Research Article

Mechanical Properties and Biodegradability of the Kenaf/Soy Protein Isolate-PVA Biocomposites

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The effective utilization of original natural fibers as indispensable components in natural resins for developing novel, low-cost, eco-friendly biocomposites is one of the most rapidly emerging fields of research in fiber-reinforced composite. The objective of this study is to investigate the interfacial adhesion properties, water absorption, biodegradation properties, and mechanical properties of the kenaf/soy protein isolate- (SPI-) PVA composite. Experimental results showed that 20 wt% poly (vinyl alcohol) (PVA) and 8 wt% glutaraldehyde (GA) created optimum conditions for the consolidation of the composite. The increase of interfacial shear strength enhanced the composites flexural and tensile strength of the kenaf/SPI-PVA composite. The kenaf/SPI-PVA mechanical properties of the composite also increased with the content of cross-linking agent. Results of the biodegradation test indicated that the degradation time of the composite could be controlled by the cross-linking agent. The degradation rate of the kenaf/SPI-PVA composite with the cross-linking agent was lower than that of the composite without the cross-linking agent.

1. Introduction

Biodegradable polymers that are derived from natural resources are potential substitutes for existing petroleum-based synthetic polymers, owing to their low cost, easy availability, and complete biodegradability [1]. Among various cellulose, starch, and protein materials, soy protein isolate (SPI) is used to form packaging and edible film because of its excellent film forming abilities, good biodegradable performance, and high barrier against oxygen and oil in low humidity conditions [2, 3]. However, the low mechanical properties and high moisture sensitivity of soy protein-based plastics have limited their usage [4]. These characteristics are attributed to the inherent hydrophilicity of natural proteins and the amount of hydrophilic plasticizer incorporated into them. Moreover, literature [5] has confirmed that SPI films that lack secondary components do not show good mechanical and barrier properties.

Soy protein is commercially available as SPI, soy protein concentrate (SPC), and soy flour (SF). Chemically, SPI contains 90% protein, while SPC contains 70% and 18% carbohydrates and 6% ash, with fiber and moisture making

up the remaining components. SF contains up to 55% protein and 32% carbohydrates. Soybean protein contains several amino acids, such as glutamic acid, arginine, lysine, cysteine, and aspartic acids, which have polar groups. These groups can act as useful cross-linking and/or hydrogen bonding sites to improve the mechanical properties of soy protein polymers. In the present research, SPI was modified with GA to increase its mechanical and physical properties and to improve thermal stability and processability as a matrix for composite fabrication. Several researchers have studied the cross-linking of GA with proteins and confirmed the reaction mechanisms [6]. Biswas et al. [7] and Gellerstedt and Gatenholm [8] have proposed that the GA reacts with the amino groups in protein to form cross-links. Zini et al. [9] used maleinized tung oil to cross-link the SPI and improve its tensile properties. A typical cross-linked structure of soy protein with the GA is shown in Figure 1 [6]. It was, however, difficult to assess the average degree of cross-linking because of the complexity of the chemistry. The cross-linking was judged based on the improved tensile strength and interfacial adhesion properties and decreased fracture strain and moisture absorption after the GA modification as well as

FIGURE 1: Schematic structure of the glutaraldehyde (GA) cross-linked soy flour.

the significant increase in the viscosity immediately following the addition of GA to the SPC solution [10].

Another approach to modify the moisture sensitivity of SPI films is to blend other natural or synthetic polymers with SPI materials [11]. Normally, biodegradable polymers such as gelatin, starch, cellulose, and protein are used as blend components to ensure that the final films can still be considered “green.” However, because natural polymers are inherently hydrophilic, these blends cannot solve the moisture sensitivity problem of SPI films. In addition to natural materials, PVA, the world’s most produced synthetic, water-soluble polymer, can be blended with natural polymers to form biodegradable composites. These PVA/natural polymer blends have promising industrial applications in many fields because of their biodegradability, biocompatibility, chemical resistance, and excellent physical properties [11].

