Fibers and Polymers 2009, Vol.10, No.1, 83-90 DOI 10.1007/s12221-009-0083-z

Fabrication of Long and Discontinuous Natural Fiber Reinforced Polypropylene Biocomposites and Their Mechanical Properties

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(Received July 3, 2008; Revised October 25, 2008; Accepted December 15, 2008)

Abstract: Natural fiber reinforced polypropylene (PP) biocomposites were fabricated by blending long-and-discontinuous (LD) natural fibers (NF) with LD PP fibers. Firstly, random fiber mats were prepared by mixing NFs and PP fibers using a carding process. Then, heat and pressure were applied to the mats, such that the PP fibers dispersed in the mats melted and flowed out, resulting in the formation of consolidated sheets upon subsequent cooling. The effect of the fiber volume fraction on the mechanical properties of the bio-composites was scrutinized by carrying out tensile and flexural tests and observing the interface between the fiber and matrix. It was observed that the natural LD fiber content needs to be maintained at less than the nominal fiber fraction of 40 % by weight for the composites fabricated using the current method, which is quite low compared to that of continuous or short fiber reinforced composites. The limited fiber fraction can be explained by the void content in the biocomposites, which may be caused by the non-uniform packing or the deficiency of the matrix PP fibers.

Keywords: Natural fiber, Long and discontinuous polypropylene, Biocomposite, Mechanical property

Introduction

The so-called advanced composites made from carbon, aramid, silicon carbide, boron, or other higher modulus fibers have been widely used in the aerospace and recreational industries, and their use in the automotive and construction industries has also been explored, where general composites incorporating glass or other relatively low modulus fibers are used [1]. The matrix and reinforcements in advanced and general composites are mostly petroleum based materials and synthetic fibers from organic or inorganic materials, respectively. Nowadays, as industry attempts to lessen its dependence on petroleum based fuels and products, there is an increasing need to investigate more environmentally friendly, sustainable materials to replace the existing ones [2]. The commonly named bio-composite is one of the results of such attempts. The bio-composites referred to herein are composites that combine natural fibers such as kenaf, jute, hemp, and sisal with either biodegradable or non-biodegradable polymers. Natural fibers have many advantages over synthetic ones; no harm to the environment, enhanced energy recovery and biodegradability, low density, high toughness, acceptable specific strength, reduced dermal and respiratory irritation, low cost, renewable resources, etc [3-6]. As regards the matrix used for bio-composites, polyolefin thermoplastics such as polypropylene and polyethylene have been used due to the limited development of biodegradable polymers, in particular for structural applications.

Various fabrication methods have been investigated for biocomposites. These may be classified into two categories according to the types of reinforcement used; particle or short fibers and continuous fibers. For continuous fiber reinforced bio-composites, woven fabric preforms processed from natural fibers have been introduced as the reinforcements. Stocchi *et al*. [7] fabricated a laminated composite with four layers of jute woven fabrics. Prior to their impregnation in the resin matrix, the jute fabrics were treated with alkali in the biaxial tensile stress state. A significant improvement of the mechanical stiffness was achieved in the composite with the fibers treated with alkali under applied stress. Pothan *et al*. [8] also prepared a woven fabric from banana and glass fibers for use in unsaturated polyester bio-composite. In their woven fabric, banana yarns were used for all of the warp yarns, whereas glass yarns comprised the weft yarns by alternating them with the banana yarns. On the other hand, Dhakal *et al*. [9] carried out the low velocity impact testing of hemp fiber reinforced composites, which were prepared using an unsaturated polyester resin and a needle punched non-woven mat of hemp fibers. It was demonstrated in [9] that the total energy absorbed by the hemp fiber reinforced bio-composites was comparable to that absorbed by E-glass fiber reinforced unsaturated polyester composites.

Chopped natural fiber reinforced PP composites have been widely studied in an attempt to benefit from the cost and mechanical properties of these natural fibers. Zampaloni *et al*. [2] reported the fabrication of kenaf fiber reinforced polypropylene sheets that could be thermoformed for a wide variety of applications using a compression molding process utilizing the layered sifting of a microfine polypropylene powder and chopped kenaf fibers. Wambua *et al*. [10] prepared kenaf fiber reinforced PP composites using compression molding by sandwiching PP film with kenaf mats, while Shibata *et al*. [11] fabricated the same composites from PP

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and kenaf fibers by the press forming of stacked layers of their mats. On the other hand, extrusion technology was also adopted to process chopped (50-80 mm) natural fibers with micron size PP powder [12].

