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The poly(butylene succinate) (PBS) biodegradable composites reinforced with jute fibers were developed. The effect of fiber content (10–60wt%) on the properties and water absorption of jute/PBS biodegradable composites was studied. The effect of alkali, silane, and combined alkali and silane surface treatment on the properties and water absorption of jute/PBS composites was investigated. The mechanical properties of surface treated jute/PBS composites were significantly higher than those of untreated ones. Compared with alkali or silane treatment, the combined alkali and silane surface treatment showed better mechanical properties of jute/PBS composites. The best mechanical properties of jute/PBS composites were achieved at 50wt% in this study, which showed an increase in tensile strength by 517.9%, tensile modulus by 3529.8%, flexural strength by 302.6%, and flexural modulus by 1949.1% compared with those of PBS resin. Fractured surface morphologies of composite specimens exhibited an improvement of interfacial fiber–matrix adhesion in the composites reinforced with surface-treated jute fibers. The surface-treated jute/PBS composites having good fiber–matrix adhesion resulted in stable composites with better thermal stability than untreated jute/PBS composites. The water absorption amount of the composites increased with increasing the fiber content. The surface-treated jute/PBS composites showed relatively lower water absorption behavior compared to untreated ones.

Keywords: natural fiber biodegradable composites; mechanical properties; thermal properties; surface treatments; water absorption

1. Introduction

Biodegradable and environment-friendly green composite produced by combination of biodegradable polymers and natural fibers have attracted great interests in recent years due to their environmentally beneficial properties and potential applications in biomedical and bioengineering fields [1]. Natural fiber biodegradable composites have some major advantages over conventional composites such as, eco-friendliness, low cost, lightweight, high specific mechanical properties, and biodegradability [2–5]. Among the biodegradable polymers, poly (butylene succinate) (PBS) is increasing commercial interest. PBS is thermoplastic aliphatic polyester which can be naturally degraded into the environment by bacteria and fungi [6]. PBS has excellent biodegradability in nature, such as in soil, lake, sea, and compost [7]. It

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can be completely combustible by fire without evolving toxic gasses [8]. It has comparable mechanical properties with several thermoplastics like polyethylene, polypropylene, and polystyrene. As a result, PBS can be a good candidate material for the matrix of biodegradable composites.

The natural fibers, such as, hemp, jute, kenaf, flax, coir, ramie, sisal, and bamboo etc., offer specific benefits such as low cost, low density, low pollutant emissions, acceptable specific properties, renewability, and biodegradability [2,3]. Among the natural fibers, plant fibers are derived from lignocelluloses which contain strongly polarized hydroxyl groups. These fibers are hydrophilic in nature, thus they are inherently incompatible with hydrophobic thermoplastics [9]. In addition, moisture absorption of the fibers is very high because of the presence of pendant hydroxyl and polar groups in various constituents of fibers. This also leads to poor interfacial bonding with the hydrophobic matrix polymers. As a result, it is necessary to decrease the moisture absorption and hydrophilic character of fibers by suitable surface modification. Among the plant fibers, jute is the second most important bast fiber after cotton because of its easy availability at low cost. It is produced from plants in the genus *Corchorus*, family *Tiliaceae*. Jute fibers are composed primarily of the plant materials cellulose and lignin. Table 1 summarizes several physical, chemical, and mechanical properties of jute fiber compared with other typical natural fibers such as flax, hemp, coir, ramie, and sisal [1–3,10].

Jute has many advantages as a home textile, because it is the strong, durable, color, biodegradable, and renewable natural fiber. Its ultraviolet protection, sound and heat insulation, low thermal conduction, and anti-static properties make it a wise choice in home furnishings and in high performance technical textiles. Traditionally, jute is used for making hessian clothes, ropes, gunny and shopping bags, wrappers, wall-coverings, upholstery, floor mats, and, etc. The major breakthrough came when the automobile, the pulp and paper, and the furniture and bedding industries started to use jute and its allied fibers with their nonwoven and composite technology to manufacture nonwovens, technical textiles, and composites. However, a major drawback of using jute fiber as reinforcing material is its hydrophilic nature and responsible for moisture absorption. To be an effective reinforcing constituent for jute fiber, it is essential to improve compatibility and bonding between the fiber and the matrix. To make them suitable reinforcing candidates with adequate bond characteristics for general applications, various chemical modifications of jute fibers have been attempted [11–13]. Several researchers have carried out chemical treatment of jute fiber to improve its hydrophilic character and mechanical properties of jute fiber reinforced polymer composites [14–19]. Among the chemical surface treatments undertaken, alkali and silane treatment (AST) is the most economically viable. In the present work, the unidirectional jute/PBS biodegradable composites were developed. The jute/PBS biodegradable composites were fabricated by compression molding method. The effect of fiber content on mechanical and thermal properties and water absorption of jute/PBS composites was studied. The influence of alkali, silane, and combined alkali and silane treatment (ST) on the properties and water absorption of jute/PBS composites was also investigated. The fiber surface morphologies and fractured surfaces of composite specimens were investigated by scanning electron microscope (SEM) providing the information to evaluate the interfacial fiber–matrix adhesion.

2. Experimental

2.1. Materials

The raw jutes were supplied by Hung Yen Jute and Garment JSC (Hung Yen, Viet Nam). Biodegradable PBS pellets (#1001) with melting temperature of 115 °C were supplied by Showa High Polymers, Ltd. (Tokyo, Japan). The density of jute fiber and PBS are 1.4 g/cm^3

Table 1. Physical, chemical, and mechanical properties of jute fiber compared with other natural fibers [1–3,10]

Properties/fibers	Jute	Coir	Flax	Hemp	Ramie	Sisal
Density (g/cm^3)	1.3–1.45	1.25–1.5	1.4	1.48	1.5	1.26–1.33
Diameter (μm)	60	100–450	100	25	40–50	100–300
Cellulose content (%)	59–71	36–43	62–72	67–75	68–76	74–75.2$^{(B)}$60–67$^{(I)}$
Hemicellulose content (%)	12–13	0.2	16–18	16–18	13–14	10–13.9
Lignin content (%)	11.8–12.9	41–45	2–2.5	2.8–3.3	0.6–0.7	8–12$^{(I)}$7.6–7.98$^{(B)}$
Microfibrillar angle (°)	7–9	30–45	10	6.2	7.5–12	10–20
Tensile strength (MPa)	400–800	105–175$^{(I)}$ 95–118$^{(B)}$	800–1500	550–900	500–870	600–700
Young's modulus (GPa)	10–30	4–6$^{(I)}$	60–80	70	44	38
Elongation at break (%)	1.16–1.8	17–47$^{(I)}$ 23.9–51.4$^{(B)}$	1.2–2.4	1.6	1.2	3.64–5.12$^{(I)}$ 2–2.5$^{(B)}$
Moisture absorption (%)	12	10	7	8	12–17	11

Note: (B) – Brazilian and (I) – Indian.