Biocomposites composed of natural fibers and synthetic or natural polymer matrices have gained recent attention due to their cost effectiveness, low density, biodegradability, ready availability, energy recovery, and CO₂ sequestration [12]. The natural fibers commonly used to reinforce biocomposites are jute, flax, hemp, ramie, sisal, bamboo, and kenaf fiber [13]. The renewable and biodegradable characteristics of these natural fibers enable disposal processes, which are not possible with most industrial fibers.

Among others, kenaf offers the particular advantages of a fiber crop, including rapid growth in various climatic conditions and, subsequently, the prompt accumulation of carbon dioxide [14]. Kenaf fiber, obtained by processing the bark of the kenaf plant, exhibits low density and nonabrasiveness during processing, highly specific mechanical properties, and biodegradability. Due to their similarities, kenaf can be used either as an alternative to or in admixture with jute. In 1995, kenaf was priced at \$400 per tonne and from \$278 to \$302 per tonne in 2000 [15]. It takes 54 MJ to produce 1 kg of glass fiber, but only 15 MJ of energy to produce 1 kg of kenaf. Kenaf has been largely used to reinforce thermoplastic polymers [16, 17] and, recently, thermoset polymers [18]. Kenaf fibers

have also been used as nonwoven mats in the automotive, textile, fiberboard, civil, and electronic industries [19–21].

Until recently, studies on SPI/PVA blends reported the use of plasticizers and cross-linking agents to increase mechanical and physical properties, improve thermal stability, reduce moisture absorption, and improve processability as a resin for composite fabrication. Biocomposites reinforced with kenaf fiber that use a SPI/PVA blend have not been reported, despite their many advantages of biodegradability, biocompatibility, chemical resistance, and excellent physical properties. In this study kenaf nonwovens were used to fabricate environment friendly biocomposites using modified SPI-based resins. SPI was modified using PVA in order to improve the interfacial bonding of kenaf nonwoven/SPI and then cross-linked with GA to improve its mechanical properties and water resistance. The biodegradation of the biocomposites was analyzed to determine the effect of PVA and the cross-linking agent on the composting of the kenaf composites.

2. Experimentals

2.1. Materials. SPI with an approximate protein content of 90% was supplied by Solea Company, USA. GA (grade II, 25% solution), glycerol, and PVA (98% hydrolyzed, average molecular weight (Mw) of 89,000–98,000) were obtained from Sigma-Aldrich Chemical Company. Kenaf nonwovens with an areal density of 400 g/m² were obtained from KumHa Co. Ltd. (Korea).

2.2. Preparation of the SPI/PVA Film and Kenaf/SPI-PVA Composite. The SPI/PVA films were prepared using a casting method. First, the SPI (10 wt%) and PVA (5 wt%–20 wt%) were dissolved in water (pH 8) and cured at 70°C for 25 minutes. Second, 20 wt% glycerol was added according to the optimum condition [22]. Prepared solution was casted on a glass plate and dried at 40°C for 24 hours. Finally, the SPI/PVA film was cured at 140°C via hot pressing for 10 minutes, under a pressure of 7 MPa.

FIGURE 2: Manufacturing process of the SPI/PVA film.

Kenaf nonwovens were dewaxed by soaking them in a mixture of ethanol and benzene (1 : 2) at 50°C for 5 hours and then washed with distilled water and air dried. The dewaxed nonwovens were immersed in a 10 wt% aqueous sodium hydroxide solution at 30°C for 1 hour and then washed with distilled water and dried. To remove any remaining moisture, the pretreated kenaf nonwoven was cut into 20 cm by 20 cm pieces and dried at 100°C for 2 hours in a dryer and at 80°C for 3 hours in a vacuum oven. Kenaf nonwoven reinforced composites were fabricated using SPI (10 wt%)/PVA (15 wt%) resins added glycerol (20 wt%)/GA (0 wt%~25 wt%). The manufacturing process of the SPI/PVA film and kenaf/SPI-PVA composite are shown in Figures 2 and 3, respectively. The purposes of adding glycerol in kenaf/SPI-PVA composite were to improve the toughness and to make the kenaf/SPI-PVA composite more flexible.