For the preparation of thermoplastic composites such as PP, a commingled technique was developed for fabricating continuous or discontinuous fiber reinforced composites. For the preparation of continuous fiber composites, both the reinforcement and matrix fibers are commingled into yarns or fabrics, while both fibers are entangled into nonwoven mats for the preparation of chopped fiber composites. To convert them into solid composites, heat and pressure are applied to the commingled preforms such that only the resin fibers within them melt and flow, forming a continuous matrix phase between the reinforced fibers. This fabrication method has been utilized to manufacture bio-composites using a carding process, which is used to make uniform blends of discontinuous natural fibers such as kenaf or jute with synthetic fibers for use as the matrix [13]. The commingled fabrication method using the carding process may be an effective means of processing bio-composites using long and discontinuous natural fibers, because it can avoid the process of converting them into continuous yarns, which allows the cost to be reduced and the fibers to be uniformly distributed in the composites. Here long fibers refer to fibers whose length is large compared to short fibers with a length of a few millimeters at most. In this study, biocomposites from kenaf and jute fibers and PP matrix fibers, in particular long and discontinuous natural and PP fibers, were fabricated using the commingling concept via the carding process and their mechanical properties were investigated.

Experimental

Materials

In the present study, staple PP fibers (melt flow index: 12 (230 ^{o}C/2,160 g)) were chosen as the matrix material. Kenaf and jute (bast) fibers were selected as the reinforcement. The mechanical and physical properties of these constituent fibers are listed in Table 1. Note that the mechanical properties of the fibers (six averaged values) were measured by using single fiber tensile testing with a specific grip method (as shown in Figure 1). Because of the sensitivity of the small load cell (20 g) used for the single fiber testing, a grip made of thin paper was designed with a hole which can be attached to the upper hook in the tensile machine. The single fiber was glued to the paper in the grip and then stretched at a rate of 10 mm/1 min. It was observed that the mechanical properties of the natural fibers were more broadly scattered than those of the synthetic PP fibers (see Figure 2), because the PP fibers were manufactured to have uniform physical and mechanical properties; however, since the kenaf and jute fibers are grown naturally, their properties can vary immensely from plant to plant.

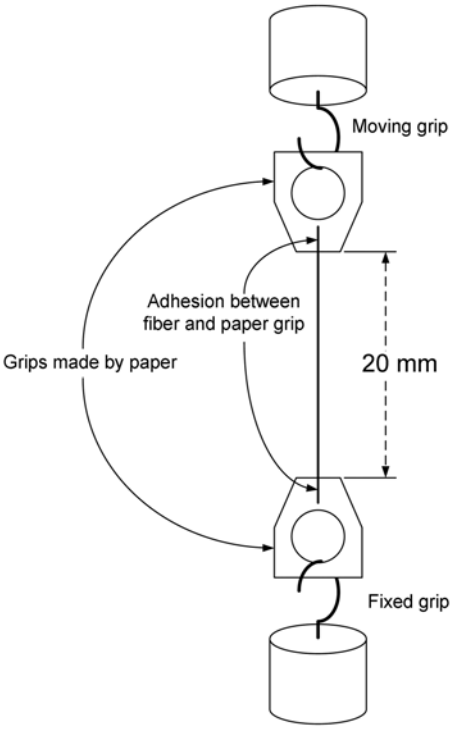

Figure 1. Gripping method for single fiber tensile testing.

Composite Fabrication

Biocomposites consisting of Kenaf/PP fibers and Jute/PP

Table 1. Mechanical and physical properties of natural and PP fibers used in this study

Materials	Young's modulus (MPa)	Tensile strength (MPa)	Density (g/cm^3)	Fiber length (mm)	Fiber diameter (μm)
Kenaf fiber	4,300 (1,600*, 50,000 [a])	250 (62*, 800 [a])	[b]1.4	60	81
Jute fiber	2,200 (660*, 20,000 [a])	345 (114*, 600 [a])	[a]1.4	60	82
PP fiber	780 (74*)	110 (5.5*)	[c]0.91	30	40

*: standard deviation, a: natural fibers, biopolymers, and biocomposites, 2005, b: Ind. Eng. Chem. Res. 1995, 34, 1889-1896, and c: Specification from LG Chem., Ltd.