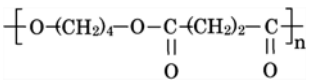

Figure 1. Chemical structure of PBS used in the present study.

and 1.26 g/cm^3, respectively. Figure 1 depicts the chemical structure of PBS used in this study.

2.2. *Surface treatment of jute fiber*

There different methods were used to treat the jute fibers before composite fabrication, including (1) alkali treatment (AT), (2) ST, and (3) AST. For AT method, firstly untreated (UT) jute fibers were treated with 2% sodium hydroxide solution in a glass beaker for different soaking times (1–6 h) at room temperature (RT). Next, the fibers were taken out of the solution and then washed several times with fresh water and subsequently with distilled water to remove any traces of sodium hydroxide sticking to the fiber surface until pH of the solution was equal to 7. Finally, the jute fibers were air-dried for two days. The reaction of sodium hydroxide with jute fiber is described as follows:

$$\text{Cell–OH} + \text{NaOH} \rightarrow \text{Cell–O} - \text{Na}^+ + \text{H}_2\text{O} \tag{1}$$

For ST method, the UT jutes were treated with γ-Methacryloxypropyltrimethoxysinale (γ-MPS) which was supplied by Shin-Etsu Chemical Co., Ltd (Tokyo, Japan). First, the aqueous solution was prepared by dissolving liquid 0.3wt.% of γ-MPS in acidified water (the pH of the solution was adjusted to 4.5 with acetic acid). Next, the solution was stirred continuously during 10 min, then UT jute fibers were immersed in the solution for 1 h at RT. Lastly, the fibers were air-dried for two days. The mean mechanical properties of PBS composites reinforced with 2% alkali-treated jute fibers for 3 h, which will be shown in next section, are higher than those of UT and other alkali-treated jute fibers.

Therefore, for AST method, firstly UT jutes were alkali-treated for 3 h and then soaked in a solution of 0.3wt.% γ-MPS at RT for 1 h. The reaction between jute fiber and silane coupling agent [11] is given as follows:

$$\begin{aligned} &\text{CH}_2 = \text{C(CH}_3\text{)COO(CH}_2\text{)}_3\text{Si(OH)}_3 + \text{Cell–OH} \rightarrow \\ &\text{CH}_2 = \text{C(CH}_3\text{)COO(CH}_2\text{)}_3\text{Si(OH)}_2\text{O–Cell} + \text{H}_2\text{O} \end{aligned} \tag{2}$$

2.3. *Composite fabrication*

The composite plates made from PBS and different UT, AT, ST, and AST jute fiber weight contents were fabricated by hot press equipment (Imoto Corp., Kyoto, Japan). To begin with, jute fibers were dried at 60 °C in a vacuum oven for 24 h. Next, the dried jutes were cut into the segments with the length of 150 mm, weighed, aligned in a parallel array, and placed in the mold between the PBS sheets as described in [20]. Then composite plates were pressed in a stainless steel mold with a thickness of 2 mm under 12 MPa pressure for 10 min at 150 °C and quickly cooled by fan. The schematic representation of the hot press for composite fabrication is shown in Figure 2. Both PBS sheets and composite plates were prepared with the same thermal history.

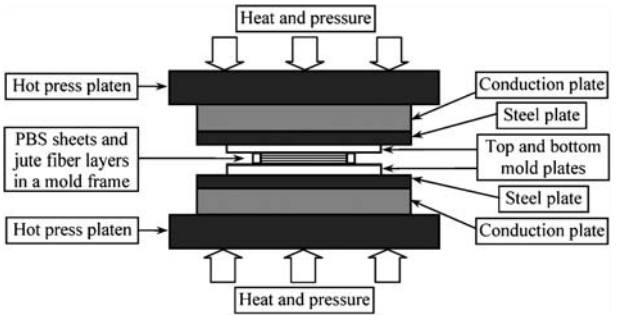

Figure 2. Schematic representation of the hot press used to fabricate composite plates [20].

2.4. Tensile test

The tensile specimens of 120 mm × 10 mm × 2 mm were cut out from pure PBS and composite plate by cutting machine AC-300CF (MARUTO Testing Machine Co., Tokyo, Japan) and kept in desiccator at 25 °C and 35% relative humidity before testing. The both clamped ends of the specimens (Figure 3) were glued by two glass fiber reinforced plastic tabs. Strain gages were glued at the center of the specimens to measure the fracture strain. Tensile properties were measured according to JIS K7113 using a universal testing machine RTF-1350 (JTT Inc., Tokyo, Japan). All the tensile tests were carried out at RT with a crosshead speed of 0.5 mm/min. The mean tensile properties of pure PBS and different surface-treated jute/PBS composites were obtained from seven specimens for each fiber content.

2.5. Flexural test

The flexural properties of pure PBS and jute/PBS biodegradable composites were measured by a three-point bending method according to JIS K7171 standard using universal testing machine Senstar SC-5H (JTT Inc., Tokyo, Japan). The flexural test was carried out at RT with a crosshead speed of 2 mm/min. The dimension of flexural specimens was 50 mm × 25 mm × 2 mm. The ratio of span width to thickness of pure PBS and composite specimens was 16. The flexural strength (σ_f) and modulus (E_f) were calculated using the following equations:

$$\sigma_f = \frac{3FL}{2bh^2} \tag{3}$$

$$E_f = \frac{L^3 m}{4bh^3} \tag{4}$$

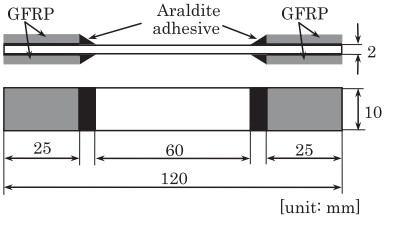

Figure 3. Shape and dimensions of tensile specimen.

where F is the maximal applied force, L is the length of support span, m is the slope of the force-deflection curve, b and h are the width and thickness of the specimen, respectively. The mean flexural properties of each composite were obtained from seven test specimens.

