface treatment. Various biocomposites were fabricated with untreated and treated, chopped henequen fibers and polypropylene using a compression molding method. The result is discussed in terms of interfacial shear strength, flexural properties, dynamic mechanical properties, and fracture surface observations of the biocomposites. The soaking (static method) and ultrasonic (dynamic method) treatments with tap water and sodium hydroxide at different concentrations and treatment times significantly influenced the interfacial, flexural and dynamic mechanical properties of henequen/polypropylene biocomposites. The alkali treatment was more effective than the water treatment in improving the interfacial and mechanical properties of randomly oriented, chopped henequen/PP biocomposites. In addition, the application of the ultrasonic method to each treatment was relatively more effective in increasing the properties than the soaking method, depending on the treatment medium and condition. The greatest improvement in the properties studied was achieved by ultrasonic alkalization of natural fibers, which was in agreement with the other results of interfacial shear strength, flexural strength and modulus, storage modulus, and fracture surfaces.

Keywords: biocomposites, polypropylene, henequen natural fibers, surface treatment, interfacial properties, mechanical properties.

Introduction

Biocomposites have increasingly attracted attention because they may be a promising material not only as a novel material for natural resource, eco-friendliness, sustainability, lightness, carbon dioxide reduction in nature and cost-effectiveness, but also as an alternative to conventional glass fiber polymer composites in many industrial and commodity applications.[1,2] Although plant bast-based natural fibers like jute, kenaf, flax and hemp have been widely utilized as reinforcement,[3-5] leaf-based natural fibers like sisal, henequen, and banana have also been increasingly used.[6,7] Of them, henequen fibers are attracting more attention due to relatively low cost and density for biocomposites.[8]

Henequen (*Agave fourcroydes*), which is a similar family with sisal, is a long, hard and strong fiber obtained from the long leaves of agave plants, which are native to Yucatan, Mexico. These natural fibers have been used traditionally to make twines, ropes, carpets and cordages for a long period of time.[9] However, a small number of papers have been reported on biocomposite materials using henequen,[10-12] compared to other natural fiber systems. Polypropylene (PP) is the most widely used thermoplastic resin among a number of polymer materials,[13,14] especially in automobile part applications. This material can be extended to extrusion and injection techniques for molding composite parts with short natural fibers.[15] Therefore, PP has been most frequently utilized as matrix resin in research and development of natural fiber composites or biocomposites for the last decade.

The interfacial adhesion between hydrophilic natural fibers and hydrophobic polymer matrices has often been an important issue in many different biocomposite systems.[16] A strong bond in the interfacial region is essential for

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achieving high mechanical performances of biocomposites. Therefore, a number of chemical and physical surface modification studies on a variety of natural fibers have been devoted to understand and enhance the interfacial adhesion between the natural fibers and the polymer matrix and to further improve the composite properties. For instance, representative chemical modification methods[17-20] are alkalization, acetylation, silane coupling and grafting, and key physical modification methods[12,21,22] are plasma treatment, electron beam irradiation and surface roughening. The chemical methods have been more frequently used because they are relatively simple, inexpensive and effective. However, generally they have been performed in static treatment conditions.

In the present study, water and sodium hydroxide (NaOH) were independently used as fiber surface treatment media. Ultrasonic technique was, for the first time, utilized as a dynamic treatment tool to further enhance the treatment efficiency, compared with a static treatment by soaking. The ultimate aim of the research is to increase the interfacial adhesion between the natural fibers and the polymer matrix in a biocomposite system and then to enhance the flexural and dynamic mechanical properties. The objectives of the present study are to investigate the interfacial shear strength, flexural strength, flexural modulus, dynamic storage modulus, tan δ, and fracture surface observations of henequen/PP biocomposites fabricated using chopped henequen fibers surface-treated with different media and methods and to elucidate how the dynamic treatment influences the properties in comparison with the static treatment.