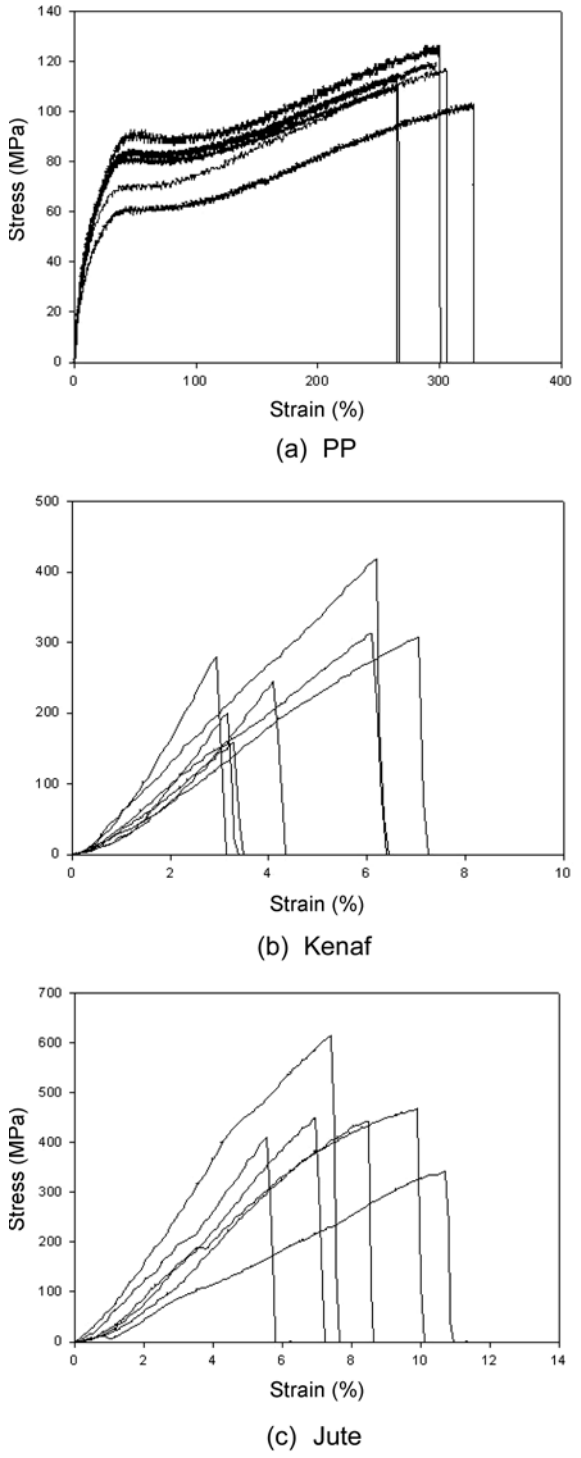

Figure 2. Stress and strain curve of single fiber.

fibers were prepared using a carding machine which enables uniform blends of the natural fibers with the PP fibers to be made. Before the carding process, the natural fibers were dried for 12 hrs at 100 °C. As shown in Figure 3, the natural fibers were fed into the machine with the PP fibers, producing

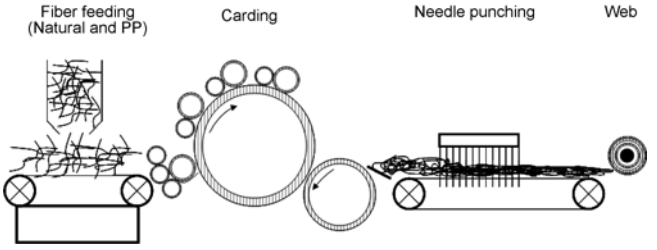

Figure 3. Carding process for mixing natural fibers with PP staple fibers.

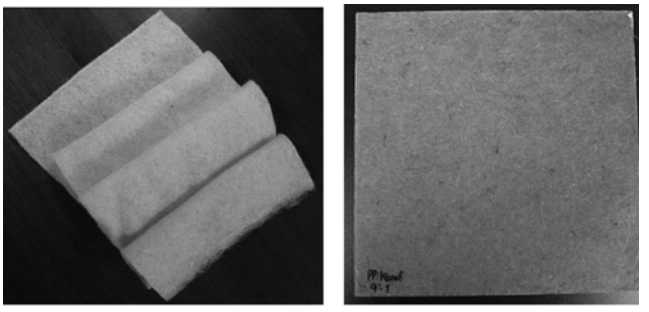

Figure 4. (a) Carded preforms of natural and PP fibers and (b) its consolidated composite.

their mixed preform (see Figure 4(a)). The carded performs were pressed tentatively by hot plates to reduce their volumes for the subsequent consolidation process. Then, the preforms were transferred into a hot press, wherein they were pressed under a pressure of 70 kgf/cm^2 for 5 minutes at 200 °C (see Figure 4(b)). During the hot pressing, the PP fibers melted and flowed out between the natural fibers, forming a continuous matrix phase. Various bio-composites were prepared using the kenaf and jute fibers by varying their nominal fiber weight fractions (10 %, 20 %, 30 %, 40 %, 50 %, and 70 %). Since the nominal fiber weight fractions were measured before the carding and hot pressing processes, they may be different in the bio-composite fabricated from the carded and hot-pressed preforms. Therefore, the fiber fraction in the bio-composites will be denoted as the real fiber fraction, in order to differentiate it from the nominal one.