2.3. Measurements

2.3.1. Water Absorption. The initial weight of the kenaf/SPI-PVA composite with the cross-linking agent was measured and the specimen submerged in distilled water at 25°C for 12 hours. Water absorption was calculated using (1), in which W_{wet} and W_{dry} represent weight of the sample after immersed in water and initial weight of the sample, respectively:

$$\text{Water absorption (\%)} = \frac{W_{\text{wet}} - W_{\text{dry}}}{W_{\text{dry}}} \times 100. \quad (1)$$

2.3.2. Mechanical Properties. To investigate the effects of PVA and the cross-linking agent on the mechanical properties of SPI, tensile and flexural (three-point bending) tests were performed at a cross-head speed of 5 mm/min using an Instron

(model 4467), according to ASTM D638 and ASTM D790, respectively. The value of each mechanical property was determined by an average of ten specimens.

2.3.3. Interfacial Adhesion Test. The interfacial shear strength (IFSS) between the kenaf fiber and SPI was measured by a microdroplet debonding test using an Instron test system (model 4467) equipped with a 500 N load cell. The microdroplet test specimen of kenaf fiber with SPI was made to composite at 80°C for 24 hours. Figure 4 shows the test grip and microvise for the microdroplet debonding test. Tests were performed at a cross-head speed of 1 mm/min at room temperature. The maximum force (F_{max}) was recorded to calculate the interfacial shear strength (τ_{max}) using (2), in which D and L represent fiber diameter and fiber embedded length, respectively:

$$\tau_{\text{max}} = \frac{F_{\text{max}}}{\pi DL}. \quad (2)$$

2.3.4. Morphology. The surface and fracture morphologies of the kenaf/SPI-PVA composite were observed with a scanning electron microscope (SEM, S4700, HITACHI).

2.3.5. FTIR Analysis. A Fourier transform infrared spectrophotometer (FT-IR, Bruker Optic GmbH, ALPHA-P) equipped with an attenuated total reflectance (ATR) accessory was used to examine the surface composition of the uncross-linked and cross-linked kenaf/SPI-PVA composite. The spectra were recorded in the transmission mode in the range of 4000–500 cm^{-1} . FT-IR spectra were measured at a spectral resolution of 4 cm^{-1} , and the spectra were obtained with an accumulation of 128 scans for a high signal-to-noise ratio.

FIGURE 3: Manufacturing process of the kenaf/SPI-PVA composite.

FIGURE 4: Photograph and schematic diagram of the microdroplet debonding test sample and test setup.

2.3.6. Biodegradation. Test specimen of the kenaf/SPI-PVA composites containing the cross-linking agent was prepared in equal amounts of the size of 20 mm × 20 mm. They were dried in a vacuum oven for 2 hours. Specimens were mixed with the compost in a test bath (as shown in Figure 5) at $58 \pm 2^\circ\text{C}$, using a constant temperature bath. To maintain aerobic environment, air was injected continuously at 200 cc per minute during the test. Weight changes were measured after the specimen was dried in a vacuum oven for 12 hours at 50°C .

3. Results and Discussion

3.1. Effect of PVA Content on SPI/Glycerol Film

3.1.1. Tensile Properties. Figures 6 and 7 show the effect of PVA content on the tensile properties of SPI film without and with glycerol. The following results can be concluded from the data: (Figure 6) For the SPI/PVA films without glycerol, tensile strength decreased with the increase of PVA

FIGURE 5: Schematic diagram of biodegradation test chamber.

content. (Figure 7) Comparing to the SPI/PVA films without glycerol, after adding glycerol (20 wt%), tensile strength of each SPI/PVA film increased and tensile modulus of it decreased with the content of glycerol. These behaviors are due to the plasticization effect of glycerol. Consequently, 15 wt% of PVA was chosen as an optimum condition. Enhancement of mechanical properties is attributed to the long-chain PVA molecules which contain many negative OH groups, forming strong intra- and intermolecule interactions with the protein molecules. These interactions may include hydrogen-bonding, dipole, and charge effects. In addition, blending long molecules in SPI could bring about molecular entanglements, which in turn will improve the mechanical properties of SPI [23].