Experimental

Materials. Straight henequen fibers of 60-70 cm long were originated from Yucatan, Mexico. It was found that the average fiber diameter was in the range of 250 to 350 μm. The fiber bundles were cut to 10 mm long in average for fabricating randomly oriented henequen/PP biocomposites. PP fibers in the entangled fluffy (or woolly) form (P-4203, 10 denier), which can transform to the matrix through melting during biocomposite processing, were supplied from Kolon Glotech Co., Korea. The average PP fiber diameter was about 20-50 μm. The tenacity was 1.5-4.5 g/denier. The henequen and PP fibers were dried at 80 $^\circ$C for 12 h in a vacuum oven prior to use.

Fiber Surface Treatment. 'As-received' henequen fiber bundles were treated with normal tap water by means of soaking and ultrasonic methods at 30 $^\circ$C for 2 h, respectively. The 'as-received' bundles were also treated with 1 wt% and 6 wt% NaOH aqueous solutions for 10 min and 60 min, respectively. The ultrasonic frequency applied was 40 kHz. After alkalization, the fiber bundles were washed with distilled water for several minutes until pH 7 was attained. Hereinafter, for both fiber surface treatments with tap water and NaOH, soaking treatment is referred to as 'static
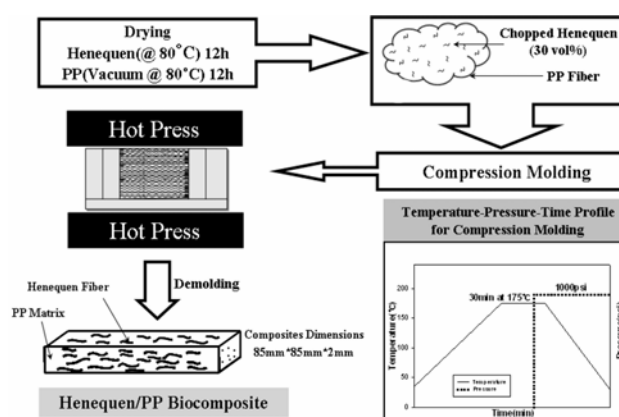

Figure 1. A diagram of the fabrication procedure of henequen/PP biocomposites and the processing profile used in this work.

method', whereas ultrasonic treatment is referred as 'dynamic method' due to the ultrasonic frequency applied.

Biocomposite Fabrication. The henequen fiber content and the average chopped henequen fiber length in henequen/PP biocomposites were 30 vol% and about 10 mm, respectively. The chopped henequen fibers and PP fibers were uniformly mixed and the mixed henequen/PP molding compound was placed in a stainless steel matched-die mold. All biocomposites used were fabricated by a compression molding technique using a hot-press. The molding temperature was 175 $^\circ$C with a holding time of 30 min. The pressure of 1,000 psi was applied after 20 min at 175 $^\circ$C. Finally, the molded biocomposite was cooled down to ambient temperature in the presence of the applied pressure. The dimensions of the henequen/PP biocomposites with randomly oriented henequen fibers were 85 mm×85 mm×2 mm. PP plaques without henequen were also made for comparison. Figure 1 presents the experimental procedure fabricating henequen/PP biocomposites and the temperature-time-pressure profile used to form the biocomposite plaques.