Mechanical Tests

The tensile tests were carried out according to ASTM D638-03 (dumbbell-shaped specimens) at 23±2 °C and 50± 5 % RH using a Universal Testing Machine (UTM, Zwick Co.). The crosshead speed was set at 5 mm/minute, and the tensile modulus and strength were extracted from the stress-strain curves. Flexural tests were also conducted according to ASTM D790-03 using the UTM with the same crosshead speed as that indicated above. The specimen geometry for the flexural test was 3 mm×12 mm×63 mm in thickness,

Table 2. Fiber fraction by weight and volume in bio-composites

Nominal fiber weight fraction (%)		10	20	30	40	50	70
Fiber weight fraction (%)	Kenaf	8.9	16.6	31.9	38.3	43.8	62.5
	Jute	10.3	19.6	31.2	39.9	52.3	71.2
Fiber volume fraction (%)	Kenaf	6.0	11.4	23.3	28.7	33.6	52.0
	Jute	7.0	13.7	22.5	30.1	41.2	61.2

width and length, respectively.

Fiber Volume Fraction, Void Content and Surface Morphology

Since the mechanical properties of the bio-composites depend on their (real) fiber volume fractions, they were measured after the molding (consolidation) operation, in order to take into consideration the loss of fibers in the carding process. After dissolving the PP in the bio-composites using a solvent, 1,2,4-trichlorobenzene, and being left to stand for 30 hours at 140 °C, the remaining natural fibers were weighed in order to calculate the fiber volume fractions in the composites, as shown in Table 2. The jute fiber reinforced composites showed better integrity during the carding and molding processes, because their weight fractions were only slightly changed compared to those of the kenaf reinforced composites.

The stiffness of fiber reinforced plastic composites is determined by the load transfer capability between the resin matrix and fibers through the shear stress. If voids are formed in the composites, e.g., in the matrix or between the fiber and matrix, they deteriorate the load transfer capability significantly, due to the stress concentration around them [14]. As such, the void content should be characterized for the current LD fiber reinforced composites. The void content of the natural fiber/PP bio-composites was determined using the following formula (ASTM D2734).

$$V = 100(T_d - M_d)/T_d \quad (1)$$

where V, T_d, and M_d are the void fraction (volume %), the theoretical density (g/cm^3) and the measured density (g/cm^3), respectively. T_d was calculated by firstly finding the weight fraction of natural fibers in the bio-composites. Since the densities of the natural and PP fibers were already known, the theoretical density of the bio-composites, i.e. the zero void content case, can be calculated using the volume of the composites and M_d, which is the measured density. As a result, the void contents are calculated using the two density values by means of equation (1).

The fracture surface of the test specimens was observed using SEM (Jeol-5410 LV). For the microscopy examination, the specimen was coated with gold (degree of purity 99.99 %) in order to inhibit electric discharges.

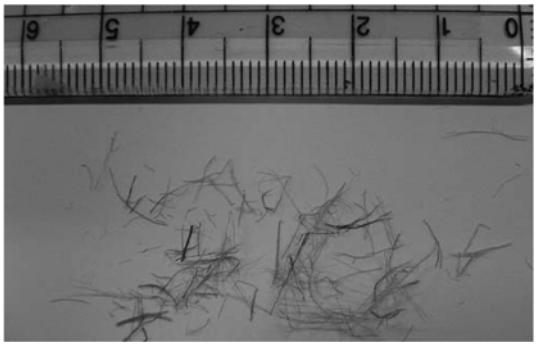

Figure 5. Natural fibers collected after dissolving PP matrix in bio-composites.

Results and Discussion

Fiber Volume Fraction and Voids Contents

Figure 5 shows the natural fibers which were collected after dissolving the PP matrix in the biocomposites using the solvent. The length of the natural fibers was set to 60 mm before the carding process, however after dissolving the PP matrix it was found to be reduced to around 10 mm. This reduction may be due to the breaking of the fibers during the carding and punching process. Another reason may be the molding temperature (200 °C) and the solvent (used for measuring the fiber weight), because the solvent may break the natural fibers which were weakened by the high processing temperature.