2.6. Thermogravimetric analysis

The thermal stability of jute fiber, PBS resin, and each jute/PBS biodegradable composite was analyzed up to 500 °C under a purging nitrogen gas with a flow rate of 100 cc/min by a thermogravimetric analyzer DTG-60 (Shimadzu Corp., Japan). About 10 mg of each specimen was loaded for each measurement at a heating rate of 10 °C/min. The thermogravimetric analysis (TGA) and differential thermal analysis curves were recorded.

2.7. Water absorption

In order to measure water absorption, the rectangular specimens having dimensions of 25 mm × 10 mm × 2 mm were prepared. The specimens were dried in an oven at 80 °C for 24 h, cooled in a desiccator to a constant weight, and immediately weighed. The water absorption tests were carried out by immersing the specimens in distilled water at RT from 1 to 256 h. At regular time intervals, each specimen then was removed from the water tank and wiped with tissue paper to remove surface water and subsequently weighted by electronic balance (ER-182A, A&D Instruments, Tokyo, Japan). The water absorption was calculated and reported as the percentage increase of the initial weight as follows:

$$\text{Water absorption } (\%) = \frac{W_t - W_o}{W_o} \times 100 \tag{5}$$

where W_t and W_o are the weights of the specimen before and after immersion in distilled water, respectively. All data from five repeated tests were averaged.

2.8. Morphological characterization

The jute surface morphologies and fractured surface of the composites after tensile tests were examined using SEM (VE-7800, Keyence Inc., Osaka, Japan).

3. Results and discussions

3.1. Surface modification of jute fiber

Figure 4 shows SEM micrographs of jute fiber surface before and after chemical surface treatments. It is observed that jute fibers after treatment have some changes in their surface structure. Natural and artificial impurities were found on the UT jute fiber surface (Figure 4(a)). However, these impurities on jute fiber surface were removed after AT making the fiber cleaner and rougher than before (Figure 4(b)). Moreover, a rougher surface morphology was typical for the treated jute fibers, because of the removal of lignin and hemicelluloses in comparison with UT fibers [13]. It can be realized that AT increases the surface roughness and exposes the cellulose on the fiber surface. Furthermore, as reported in [15] surface modification by alkalization can also change the fine structure of the native cellulose I to cellulose II and the crystallographic structure, which is likely to result in stiff and strong fibers of interest in the formation of biocomposites. The hydrophilic behavior of jute fiber induced by predominance of –OH groups will thus be weakened, and its compatibility with hydrophobic PBS will accordingly be improved [11,21].

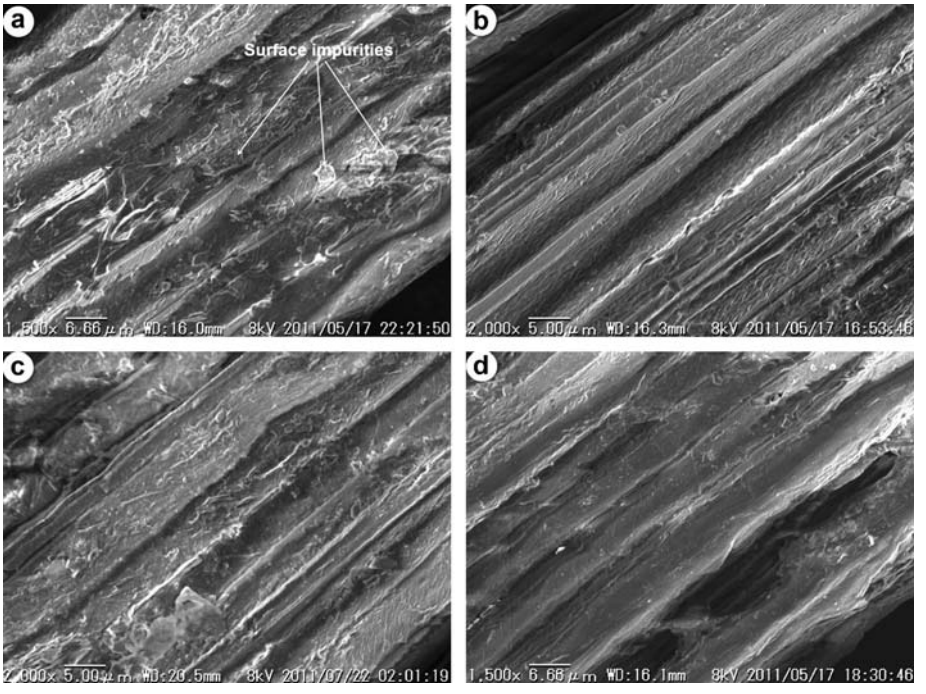

Figure 4. SEM micrographs of jute fiber surface: (a) UT jute, (b) AT jute, (c) ST jute, and (d) AST jute.

For ST, jute fibers were treated with hydrolyzed silane solution to allow silane penetrates into the fiber lumina and further diffuse into the cell walls. As a result, after ST jute fiber surfaces were modified, as seen in Figure 4(c) and (d). This can be explained that when hydrolyzed silane solutions are mixed with natural fibers, the reactive silanol groups have a high affinity for each other, forming –Si–O–Si– bonds, and also for the hydroxyl sites of fibers via hydrogen bonds [22,23]. The silanols of γ-MPS firstly form a monolayer on the fiber, and then are further adsorbed resulting in the formation of a rigid polysiloxane layer on the fiber surface [24]. In general, the main defects of natural fibers as reinforcing fillers, such as high moisture absorption, poor wettability, and incompatibility with hydrophobic polymer, can be improved by surface modification. This demonstrates that surface modification of natural fibers can improve the properties of their composites.