Resin Microdroplet Formation. A single PP filament taken from the PP fibers was placed to be perpendicularly across with a single henequen fiber on a hot plate of about 190 $^\circ$C. The henequen fiber was lifted up as soon as the PP filament was melted again. On this occasion, a tiny amount of the PP resin was covered over the single henequen fiber surface. The resin was rotated about the fiber axis and remelted holding both ends in order to make the PP microdroplet uniform. Then it was cooled down to ambient temperature to be consolidated. Finally, the PP microdroplet was uniformly formed around the single henequen fiber embedded. The procedure for forming a resin microdroplet on a single henequne fiber modified by both static and dynamic methods was repeated to prepare a sufficient number of henequen/PP biocomposite model specimens for single fiber microbonding tests. The formation of henequen/PP microdroplets was observed with an optical microscope
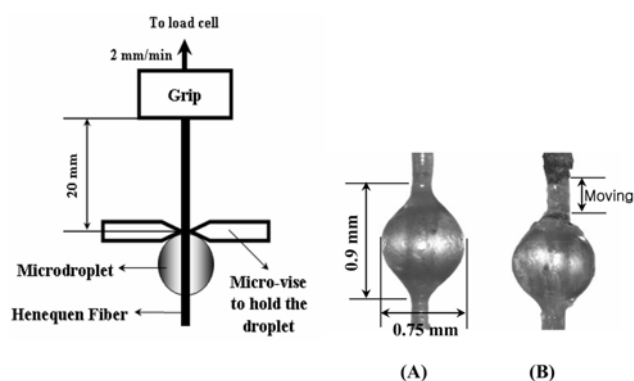
Figure 2. A brief illustration of a single fiber microbonding test method and the optical microscopic views of the typical change of the single henequen fiber embedded in the PP resin microdroplet before (A) and after (B) the test (magnification: ×40).

(Nikon Model ECLIPSE E200).

Single Fiber Microbonding Test. Single fiber microbonding tests were performed with a number of henequen/PP biocomposite model specimens using a universal testing machine (Instron 4467) equipped with a tailor-made testing device, as schematically shown in Figure 2. A load cell of 100 N was used. The crosshead speed was 2 mm/min. The microvise-to-grip distance was 20 mm. The interfacial shear strength (IFSS) for each sample was obtained from the following equation.

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

where τ is the IFSS, F is the force required for debonding the PP microdroplet from the single henequen fiber while tensile loads are applied, D_f is the diameter of the measured natural fiber including the microdroplet, and L_e is the fiber length embedded in the PP microdroplet. The average IFSS value of each sample was obtained from about 30 specimens.

Flexural Test. Three-point bending tests were performed according to ASTM D790M-86 using a universal testing machine (Instron 4467). 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 biocomposite were obtained from 10 specimens. The specimen dimensions were 50 mm×25 mm ×2 mm.

Dynamic Mechanical Analysis. Dynamic mechanical analysis (DMA Q800, TA Instruments) was carried out purging liquid nitrogen with a heating rate of 5 °C/min at a fixed frequency of 1 Hz and with an oscillation amplitude of 20 μm. A single cantilever bending mode was used. The specimen dimensions were 35 mm×12 mm×2 mm. The measurements were conducted from -30 to 100 °C.

Fracture Surface Observation. The fractured surfaces

of each biocomposite without and with fiber surface treatments were observed using a scanning electron microscope (JEOL, JSM-6380). The fracture was done after resting the samples for a sufficient period of time in a liquid nitrogen bath.