After collecting the natural fibers in the biocomposites, the void contents of the bio-composites were calculated using equation (1), as shown in Figure 6. It is clear that the voids in the bio-composites increase as the nominal fiber fraction by weight increases. This tendency is observed in both the kenaf and jute fiber reinforced PP composites, however the void contents in the latter composites were reduced slightly. In general, the voids are closely related to the processing conditions, because they can be formed by gases which may be generated in the thermal process. Since the current study adopted thermoplastic PP and there are few possible sources of gas, the voids may be formed due to the discontinuous resin matrix inside the composites, resulting from the uneven distribution of the PP fibers or their failure to form a continuous phase in the bio-composites. Note that the void content increased significantly as the nominal fiber fraction

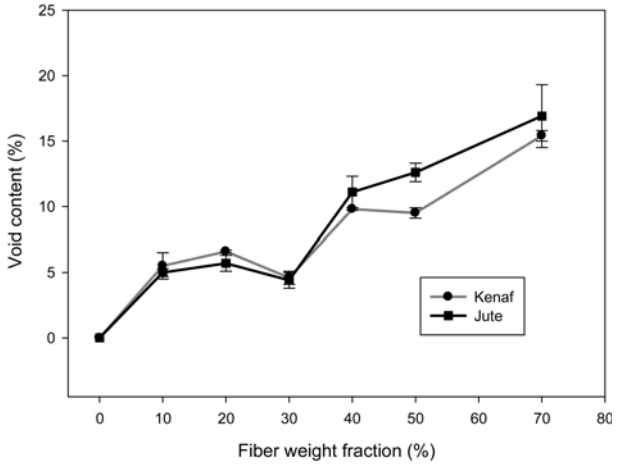

Figure 6. Void contents of natural fiber reinforced PP composites according to nominal fiber fraction by weight.

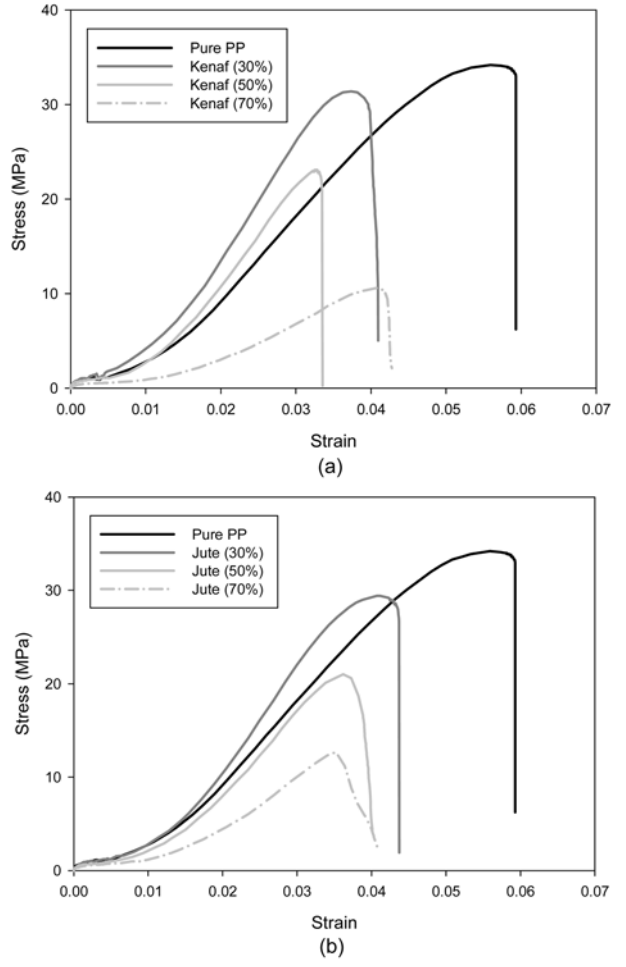

Figure 7. Tensile deformation behavior of the bio-composites; (a) kenaf and (b) jute fiber reinforced composites.

inversion in the mechanical properties can be expected around this fiber fraction.