FIGURE 6: Tensile properties of the SPI film without PVA content.

FIGURE 7: Tensile properties of the SPI/glycerol film with PVA content.

3.1.2. FTIR Analysis. The FTIR spectra of the SPI/glycerol film with and without PVA are shown in Figure 8(a). Pure glycerol is known to show absorption peaks corresponding to C-C and C-O groups in the fingerprint region from 800 cm^{-1} , 1150 cm^{-1} wavenumbers as reported by Lodha and Netravali [24]. The FTIR spectra of the SPI/glycerol without PVA show five peaks at 850 cm^{-1} , 900 cm^{-1} , 925 cm^{-1} (C-C skeletal vibrations), 1045 cm^{-1} (C-O stretch at C^1 and C^3), and 1117 cm^{-1} (C-O stretch at C^2) wavenumbers indicating the presence of glycerol on the specimen surface. Similar results for SPI/glycerol film have also been previously reported [24, 25]. Also, the absorption peak of 1480 cm^{-1} to 1570 cm^{-1} , 1600 cm^{-1} to 1700 cm^{-1} , and 3294 cm^{-1} in SPI refers to the hydrogen bond between protein chains and moisture in protein and carbonyl groups [23].

Figure 8(b) shows the FTIR spectra of SPI/glycerol film with PVA. The basic structure of the PVA molecule is -OH groups on carbon chains. The broad -OH absorption band was observed in the wavenumber range of 2918 cm^{-1} to 3565 cm^{-1} . As can be seen in spectra of the SPI/glycerol film with PVA, new absorption bands appeared in comparison

(a) SPI/glycerol without PVA
(b) SPI/glycerol with PVA

FIGURE 8: FTIR spectra of SPI/glycerol without and with PVA.

to the SPI/glycerol film without PVA. The absorption bands at 1600 to 1400 cm^{-1} and 1150 to 1250 cm^{-1} are attributable to -NH-, C-N stretching, and N-H bending (amide III) vibrations, respectively. A typical characterization of these spectra is the disappearance of the strong hydrogen bond appearing both in SPI and PVA spectra, whereas a new absorption band at 2900 to 3100 cm^{-1} appears. Normally, the absorption band of 2918 to 3565 cm^{-1} accords with the lapped characteristic band of -OH for moisture in SPI and PVA as mentioned earlier [23].

3.1.3. Morphology Observation. Figures 9(a) and 9(b) typically show the SEM photographs of SPI/glycerol film without and with PVA. Obviously, SPI/glycerol films can be fabricated smoothly without cracks and microholes. With the addition of PVA in SPI/glycerol, the surfaces have some ripples, which might owe to the toughening effect of little molecules.

SEM photographs of the fractured surface of the SPI/PVA film after the tensile test are shown in Figure 10(a). The SPI film without PVA showed a brittle fracture behavior and a shear fractured cross section due to fast crack propagation. With the increase of PVA content in the SPI, the blend materials become less brittle, as shown in Figures 10(b), 10(c), and 10(d).

3.2. Effect of GA Content on Kenaf/SPI-PVA Composites

3.2.1. FTIR Analysis. The FTIR spectra of the uncross-linked and cross-linked kenaf/SPI-PVA composite are shown in Figure 11. As can be seen in spectra of the cross-linked kenaf/SPI-PVA composite, most characteristic absorption bands appear with addition of SPI, PVA, and GA. As mentioned earlier in Section 3.1.2, the cross-linking of the kenaf/SPI-PVA composite was confirmed by strong hydrogen bond, -NH-, C-N stretching, and N-H bending vibrations after curing treatment, as compared to uncured kenaf/SPI-PVA composite.