3.2. Effect of fiber content on mechanical properties of UT jute/PBS composites

Mechanical properties of PBS resin and UT jute/PBS composites with different fiber weight contents were given in Table 2. As seen in Table 2, the incorporation of jute fibers improved mechanical properties of PBS resin. It can be realized that tensile and flexural strength gradually increased with increasing the fiber content from 0 to 50wt.%, but decreased with upper fiber content. Tensile modulus of the composites gradually increased with increasing the fiber content up to 60wt.% while flexural modulus increased with increasing the fiber content to 50wt.%, but it decreased at 60wt.% fiber content. The increase in mechanical strength and modulus of the composites is due to the reinforcement of jute fibers in PBS matrix, because the strength and modulus of jute fiber are higher than those of PBS matrix. The decrease in mechanical strength and flexural modulus at 60wt.% fiber content probably resulted from

Table 2. Mechanical properties of PBS and UT jute/PBS biodegradable composites.

Fiber content (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Fracture strain (%)	Flexural strength (MPa)	Flexural modulus (GPa)
0	37.5 ± 1.1	1.0 ± 0.06	9.0 ± 0.91	49.4 ± 2.4	0.9 ± 0.2
10	66.6 ± 5.3	10.2 ± 1.6	0.97 ± 0.07	70.3 ± 4.8	2.9 ± 0.5
20	98.0 ± 6.5	13.2 ± 2.0	0.86 ± 0.05	115.9 ± 6.9	7.4 ± 0.6
30	131.1 ± 10.5	20.5 ± 2.2	0.85 ± 0.06	137.2 ± 8.9	9.0 ± 0.7
40	143.3 ± 11.3	23.2 ± 2.3	0.79 ± 0.07	161.1 ± 8.7	11.6 ± 0.9
50	157.7 ± 10.5	27.0 ± 2.4	0.75 ± 0.08	185.1 ± 8.6	14.4 ± 0.8
60	147.5 ± 9.90	30.8 ± 2.8	0.63 ± 0.08	150.1 ± 9.5	12.7 ± 0.8

incomplete fiber wetting, because PBS content is not sufficient to wet all fiber surfaces. The high mechanical strength at 50wt.% fiber content might be due to adequate fiber content in the composites, which leads to greater wetting. Compared with PBS, UT jute/PBS composites at 50wt.% fiber content shows an increase in tensile strength by 320%, tensile modulus by 2594%, flexural strength by 274.7%, and flexural modulus by 1577.1%. The best mechanical properties of UT jute/PBS biodegradable composite were obtained at the fiber content of 50wt.% in this study.

The incorporation of high fiber content reduced fracture strain of the composites, because increasing the amount of reinforced fibers will lead to the decrease in the amount of polymeric matrix available for the elongation. The decrease in fracture strain, as seen in Table 2, is mainly due to the structural integrity of PBS being destroyed by jute fiber loading and increasing fiber content imply poor interfacial fiber–matrix bonding leading to quicker fracture than pure PBS [15]. Compared with PBS, fracture strain of UT jute/PBS composites with 10wt.% fiber content significantly reduces to 89%. After such initial drop, the percent fracture strain decreases inconsiderably with increasing the fiber content. This can be explained due to the lower fracture strain of jute fiber than that of PBS resin. This also indicates that the ductile nature of PBS resin strongly decreases with the addition of jute fibers.

3.3. Effect of soaking time on mechanical properties of alkali-treated jute/PBS composites

The effect of soaking time on tensile and flexural properties of alkali-treated jute/PBS composites at 30wt.% fiber content is shown in Figures 5 and 6. As seen in Figure 5, the mean tensile strength and modulus of the composites increase with increasing the soaking time from 1 to 3 h, but they decrease beyond 3 h. When the soaking time increases from 1 to 6 h, the tensile strength and modulus of alkali-treated jute/PBS composites range from 147.4 to 151.9 MPa and 21.6 to 22.9 GPa, respectively. The standard of deviation ranges from 9.4 to 12.4 for tensile strength and from 1.7 to 3.1 for tensile modulus. Consequently, statistically within the error bars the tensile strength and modulus change inconsiderably. The trend was similar for the flexural strength and modulus of the composites (Figure 6). The mechanical strength and moduli of composite reinforced with 3 h alkali-treated fiber were found to be superior in comparison with the UT and other alkali-treated jute fibers (as shown in Table 2 and Figures 5 and 6). This can be explained that when increasing the soaking time AT will makes the fiber rigid and somewhat brittle afterward owing to the development of crystallinity. On the application of stress, these fibers suffered breakage due to increased brittleness and could not take part in effective stress transfer at the interface, thus lowering the strength and modulus of the composite as reported in [19]. Concurrently, Ray and Sakar [12] showed that most of the changes of jute fibers occurred within 2–4 h of AT. In this study, maximum

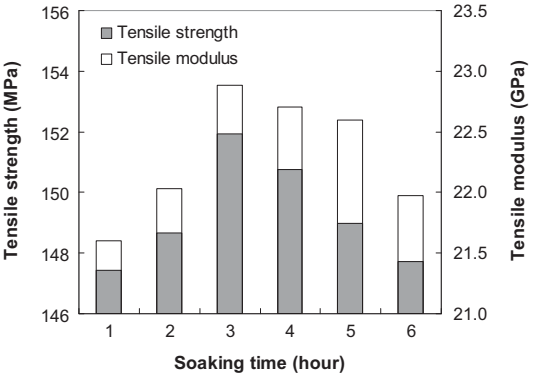

Figure 5. Effect of soaking time on tensile properties of 30wt.% alkali-treated jute/PBS composites.

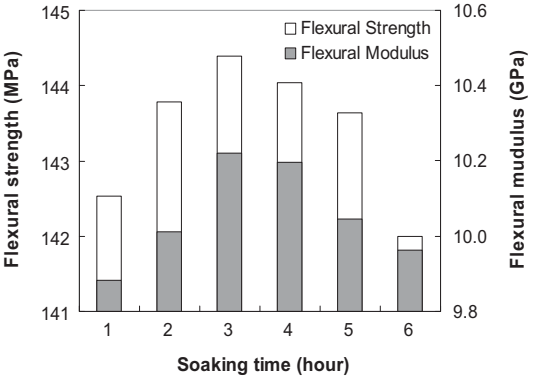

Figure 6. Effect of soaking time on flexural properties of 30wt.% alkali-treated jute/PBS composites.

improvement in mechanical properties was observed for the composite prepared with 3 h alkali-treated jute fiber.