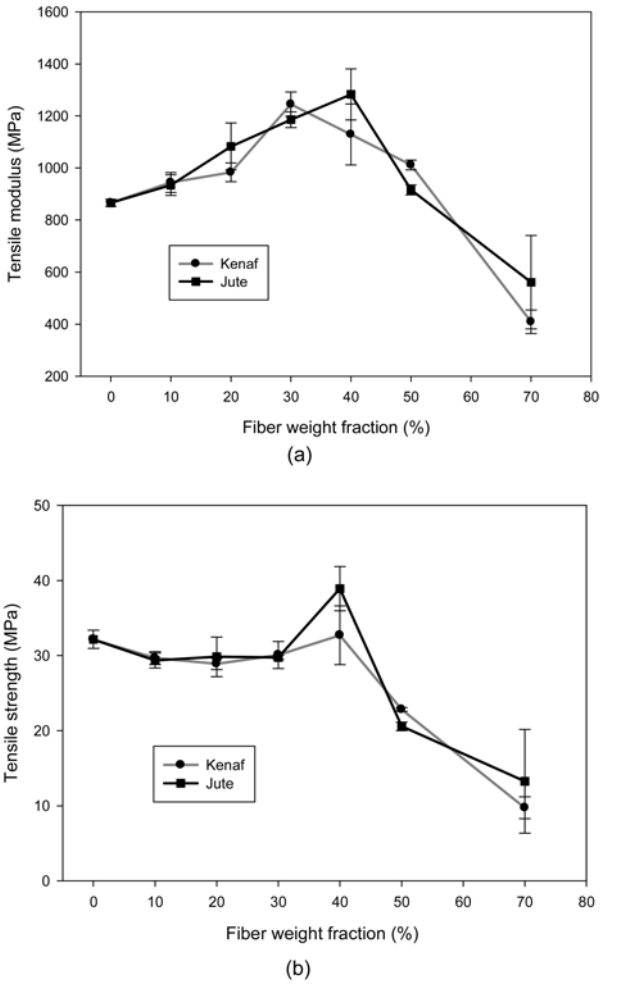

Figure 8. Tensile properties of the bio-composites according to nominal fiber fraction by weight; (a) tensile modulus and (b) strength.

Mechanical Properties of Bio-composites

The tensile behavior of the bio-composites is illustrated in Figure 7 and 8, from which two noticeable features can be identified. Firstly, it is obvious that the tensile strength and breaking strain of the natural fiber reinforced composites becomes lower than that of the pure PP matrix as the nominal fiber fraction increases. This can be explained by both the interfacial adhesion between the matrix and fiber surface and the voids in the composites. Since no coupling agent was introduced to improve the interfacial bonding in this study, mechanical interlocking without any chemical bonding may be responsible for the adhesion. The strength of this mechanical interlocking seems to be insufficient to hold the fiber and matrix together as the composite undergoes large tensile deformation, resulting in low load transfer and, subsequently, low tensile strength and breaking strain. The void contents may be another source of the low tensile strength, because the voids act as a stress raiser (i.e., stress

concentration) that can bring about rapid failure. This tendency can be observed in both the kenaf and jute fiber reinforced composites. A slight increase in strength at the nominal fiber fraction of 40 % (see Figure 8(b)) was observed, which could not be explained at the moment.

Secondly, it can be observed that as the fiber volume fraction increases, the tensile modulus increases until it meets the real fiber volume fraction at about 23 %, beyond which the modulus decreases (see Figure 8(a)). As explained above, the adhesion of the fibers to the matrix came from mechanical interlocking in the current bio-composites and, therefore, the interfacial bonding was not strong enough to bear the large stress build-up. The initial increase of the modulus indicates that the mechanical interlocking was good enough to transfer small loads from the matrix to the fiber. As the fiber volume fraction increases further, the modulus was also decreased, for the same reason (void content) as that in the strength analysis. As a result, it can be concluded that when the current fabrication method (carding process) is employed to fabricate long and discontinuous natural fiber/PP fiber composites, the fiber volume fraction should be maintained at less than the critical value, e.g., 23 % in the current study or 30 % in terms of the nominal fiber fraction by weight, to ensure that minimum voids are formed from the discontinuous matrix resin.

The fracture surfaces of the jute/PP bio-composite were investigated using the SEM micrograph in Figure 9. The jute/PP bio-composite with a nominal fiber fraction of 30 % (by weight) contains more continuous PP matrix than any of the higher fiber fraction bio-composites. The jute fibers in Figure 9(a) seem to be surrounded by the PP matrix, whereas some of them were pulled out during the tensile test. Note that the voids can be observed in Figure 9(b) and (c), which is consistent with the quantitative analysis of the void content (see Figure 6).

The flexural properties of the natural fiber/PP bio-composites were characterized using ASTM D638-03. The flexural test allows the combination of the tensile and compressive properties to be examined. As the fiber volume fraction increases, the flexural strength decreases, as shown in Figure 10, which is the same tendency as that observed in the tensile test. However, the flexural modulus of both the kenaf and jute fiber reinforced bio-composites, in particular those with a volume fraction of around 23 % (or a nominal fiber fraction of 30 % by weight), was improved. As a result, it can be confirmed that the resistance to bending (combined tensile and compressive deformation) was improved by the natural fibers in the case of a small deformation, indicating that the adhesion plays the same role in the case of both tensile and compressive deformation.