Results and Discussion

Interfacial Properties. Figure 3 presents the effect of natural fiber surface treatments on the interfacial shear strength obtained by means of microbonding test for henequen/PP biocomposite model specimens with a single henequen fiber embedded in the PP microdroplet. Here we, for the first time, introduced ultrasonic technique, which is a dynamic method, to treating natural fiber surfaces and compared with soaking technique, which is relatively a static method. The IFSS value of untreated (A) henequen/PP biocomposite was remarkably improved by both static and dynamic surface treatments with tap water and NaOH, respectively. It is noticeable that although the alkali-treated biocomposites (C to F) exhibited higher IFSS values than the water-treated one (B), the latter was also more or less effective to increase the interfacial adhesion between the henequen fibers and the PP matrix. The alkali-treated henequen/PP biocomposite (F) with 6 wt% NaOH for 60 min showed the much improved IFSS value about 134% (static soaking) and about 138% (ultrasonication) greater than the untreated counterpart (A). The water-treated henequen/PP biocomposite (B) at 30 °C for 120 min showed the IFSS value about 19% (soaking) and about 48% (ultrasonication) greater than the untreated case. For the corresponding henequen/PP biocomposite system, the percent improvement of the IFSS was greater by the dynamic ultrasonic method than by the static soaking method. In both static and dynamic methods, the IFSS was increased with the increased NaOH
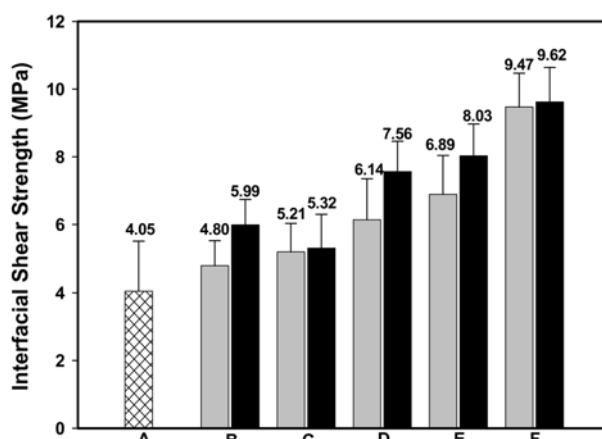

Figure 3. Interfacial shear strengths of henequen/PP biocomposites untreated (A) and treated by static (gray) and dynamic (black) methods with tap water (B) and NaOH (C: 1 wt%, 10 min; D: 1 wt%, 60 min, E: 6 wt%, 10 min, F: 6 wt%, 60 min).
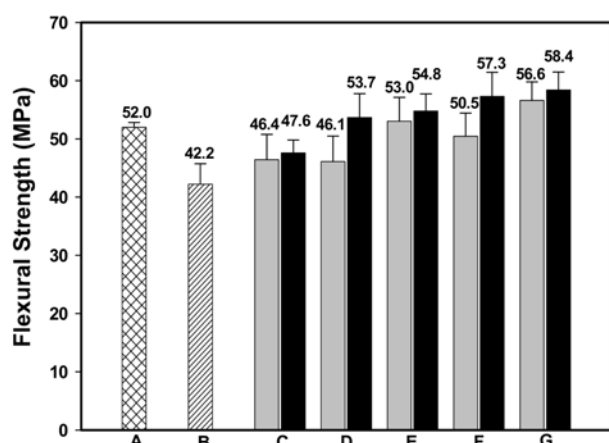
Figure 4. Flexural strengths of PP control (A) and henequen/PP biocomposites untreated (B) and treated by static (gray) and dynamic (black) methods with tap water (C) and NaOH (D: 1 wt%, 10 min; E: 1 wt%, 60 min, F: 6 wt%, 10 min, G: 6 wt%, 60 min).

Figure 5. Flexural moduli of PP control (A) and henequen/PP biocomposites untreated (B) and treated by static (gray) and dynamic (black) methods with tap water (C) and NaOH (D: 1 wt%, 10 min; E: 1 wt%, 60 min, F: 6 wt%, 10 min, G: 6 wt%, 60 min).

concentration and treatment time.

Flexural Properties. In general, flexural deformation occurs at the outside surface of the test specimen. In a fiber-reinforced composite system, the specimen is deflected until a rupture occurs in the outer fibers and the fibers located in the specimen surface may sensitively respond for the applied load. The flexural properties also more importantly influenced with the through-the-thickness behavior in the specimen during the test, depending on the fiber-matrix interaction therein. Therefore, the flexural behavior may reflect the effect of interfacial strength between the reinforcing fibers and the matrix in a composite system. In addition, the flexural test is more relevant with advantages for material design or specification purposes than a tensile test in many practical applications.[23]