FIGURE 9: SEM photographs of surface of (a) SPI/glycerol without PVA, (b) SPI/glycerol film with PVA.

FIGURE 10: SEM photographs of tensile fracture surface of (a) neat SPI/glycerol film, (b) SPI/glycerol film with PVA (5 wt%), (c) SPI/glycerol film with PVA (15 wt%), and (d) SPI/glycerol film with PVA (20 wt%).

3.2.2. Interfacial Adhesion Properties. The interfacial adhesion strength was examined whether the addition of GA could improve adhesion between the kenaf fiber and the SPI. The average of thirty specimens was used to evaluate the IFSS as a manifestation of adhesion strength. The IFSS of the kenaf/SPI-PVA composite according to the added amount of GA is shown in Figure 12. The IFSS showed a maximum value of 15.03 MPa with the addition of 8 wt% GA. However, the IFSS of the kenaf/SPI-PVA composite with an added GA

of 16 wt% and 25 wt% decreased, indicating that the blend becomes brittle when highly cross-linked with GA.

The effect of the IFSS on tensile properties of the kenaf/SPI-PVA composites is shown in Figure 13. There is a proportional relationship between tensile strength and IFSS. Tensile strength of the kenaf/SPI-PVA composites increased from 17 MPa to 23 MPa when the IFSS was raised from 10.14 MPa to 15.03 MPa, as compared to 10.14 MPa IFSS. The increased tensile strength of the kenaf/SPI-PVA composites

FIGURE 11: FTIR spectra of the uncross-linked and cross-linked kenaf/SPI-PVA composite.

FIGURE 12: Interfacial adhesion properties of the kenaf/SPI-PVA composites with GA content.

FIGURE 13: Relationship between tensile strength and IFSS of the kenaf/SPI-PVA composites.

indicates a good adhesion between kenaf fiber and SPI/PVA matrix when 8 wt% GA is added.

3.2.3. Mechanical Properties. Flexural and tensile properties of the kenaf/SPI-PVA composite with GA are shown in Figures 14 and 15. In cases of 16 wt% GA content, the flexural strength and modulus of the kenaf/SPI-PVA composite increased to 39 MPa and 2662 MPa, respectively. However, the flexural strength and modulus slightly decreased at the higher GA content, as shown in Figure 14. The slight decrease of flexural properties is well known such that internal stresses occur during the excessive cross-linking of thermosets and these residual stresses play an important role in flexural fracture. Residual stress build-up has been extensively studied in epoxy polymers and acrylate networks, and so forth [26]. It has to be noticed that excessive residual stresses in matrix and interface have been a reason of low compressive or flexural stress.

In Figure 15, while there appears to be little difference in tensile strengths of the kenaf/SPI-PVA composites with GA content, most of tensile strength is higher than those of the uncross-linked composite. Tensile modulus of the kenaf/SPI-PVA composite showed an increasing tendency with GA, while tensile strength did not show it. The increase in tensile modulus with GA addition can be attributed to the cross-links formed by GA with kenaf/SPI-PVA. Park et al. [27] showed that cross-linking SPI with GA increased the tensile properties from 8.3 to 14.9 MPa. They suggested that the covalent intermolecular and intramolecular cross-linking between soy protein and GA increased the mechanical properties of SPI/GA films.

3.2.4. Water Absorption. Water absorption properties of the cross-linked kenaf/SPI-PVA composite with the cross-linking agent after immersion in a water tank at 25°C for 12 hours are shown in Figure 16. Most of the water absorption of kenaf/SPI-PVA composite decreased with the cross-linking agents. As the GA content increased, water absorption decreased from 40.5% to 22.2%. Due to the cross-linking effect of GA, compact structure of the composite less absorbs the external moisture.