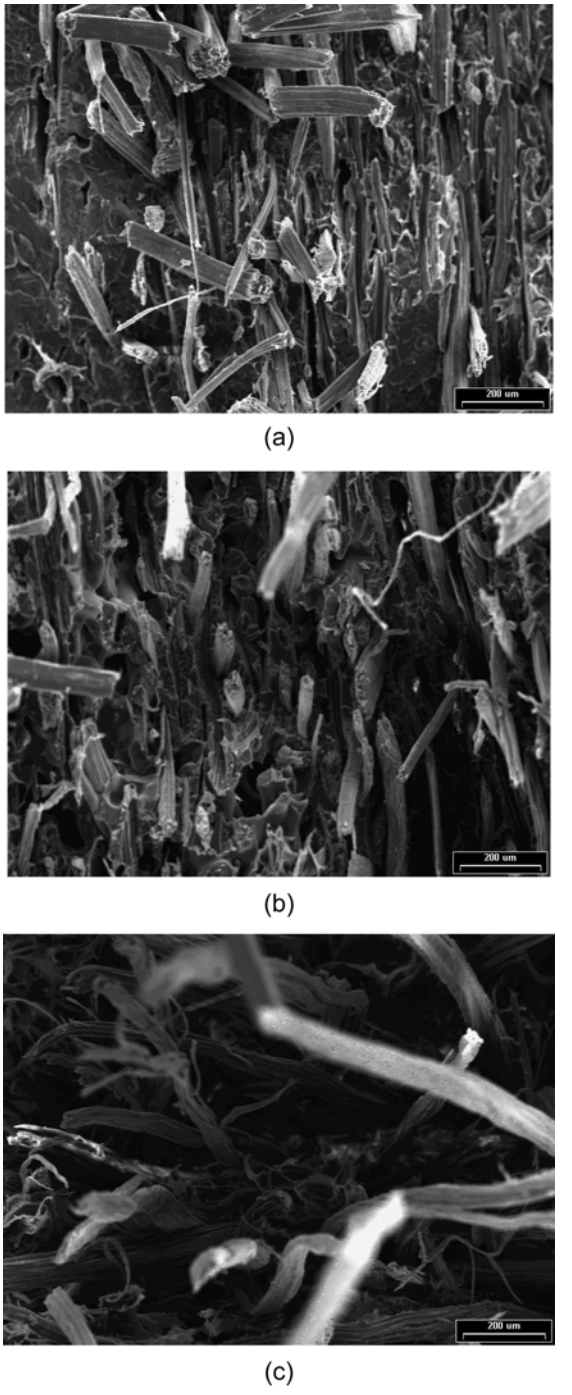

Figure 9. SEM micrographs of the fracture surface of jute fiber reinforced bio-composite with nominal fiber fractions of (a) 30 %, (b) 50 %, and (c) 70 %.

Prediction of the Elastic Modulus

The elastic modulus of the short fiber reinforced composites can be predicted using the various equations and models developed by Eshelby, Mori-Tanaka, Halphin-Tsai, Cox, etc [15]. Herein, Tsai and Pagano's model [16] was adopted to investigate the possibility of predicting the elastic modulus of the LD natural fiber reinforced composites. Tsai and Pagano's equation is given by:

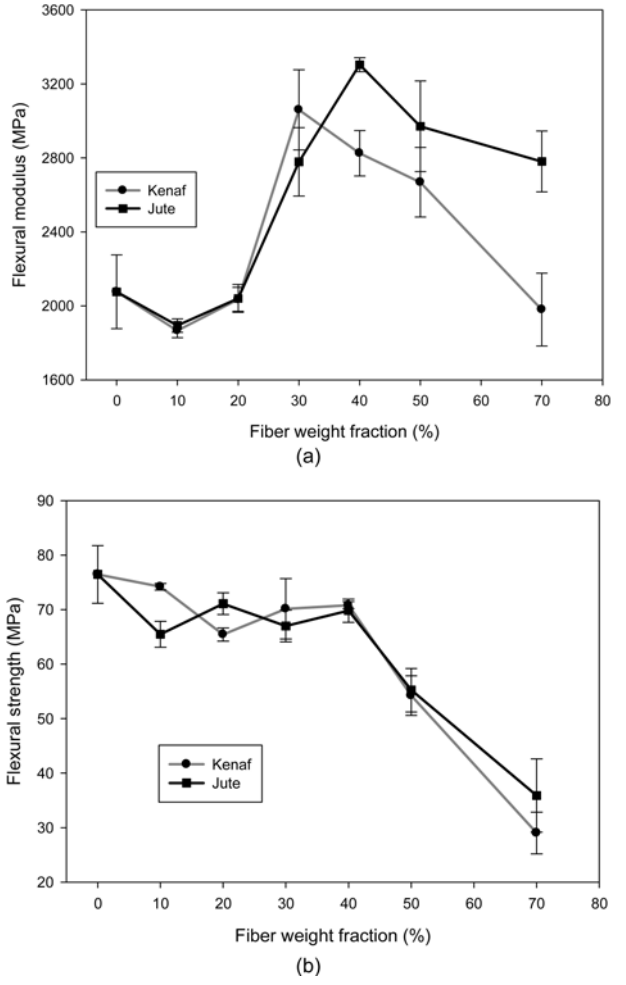

Figure 10. The flexural deformation behavior of the bio-composites according to nominal fiber fraction; (a) flexural modulus and (b) strength.