3.4. Effect of fiber surface treatment and fiber content on mechanical properties of jute/PBS composites

AT improves the fiber–matrix adhesion due to the removal of natural and artificial impurities from the fiber surface as well as changing in the arrangement of units in the cellulose macromolecule [25]. As described above, AT increases the surface roughness and the amount of cellulose exposed on the fiber surface resulting in better mechanical interlocking. This was well depicted in Figure 4 by comparing the SEM micrograph of alkali-treated fiber with the UT fiber. Therefore, the development of a rough surface tomography and the enhancement in aspect ratio offer better fiber–matrix interfacial bond resulting in increasing mechanical properties. Silane coupling agent is a chemical which functions at the interface to create a chemical bridge between the reinforcement and the matrix. It improves the interfacial adhesion when one end of the molecule is tethered to the reinforcement surface and the functionality at the other end reacts with the polymer phase [24].

The effect of fiber surface treatment and fiber content on the mechanical properties of jute/PBS biodegradable composites is shown in Figures 7 and 8. The tensile strength,

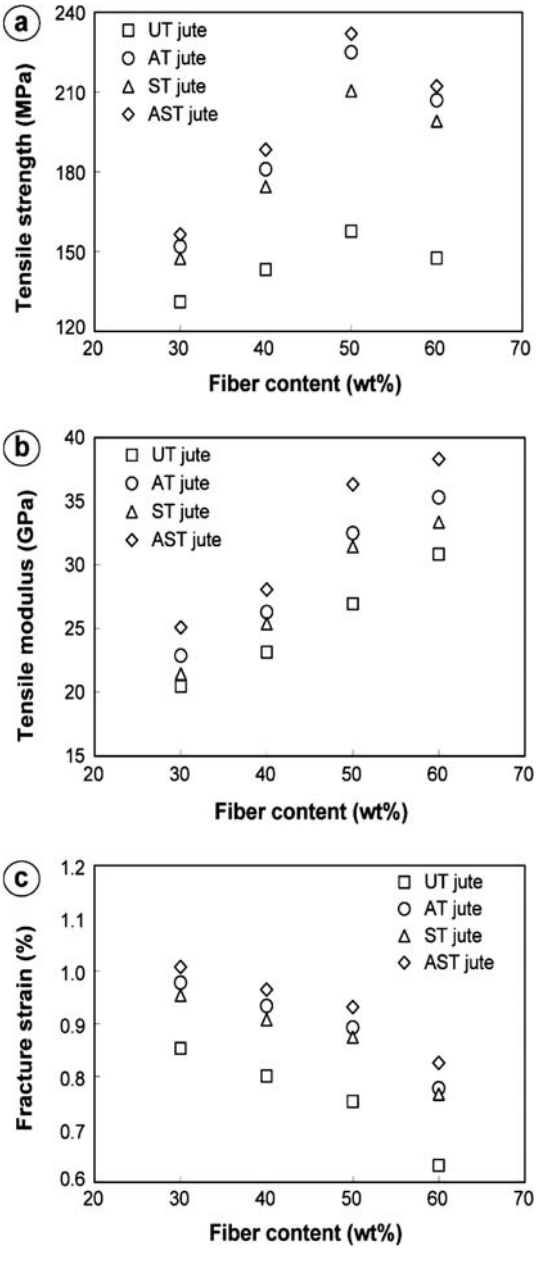

Figure 7. Effect of fiber surface treatment on: (a) tensile strength, (b) tensile modulus, and (c) fracture strain of jute/PBS biodegradable composites with different fiber weight contents.

modulus, and fracture strain of jute/PBS composites at 50wt.% fiber content range from 157.7 to 231.9 MPa, 26.9 to 36.3 GPa and 0.8 to 0.93%, respectively. The coefficient of variation ranges from 4.0 to 6.7% for tensile strength, from 7.1 to 11.6% for tensile modulus, and from 6.9 to 11.0% for fracture strain. As seen in Figure 7, the surface modifications of jute fibers by AT, ST, and AST improved tensile properties of jute/PBS composites. Compared with UT jute, AT, ST, and AST jute/PBS composites at 50wt.% fiber content exhibited an

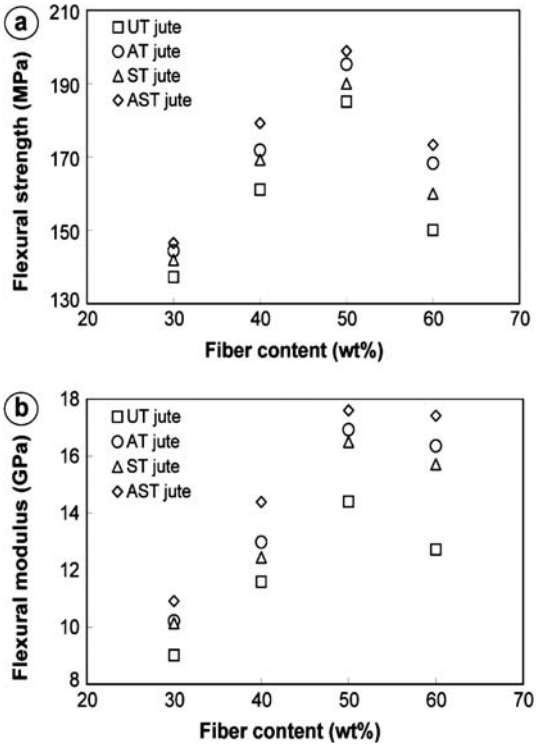

Figure 8. Effect of fiber surface treatment on: (a) flexural strength and (b) flexural modulus of jute/PBS biodegradable composites with different fiber weight contents.

increase in tensile strength by 42.6, 33.5, and 47.0%, in tensile modulus by 20.6, 16.7, and 34.8% and in fracture strain by 18.6, 16.2, and 23.7%, respectively. Moreover, as demonstrated in Figure 8, the surface treatment of jute fibers improved also the flexural properties of jute fiber reinforced PBS biodegradable composites. AT, ST, and AST jute/PBS composites yielded higher mean value of flexural properties when compared to UT jute/PBS composites. This reflects the contribution of alkali or silane in terms of changes of fiber properties and the enhancement of interfacial fiber–matrix adhesion. The flexural strength and modulus of jute/PBS composites at 50wt.% fiber content range from 185.1 to 198.9 MPa and 14.4 to 17.6 GPa, respectively. The coefficient of variation ranges from 3.7 to 4.6% for flexural strength and from 5.2 to 5.8% for flexural modulus. AT, ST, and AST jute/PBS composites at 50wt.% fiber content showed an increase in flexural strength by 5.5, 2.7, and 7.5% and flexural modulus by 17.5, 14.5, and 22.2% compared to the one with UT jute fibers, respectively. The results indicate that surface modification by AT, ST, and AST has less influence on flexural properties compared to tensile properties. This can be explained that the flexural failure mode usually shows little fiber pull-out [15], because applied force is perpendicular to reinforced fibers of the composite specimens in flexural test. It is interesting to note that flexural strength has the same trend as tensile strength with increasing the fiber content.