Figure 4 demonstrates that the flexural strength of PP was slightly reduced with incorporation of untreated chopped henequen fibers in the present biocomposite system. This may be because the presence of henequen with cellulosic structure in the PP matrix rather decreased the strength, which can be affected by pores and voids therein, especially in the through-the-thickness direction being subject to the three-point bending loads. However, the flexural strength of the untreated henequen/PP biocomposite was improved with both static and dynamic methods of henequen fiber treatment. The improvement was greater with water than with NaOH and also by the dynamic method than by the static one. It was also greater at a higher NaOH concentration and with a longer treatment time, as found in the IFSS result. Compared with the untreated specimen, the greatest improvement of the flexural strength was observed to be about 34% by static soaking and about 38% by dynamic ultrasonication of henequen fibers in 6 wt% NaOH solution for 60 min prior to biocomposite fabrication, respectively.
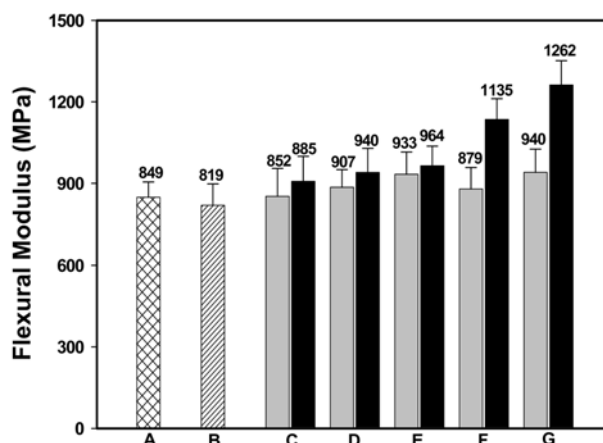
With this treatment, the decreased flexural strength of PP was completely recovered, showing the strengths 9% (static) and 12% (dynamic) greater than the PP control. As a result, it can be said that in the static and dynamic methods, the alkali treatment of henequen fibers significantly contributed to enhancing the flexural strength of untreated henequen/PP biocomposites and it may be ascribed to the improved interfacial strength between the henequen fibers and the PP matrix described earlier.

Figure 5 shows the effect of static and dynamic methods used to treat the henequen fiber surfaces on the flexural modulus of untreated henequen/PP biocomposite. Unlike the flexural strength, the modulus of PP was increased not only by static method but also by dynamic method. This was due mainly to the incorporated natural fibers with a high aspect ratio, which more influence the modulus than the strength. Therefore it is obvious that the biocomposites with the treated fibers had the greater flexural modulus than that with the untreated. The greatest flexural modulus was obtained from the specimens treated with 6 wt% NaOH for 60 min, indicating the improvement of 11% (static) and 49% (dynamic), respectively. The increase of the flexural modulus was greater at a higher alkali concentration and with a longer treatment time. This indicates that the ultrasonic vibration at a high frequency exposed to the henequen fibers during the treatment may modify to greater extent the henequen fiber surfaces making them rougher, as similarly found earlier with different types of natural fibers.[24,25] Therefore it has been suggested that the effect of the present surface treatment method and treatment condition on the interfacial and mechanical properties of a biocomposite system may also depend on the type of natural fibers used.

The increase of the interfacial and flexural properties of the biocomposite by incorporating the treated henequen

fibers may be explained as follows. The henequen fiber surfaces became rougher with more undulation and a greater surface area after alkali treatment. This may lead to the formation of the surface asperity with some troughs along with the longitudinal direction of the fiber, which was resulted from the removed cementing components like hemicellulose and lignin.[26] Such the topography can allow the penetration and wetting of the matrix resin to the fiber surfaces, enhancing mechanical interlocking between the henequen fibers and the PP resin. It may contribute to increasing the interfacial bonding and integrity of the biocomposite. As a result, the flexural strength and modulus were increased, compared with the untreated counterpart.