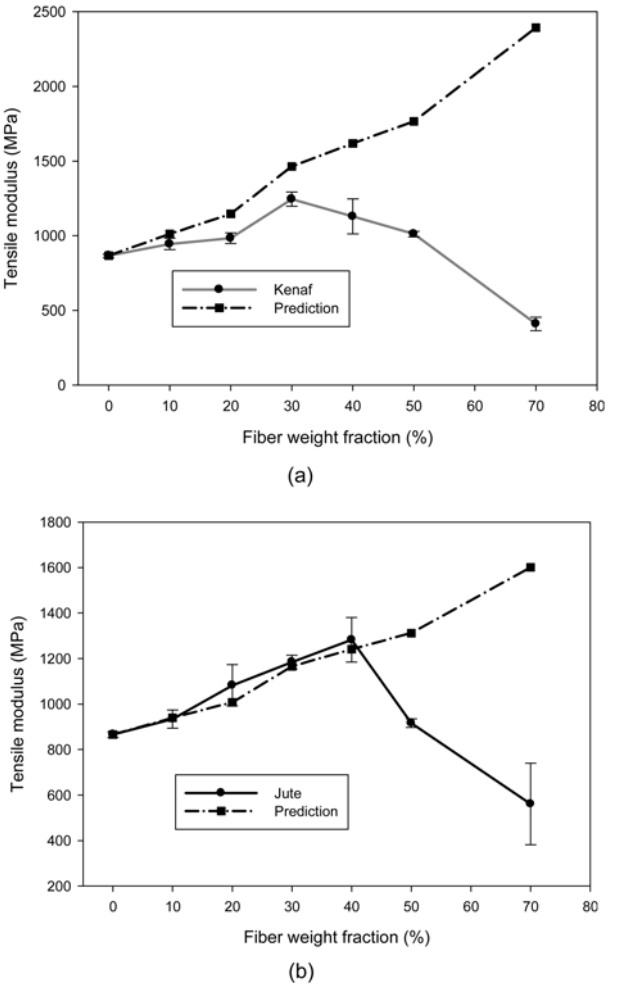

Figure 11. Theoretical prediction of the tensile modulus of the bio-composites; (a) kenaf and (b) jute fiber reinforced composites.

$$E = \frac{3}{8}E_1 + \frac{5}{8}E_2 \tag{2}$$

where E_1 and E_2 are the elastic moduli of randomly oriented fiber reinforced composites given by the Halpin-Tsai equations [17] as follows.

$$\frac{E_i}{E_m} = \frac{1+\xi_i \eta_i v_f}{1-\eta_i v_f},\ \eta_i = \frac{E_f/E_m - 1}{E_f/E_m + \xi_i},\ \xi_i = \begin{cases} 2(l_f/d_f) & i=1 \\ 0.5 & i=2 \end{cases} \tag{3}$$

where E_f and E_m are the moduli of the fiber and matrix, respectively, and v_f, l_f, and d_f are the fiber volume fraction, fiber length, and fiber diameter, respectively. Using the data in Table 1 and 2, the tensile moduli were calculated for both the kenaf and jute reinforced biocomposites (see Figure 11). The prediction matched well with the experimental values for the low fiber volume fraction case (less than 30-40 % by weight) where the void contents were not high. These results indirectly confirm that the LD bio-composites fabricated from the carding, punching and pressing operations are randomly oriented fiber composites and that further consideration of the equation may be required to properly predict the elastic modulus of the composites including the voids.

Conclusion

Long and discontinuous natural fiber (kenaf and jute) reinforced polypropylene composites were fabricated using the carding and punching processes followed by hot press compression molding. It was concluded that the kenaf fiber reinforced PP composites have an optimum nominal fiber fraction of 30 % by weight at which the tensile and flexural modulus are the highest, while the reduction in strength is minimal. For the jute fiber reinforced composites, a fiber fraction of 40 % (by weight) seems to be the optimum value. The limited fiber fraction was explained by the void content

in the bio-composites, which may be caused by the non-uniform packing or the deficiency of the matrix PP fibers. To incorporate more natural fibers into the bio-composites, the fiber length of both the natural and PP fibers may need to be shortened; however, too short fibers may spoil the processibility of the carding operation, thus further experimental or theoretical studies are necessary to determine the optimum fiber fraction.

Acknowledgements

This project work was initiated through research grants (2006-A046-03) supported by the Korea National Cleaner Production Center (KNCPC). The author (W.-R. Yu) also would like to thank the Korea Science and Engineering Foundation (KOSEF) for sponsoring this research through the SRC/ERC Program of MOST/KOSEF (R11-2005-065).

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