Compared with AT or ST, AST method showed better mechanical properties of the jute/PBS composites. The increase in mechanical properties of surface-treated jute/PBS composites may be due to greater fiber–matrix interfacial and physical bonding. As described above, the surface treatment can improve the compatibility between jute fiber and PBS matrix leading to less interfacial fiber–matrix debonding. As shown in Figure 9(a), the pulled-out fibers

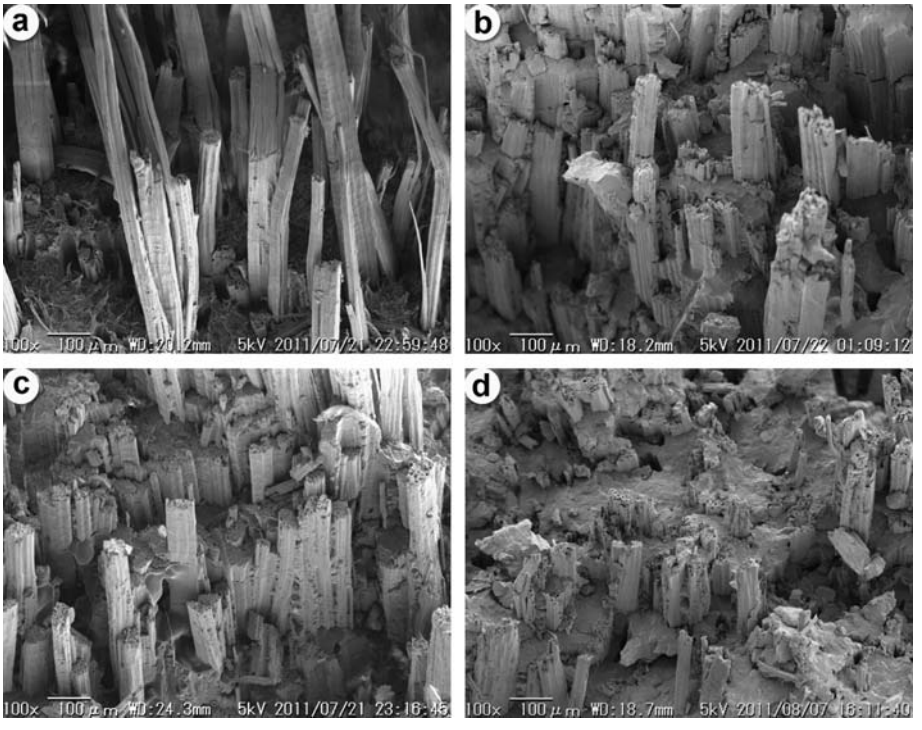

Figure 9. SEM micrograph of fractured surface of PBS biodegradable composite reinforced with 30wt. % fiber content of: (a) UT jute, (b) AT jute, (c) ST jute, and (d) AST jute.

can be found on the fracture surface of UT jute/PBS, suggesting poor interfacial fiber–matrix adhesion. For the AT (Figure 9(b)) and ST jute/PBS composites (Figure 9(c)), several jute fibers were pulled-out and broken to some extent during the fracture process, and a little PBS matrix remained and adhered to the surface of the jute fibers. However, lots of pulled-out fibers disappeared in the case of AST jute (Figure 9(d)), proving good compatibility being formed in PBS composites. For AST jute/PBS composites, it was also noted that the jute fibers failed by tearing, but that no complete interfacial failure occurred, a result which indicated that the adhesion between AST jute fibers and PBS matrix was strong enough. Therefore, AST of the jute fiber improved its surface properties and strengthened the fiber–matrix interaction.

It is obvious that UT jute fiber can be easily pulled-out from the interfacial region with poor compatibility resulting in rapid partial-collapse of PBS composite. Nevertheless, the surface-treated jute fiber having a good adhesion with PBS matrix can effectively disperse and transfer stress, leading to the improvement in mechanical properties of jute/PBS biodegradable composites. Consequently, the results suggest that surface treatment of jute fiber is necessary to enhance the interfacial fiber–matrix adhesion prior to composite processing. The mechanical strength and modulus of unidirectional jute/PBS biodegradable composites showed an optimum fiber content. The optimum fiber content varies with the nature of both fiber and matrix, fiber aspect ratio, fiber–matrix interfacial adhesion, fiber agglomeration, processing technique, etc [15]. Similar investigations have also been reported by Mohanty et al. [26] for jute fabrics/polyester composites in which the optimum fiber content is 32wt.% and by Roe and Ansell [27] for jute/polyester composites is about 60 vol.%. In this study, the addition of 50wt.% fiber content showed the best mechanical properties of jute/PBS

composites. In short, the results of mechanical properties point out the importance by using the right amount of fiber as reinforcement in the composites.

3.5. TGA analysis

TGA is a useful method that based on the measurement of weight change related to temperature for the quantitative determination of the degradation behavior and the composition of the fiber and the matrix in a composite. The magnitude and location of peaks found in the derivative thermogravimetric (DTG) curve also provide information on the component and the mutual effect of the composite components on the temperature scale [28]. The thermogravimetric (TG) and the DTG curves of UT jute, PBS and UT jute/PBS composites are shown in Figure 10. The peak temperatures and the weight loss percentage of UT jutes, PBS resin, and UT jute/PBS composites obtained from DTG curves are given in Table 3. The peak temperatures and the percentage of weight losses of different surface-treated jute/PBS composites with different fiber weight contents are shown in Table 4.