Dynamic Mechanical Properties. DMA technique is a powerful tool to investigate the dynamic mechanical response on the composite material subject to sinusoidally oscillating deformation at a frequency. Figure 6 exhibits the variations of the storage modulus (E') of PP control and the henequen/PP biocomposites untreated and treated by static soaking

(top) and dynamic ultrasonication (bottom) with tap water and NaOH at different concentration and treatment time, respectively. It is clearly seen that the storage modulus of PP was greatly improved over the whole temperature range by incorporating the chopped henequen fibers into the PP matrix, indicating the higher dynamic stiffness. The storage modulus of untreated henequen/PP biocomposite was significantly increased by both static and dynamic methods, as found from the flexural result above-mentioned. The storage modulus of henequen/PP biocomposite obtained by the dynamic method was greater than that obtained by the static method. In both methods, the E' values of the biocomposites alkali-treated were greater than those treated with tap water.

The greatest improvement was achieved by the dynamic method with 6 wt% NaOH. Especially, in the case of ultrasonication, the alkali treatment was more effective to increase the storage modulus than the water treatment. With a closer inspection, it was found that the higher NaOH concentration gave the greater storage modulus to the biocomposite. Here in the DMA result, the alkali concentration may more contribute to increasing the storage modulus than the treatment time. This dynamic mechanical result is quite consistent with the interfacial shear strength and the flexural properties result in terms of the technical method, the medium, and the condition of the fiber surface treatment given.

Figure 7 shows the variations of the tan δ of PP control and the henequen/PP biocomposites untreated and treated by soaking (top) and ultrasonication (bottom) with tap water and NaOH at different concentration and treatment time, respectively. The intensity of the tan δ peak, which is related to the damping property of a material, of PP control was significantly decreased by incorporating the henequen fibers to the PP matrix resin.

The peak maximum is related with the glass transition temperature (T_g) of the material measured. The T_g of PP control was measured to be about 3 °C and that of the untreated henequen/PP biocomposite was about -1 °C. The T_g of the treated biocomposites with the exception of soaking treatment with tap water was in the range of -9~-5 °C, as listed in Table I. However, the T_g value of the biocomposites was not significantly changed with the surface treatments done. tan δ is defined as the ratio of the loss modulus to the storage modulus or the ratio of the energy dissipated to the energy stored during a dynamic loading cycle.[27] A gradual increase of tan δ beyond 40 °C was found as shown in Figure 7. This is because the change of the loss modulus in PP control and henequen/PP biocomposites was greater than that of the storage modulus at the corresponding temperature. The loss modulus, which was not shown here, was rather slightly increased with increasing temperature beyond 40 °C.