3.2.5. Fracture Surface. SEM photographs of the surface and fracture surface of the kenaf/SPI-PVA composites are shown in Figure 17. The surface and fracture surface of the composite without a cross-linking agent are shown in Figure 17(a), in which the kenaf fiber does not fully adhere to the SPI/PVA resin. The kenaf/SPI-PVA composites with a cross-linking agent were observed smooth interfacial bond between kenaf and SPI-PVA. As the interfacial bond mainly depended on the number of entangled chains forming connections across the interface, the strong bond indicated that the molecules of kenaf/SPI-PVA composites could yield more entanglement at 8 wt% GA. Obviously, many of cracks were observed in Figure 17(d) of the composite having 25 wt% of glutaraldehyde. These cracks might affect mechanical properties and fracture of the composite. Also, internal stresses in composites might occur during the excessive cross-linking as mentioned earlier.

FIGURE 14: Flexural properties of the kenaf/SPI-PVA composites with GA content.

FIGURE 15: Tensile properties of the kenaf/SPI-PVA composites with GA content.

FIGURE 16: Water absorption of cross-linked kenaf/SPI-PVA composites with GA content.

3.2.6. Biodegradation Properties. The weight changes of the kenaf/SPI-PVA composite specimen, according to biodegradability, are shown in Figure 18. The weight of the kenaf/SPI-PVA composite without GA (a) decreased by approximately 22% after 5 days and approximately 43% after 20 days in a compost condition. The kenaf/SPI-PVA composite film with 8 wt% GA (b) showed a lower value of weight loss than the uncross-linked composite. Weight of the cross-linked kenaf/SPI-PVA composite decreased by approximately 12% after 5 days in a compost condition and 16% after 20 days. This indicated that the durability of the kenaf/SPI-PVA composite in a compost condition was enhanced by cross-linking with GA.

Photographs of the specimen under biodegradation conditions are shown in Figure 19. In the biodegradation test, moisture and temperature affected the biodegradation of the specimen. The kenaf/SPI-PVA composite without a cross-linking agent (a) was decomposed more in the compost than the cross-linked composite. In the kenaf/SPI-PVA composite with a cross-linking agent (b), however, biodegradability depended on the addition of GA rather than PVA [6, 20]. Considering biodegradation behavior, it can be concluded that the cross-linking of the kenaf/SPI-PVA composite with GA might govern the biodegradability. According to the biodegradation, by-products of microorganism appeared on the specimen's surface.

4. Conclusions

In this study, the kenaf/SPI-PVA composites were prepared with plasticizers and a cross-linking agent. Their interfacial adhesion properties, water absorption, biodegradation, and mechanical properties were analyzed. Results were as follows:

- (i) Increase of tensile strength in the kenaf/SPI-PVA composites indicated a good adhesion between kenaf fiber and SPI/PVA when GA of 8 wt% was added to the kenaf/SPI-PVA composites.
- (ii) Through the use of the cross-linking agent, the water absorption of the kenaf/SPI-PVA composite decreased. When GA 16 wt% was added, water absorption of the kenaf/SPI-PVA composite decreased significantly.
- (iii) The optimum preparation condition for the kenaf/SPI-PVA composite was established at PVA 15 wt% as a plasticizer and GA 8 wt% as a cross-linking agent.
- (iv) In the biodegradation test, degradation was controlled by the cross-linking agent GA. The degradation rate of the kenaf/SPI-PVA composite with GA was lower than the composite without GA, because the cross-linking between the kenaf fiber and SPI/PVA in the composite restricted its biodegradation.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

FIGURE 17: Surfaces (left, ×500) and fracture surfaces (right, ×100) of the kenaf/SPI-PVA composites with GA content: (a) 0 wt%, (b) 8 wt%, (c) 16 wt%, and (d) 25 wt%.

FIGURE 18: Weight loss (%) of (a) kenaf/SPI-PVA composite without GA and (b) kenaf/SPI-PVA composite having 8 wt% of GA with compost time.

FIGURE 19: Photographs of the kenaf/SPI-PVA composite with compost time: (a) kenaf/SPI-PVA composites without GA, (b) kenaf/SPI-PVA composites with GA.

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