As observed in Figure 10, UT jute fiber degrades through three main stages. The first stage, from 30 to about 250 °C, is attributed to the release of absorbed moisture in the jute fiber. About 5% weight loss of UT jute was observed in first stage because of absorbed moisture. The initial weight loss around 60 °C can be due to the evaporation of water in the jute fiber; therefore, it is not significant weight loss in the range of 100–250 °C approximately

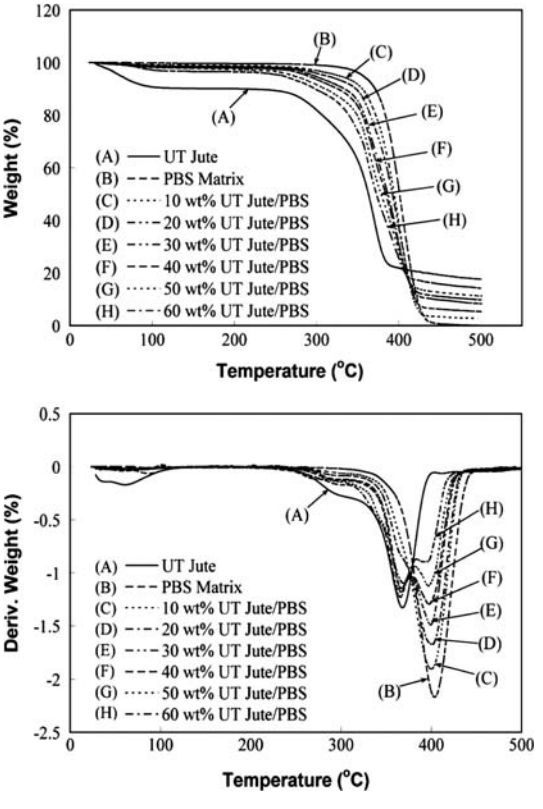

Figure 10. TG (top) and DTG (bottom) thermograms showing the thermal properties of UT jute fiber, PBS and UT jute/PBS biodegradable composites.

Table 3. Peak temperature and percent weight loss of PBS and UT jute/PBS composites.

Fiber content (wt.%)	1st peak temperature (°C)	% of weight loss at 1st peak	2nd peak temperature (°C)	% weight loss at 2nd peak	% weight loss at 500 °C
0			403.6	55.7	99.8
10			400.3	60.6	97.3
20			400.0	63.2	94.7
30	367.4	28.2	399.3	65.5	91.7
40	367.0	31.9	398.4	67.3	90.2
50	367.0	37.6	398.1	68.9	88.8
60	365.4	41.5	393.9	69.7	85.7

Table 4. Peak temperature and percent weight loss of jute/PBS composites with different surface treatments.

Fiber content (%)	1st peak temperature (°C)			% of weight loss at 1st peak			2nd peak temperature (°C)			% of weight loss at 2nd peak		
	AT	ST	AST	AT	ST	AST	AT	ST	AST	AT	ST	AST
30	369.5	364.2	367.9	27.8	25.8	24.0	402.4	400.4	401.8	65.3	64.7	64.0
40	368.3	364.1	367.2	32.8	28.8	27.4	399.3	399.3	401.4	66.7	65.7	65.9
50	367.1	362.8	365.7	35.4	31.6	33.0	396.7	395.0	398.4	66.5	64.8	67.1
60	367.0	363.9	364.8	39.7	36.7	36.8	389.6	390.9	394.6	64.2	64.3	66.9

similar with the decomposition behavior of other natural fibers, such as kenaf [29], coir [30], silk [31], and wood [32]. The second transition occurs from 250 to 370 °C, jute fiber undergoes thermal degradation of cellulose and hemicellulose substances. In this stage, the thermal degradation rate is slow below 280 °C, but above 280 °C the weight loss becomes fast. The main degradation peak occurs between 280 and 370 °C, where about 55% of the degradation occurs. The third stage occurs above 370 °C, and jute fiber starts to decompose with a lower rate of the weight loss. In this stage, all the volatile materials are driven off from the sample resulting in the residual char.

Figure 10 also showed that the thermal degradation of pure PBS starts near 300 °C and the fastest weight loss occurs with a peak of temperature at 404 °C. About 56% weight loss of PBS is observed at the peak of temperature, while 99.8% weight loss occurs at 500 °C (Table 3). The thermal stability of jute/PBS biodegradable composites is likely to be intermediate between the PBS matrix and the jute fiber depending on the fiber weight content. Below about 400 °C, the thermal degradation of the composites increases with increasing the fiber content, but it decreases above 400 °C. As a result, at low temperature region the thermal stability of the composites decreases with increasing the jute fiber content similar to the results reported in [30] for coir/PBS biodegradable composites. The percentage weight loss of jute/PBS composites decreases with increasing the fiber content at 500 °C (Table 3). As seen in Figure 10 and Table 3, jute/PBS composites have two peaks at high-fiber weight content in which the 1st peak corresponding to the degradation peak of jute fibers and the 2nd peak corresponding to degradation peak of PBS resin. In general, in the jute/PBS composites the less thermal stability of jute fiber in the low-temperature region can be compensated by the PBS, while the less thermal stability of the PBS in the high-temperature region can be also compensated by the presence of jute fibers.

As seen in Table 4, the surface treatment decreased a little percentage of weight loss and enhanced percentage residue resulting in increasing the thermal stability of the composites. In the case of AT, the PBS resin is well penetrated into the alkali-treated jute fiber thereby resulting in a strong fiber–matrix interface [33]. This resulted in the increase of thermal stability of the alkali-treated jute/PBS composites. For treatment with γ-MPS, the γ-MPS may act as a bridge at the interface between the treated jute fibers and the PBS resin. Since, γ-MPS is a bifunctional silane containing a methacrylate reactive organic group and a trimethoxysilyl inorganic group [16]. In the treated process with γ-MPS, first methoxyl groups are hydrolyzed to hydroxyl groups in distilled water. Then silanol groups react with hydroxyl groups that exist on the surface of jute fiber due to cellulose structure and form strong covalent bonds or H-bonds with OH group of cellulose [33]. Consequently, silane coupling agent increases the adhesion between ST jute fibers and PBS resin. Overall, surface-treated jute/PBS composites having good fiber–matrix adhesion resulted in stable composites with better thermal stability compared to UT jute/PBS composites.