Fracture Surfaces. Figure 8 illustrates the scanning electron micrographs of the fracture surfaces of untreated and
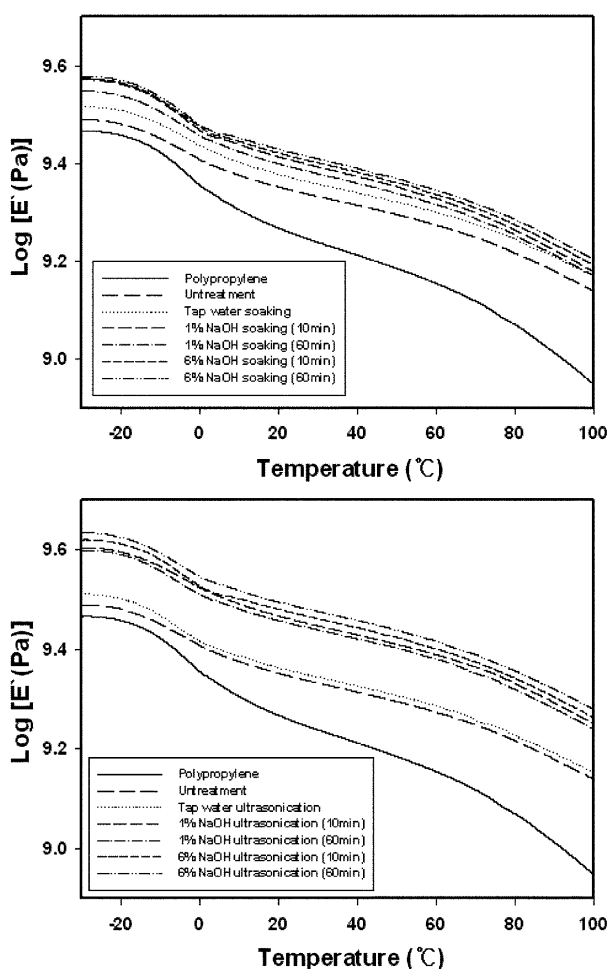
Figure 6. Variations of the storage moduli of PP control and henequen/PP biocomposites untreated and treated by static (top) and dynamic (bottom) methods with tap water and NaOH, respectively.
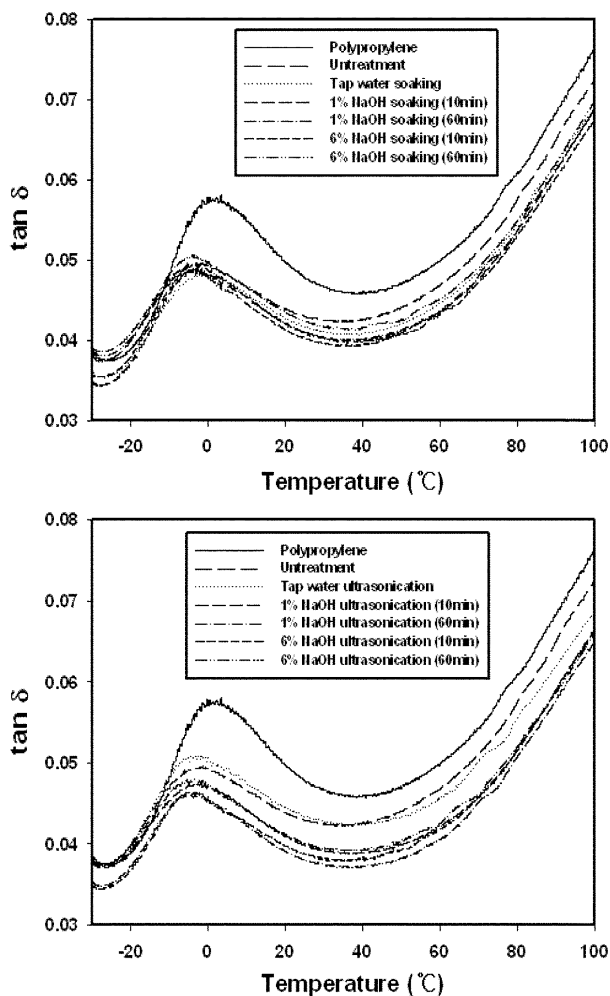
Figure 7. Variations of the tan δ of PP control and henequen/PP biocomposites untreated and treated by static (top) and dynamic (bottom) methods with tap water and NaOH, respectively.

Table I. A Summary of the Glass Transition Temperatures of Polypropylene and Untreated and Treated Henequen/PP Biocompoistes Determined from the tan δ Peak Temperature

Polypropylene		3 ℃
Untreated Henequen/PP		-1 ℃
Treated Henequen/PP	Soaking Method	Ultrasonic Method
Tap Water	0 ℃	-5 ℃
1% NaOH (10 min)	-8 ℃	-7 ℃
1% NaOH (60 min)	-5 ℃	-6 ℃
6% NaOH (10 min)	-7 ℃	-9 ℃
6% NaOH (60 min)	-5 ℃	-7 ℃

treated henequen/PP biocomposites. The SEM observation was focused on the representative single henequen fiber surrounded by the PP matrix in the biocomposite in order to inspect the interfacial bonding between the fiber and the
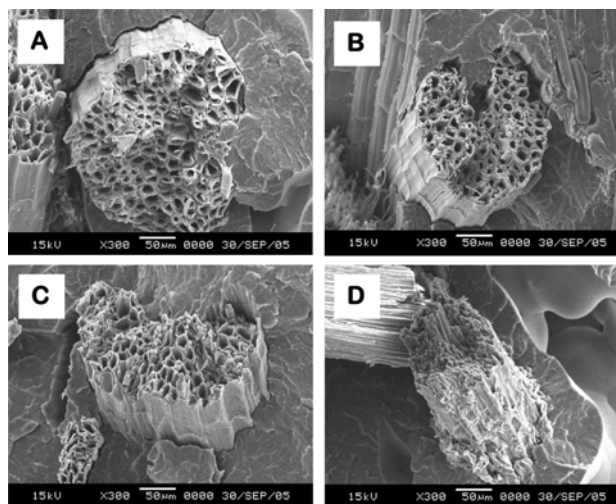

Figure 8. SEM micrographs (×300) showing the fracture surfaces of henequen/PP biocomposites untreated (A) and treated by a static method with tap water (B) and 1 wt% NaOH (C), and by a dynamic method with 6 wt% NaOH (D), respectively.

matrix more closely. The result confirms that the single henequen fiber consists of a number of hollow-type cells forming the cellular structure, as reported earlier.[12] It was found with a closer inspection that the untreated composite specimen (A) had the poor interfacial adhesion between the henequen fiber and the PP matrix. One can see the gap or interstice between the single henequen fiber and the surrounding PP matrix from the micrograph (A).

The interfacial adhesion was more or less enhanced by the soaking treatment with tap water (B) and 1 wt% NaOH for 60 min (C). The adhesion was microscopically observed to be the best by the ultrasonic treatment with 6 wt% NaOH for 60 min (D) among all the specimens. The henequen fiber was broken together with the PP matrix upon fracture, indicating the strongest interfacial adhesion between the fiber and the matrix. The microscopic observations well supported the interfacial and mechanical results, demonstrating that the better interfacial adhesion between the henequen fibers and the PP matrix in the henequen/PP biocomposite significantly contributed not only to increasing the interfacial shear strength but also to improving the flexural and dynamic mechanical properties.

Conclusions

The interfacial, flexural, and dynamic mechanical properties of henequen/PP biocomposites were significantly increased by the surface treatment of henequen fibers with both normal tap water and sodium hydroxide solutions using either soaking treatment, which is a static method, or ultrasonic treatment, which is a dynamic method. The result was well agreed with each other among the interfacial shear strength,

flexural strength and modulus, storage modulus, and fracture surfaces studied with various henequen/PP biocomposites.

It was concluded that the alkali treatment was more effective than the water treatment to improve the interfacial and mechanical properties of randomly oriented chopped henequen/PP biocomposites and, in addition, the ultrasonic method done to each treatment was more effective to improve the properties than the soaking method. The greatest improvement on the interfacial shear strength, flexural properties and storage modulus was achieved by ultrasonic alkalization done with 6 wt% NaOH for 60 min.

The flexural modulus and the storage modulus of PP were significantly enhanced by incorporating chopped henequen fibers and further improved with the treatments performed, due not only to the reinforcing effect but also to the increased interfacial adhesion between the henequen fibers and the PP matrix.

The present result suggests that the interfacial adhesion between a polymer matrix and natural fibers and the related properties, which have been the most important issue in biocomposite systems, may be effectively improved by ultrasonic alkalization at appropriate alkali concentration and treatment time, depending on the nature of natural fibers. Also, it is noticeable that ultrasonic technique for treating the natural fibers has been applied for the first time in the present study. It is also interesting that natural fiber surface treatment with normal tap water may be utilized in combination with ultrasonic technique to enhance the interfacial adhesion of a biocomposite. This treatment may be favorable in practical applications with advantages like cost-effectiveness, environmentally-friendliness, easy handling, and processibility.

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