3.6. Water absorption

Figure 11 showed the water absorption behavior of PBS and UT jute/PBS biodegradable composites as a function of fiber content. It was found that the water absorption amount of the composites increased with increasing the fiber content. This can be explained that with increasing fiber content the number of hydroxyl groups in the composites increased resulting in increasing the water absorption [21]. The increase of jute fiber content caused the absorption rate to quicken due to its strong hydrophilicity [34]. Furthermore, PBS resin can absorb only the water of about 1%, thus jute fiber content will be the major factor affecting the water absorption of the composites. The effect of surface treatments on the water absorption behavior of jute/PBS composites with different fiber contents was investigated and indicated in Figure 12. As seen in Figures 11 and 12, the water absorption for all composites increased greatly during the first 30 h, then slightly increased and leveled-off. The surface-treated jute/PBS composites show relatively lower water absorption behavior compared to UT ones. This can be explained due to the improvement in interfacial adhesion between the PBS and the jute fiber after chemical surface treatment. This indicates that hydrophilic nature of the

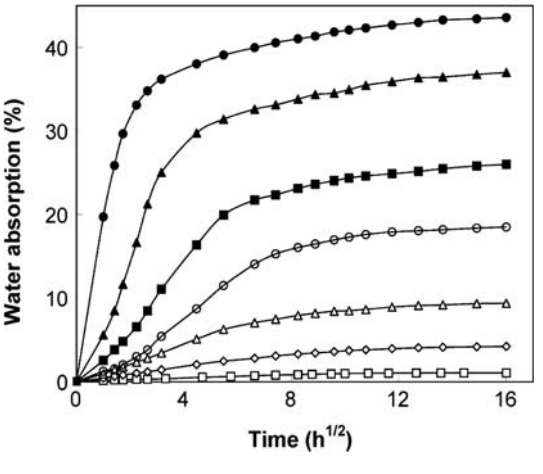

Figure 11. Effect of fiber content on water absorption of PBS and UT jute/PBS composites with different fiber contents: □ PBS; ◊ 10wt.%; △ 20wt.%; ○ 30 wt.%; ■ 40 wt.%; ▲ 50wt.%; ● 60wt.%.

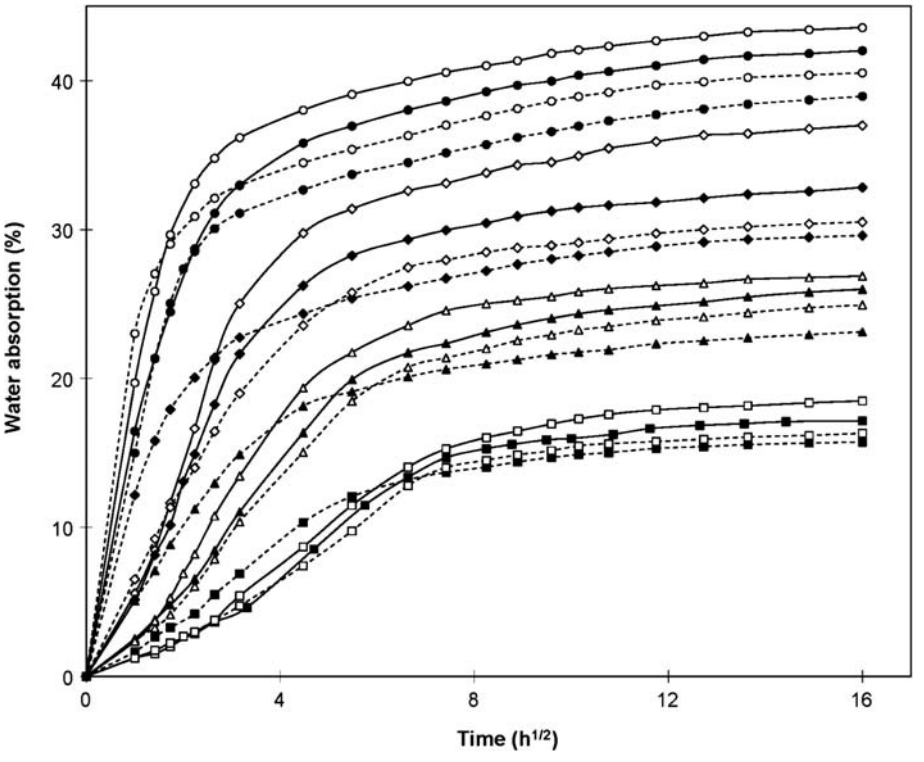

Figure 12. Effect of surface treatment on water absorption of jute/PBS composites with different fiber contents: □ ■, 30wt.%; △ ▲, 40wt.%; ◊ ◆, 50wt.%; ○ ●, 60wt.%; white for UT (solid line) and AT (dash line); and black for ST (solid line) and AST (dash line).

composites has substantially reduced during alkali and ST of jute fiber leading to decreasing the water absorption. Compared with AT or ST, AST method showed lowest water absorption of jute/PBS composites, because AST of the jute fiber improved its surface properties and strengthened better fiber–matrix interaction in the composites as explained above.

4. Conclusions

Biodegradable composites made from PBS resin and jute fibers have been developed. Effect of AT, ST, and AST on the properties and water absorption of jute/PBS biodegradable composites has been studied. The following conclusions can be drawn from this study:

(1) The mechanical properties of surface-treated jute/PBS composites are significantly higher than those of UT ones.
(2) AT increased the surface roughness and exposed the cellulose on jute surface leading to the increase of mechanical interlocking and interfacial bonding. In this study, the best mechanical properties of alkali-treated jute/PBS composites obtained when the jute fibers are soaked in 2% sodium hydroxide for 3 h.
(3) Surface modification by AT, ST, and AST increased mechanical properties of jute/PBS composites, in which AST method showed highest mechanical properties of jute/PBS composites.

(4) Mechanical strength of jute/PBS biodegradable composites increased with increasing fiber content up to 50wt.%, but decreased with upper fiber content. The authors propose that the 50wt.% jute fiber content reinforced PBS biodegradable composites have the best mechanical properties in this study.
(5) Thermal stability of jute/PBS composites is remarkably to be intermediate between PBS resin and jute fiber depending on the fiber content. The surface-treated jute/PBS composites having good fiber–matrix adhesion resulted in stable composites with better thermal stability compared to UT ones.
(6) The surface-treated jute/PBS biodegradable composites resulted in lower amount of water absorption compared to UT jute/PBS composites. The present results suggest that a useful composite with good properties could be successfully developed using jute as a reinforcing agent for the PBS matrix.

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