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Advanced Composite Materials

ISSN: 0924-3046 (Print) 1568-5519 (Online) Journal homepage: http://www.tandfonline.com/loi/tacm20

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To cite this article: Tran Huu Nam, Shinji Ogihara, Satoshi Kobayashi & Ken Goto (2015) Effects of surface treatment on mechanical and thermal properties of jute fabric-reinforced poly(butylene succinate) biodegradable composites, Advanced Composite Materials, 24:2, 161-178

To link to this article: http://dx.doi.org/10.1080/09243046.2014.882538

Advanced Composite Materials, 2015
Vol. 24, No. 2, 161–178, http://dx.doi.org/10.1080/09243046.2014.882538

Effects of surface treatment on mechanical and thermal properties of jute fabric-reinforced poly(butylene succinate) biodegradable composites

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(*Received 24 September 2013; accepted 26 December 2013*)

Biodegradable composites based on poly(butylene succinate) (PBS) and unidirectional plain jute fabrics have been developed. These composites were fabricated by compression molding of sandwiching 4–7 jute fabric layers between five and eight layers of PBS sheets. Surface modification of the jute fabric by alkali and combined alkali-silane treatments was investigated. The effects of surface modification on the mechanical and thermal properties of jute fabric/PBS biodegradable composites were studied. The mechanical properties of surface-treated jute fabric/PBS biodegradable composites were significantly higher than those of untreated ones. Compared with the alkali treatment, the combined alkali-silane treatment showed higher mechanical properties of the jute fabric/PBS biodegradable composites. The alkali-silane-treated jute fabric/PBS biodegradable laminated composite with six reinforced fabric layers (47.5 wt.%) achieved the best mechanical properties in this study, which showed an increase in tensile strength by 16.4%, tensile modulus by 10.8%, flexural strength by 24.2%, and flexural modulus by 21.9% compared with those of untreated one. Fractured surface morphologies of tensile specimens exhibited an improvement of interfacial fiber-matrix adhesion in the PBS biodegradable composites reinforced with surface-treated jute fabric. Thermal stability of the jute fabric/PBS biodegradable composites was remarkably to be intermediate between the PBS resin and the jute fabric. Surface-treated jute fabric/PBS biodegradable composites having good interfacial fiber-matrix adhesion resulted in stable composites with better thermal stability than that of untreated ones.

Keywords: natural fibers; biodegradable composites; surface treatments; mechanical properties; thermal properties

1. Introduction

Recently, natural fiber-reinforced polymer composites have been great interest both in terms of their industrial applications and fundamental research. They are renewable, low-cost, eco-friendly, completely or partially recyclable, and biodegradable.[1] Natural fiber composites based on petroleum-based thermoplastics or thermosets matrices have

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been used in many applications such as automobiles, railway coaches, aerospace, military, building and construction, packaging, consumer products, etc. [2,3]. However, a number of natural fiber composites are not fully environmentally friendly, because their matrix resins are not completely biodegradable. Therefore, the development of fully biodegradable composites [4–7] made of natural fibers and biodegradable polymers, such as poly(butylene succinate) (PBS),[8–11] has increased because of their potential environmental and economic benefits for wide range applications.

The advantages of natural fibers over traditional reinforcing fibers, such as glass and carbon fibers, are low cost, low density, high toughness, acceptable specific strength, good thermal properties, renewable characteristics, enhanced energy recovery, and complete biodegradability.[1] In general, natural fibers themselves usually have limited length by their natural origin, e.g. the plant stem has restricted length. As a result, natural fibers are often found as short reinforcements in the composites. Nevertheless, the short fiber composites have shown the moderate or low mechanical properties and can be used in non-structural applications.[12] Therefore, the alternative to the use of short natural fibers reinforcing in the composites is the production of long natural yarns.[13] Natural yarn is a long continuous assembly of relatively short interlocked natural filaments. The main advantage of using natural yarns is the possibility to weave them into fabrics with tailored yarn orientations.

These days, natural fabrics are widely used as reinforcements for composite manufacturing, because they allow the fabrication of stiff parts that are easily handed. Among commercial natural fabrics, jute fabric is extensively used in high-performance technical textiles and home furnishings. Jute fabric is economical and strong, reusable and repairable, and considered as a good insulator of electricity and heat. Because of these advantages, jute fabric is used for making a wide variety of fashion apparel, soft luggage, backing for carpets, inner spring webbing for car seats, bags, or sacks for packing, etc. Moreover, jute fabric is highly breathable and comfortable to use. Therefore, using jute fabric can easily produce structural composite materials for many industrial applications.[14]

Nevertheless, the major disadvantages of jute fabric include brittleness, coarseness, low extensibility, poor drapability and crease resistance, high fiber shedding, photo yellowing in sunlight, susceptibility to microbial attack in humid conditions, and especially hydrophilic nature and high moisture absorption.[15,16] To reduce the disadvantages of the jute fabric and enhance their reinforced efficiency in the composites, the chemical treatment methods were applied for the jute fabrics.[17–19] Among the chemical surface treatments undertaken for natural fibers, alkali and silane treatment are the most economically viable.[20–22] Surface modification by alkali and silane treatment would enhance interfacial adhesion as well as the compatibility between the reinforced fibers and polymer matrix, resulting in improving the composite properties.

In our previous research,[11] unidirectional jute fiber-reinforced PBS biodegradable composites were developed, but we might experience the difficulties to produce bulk composites for industrial/commercial applications. As a result, in this study, unidirectional plain jute fabric was used as reinforcement in PBS resin to develop novel biodegradable composites. The unidirectional jute fabric/PBS biodegradable laminated composites were produced by hot compression molding method. Surface modification of jute fabrics by alkali and silane treatment was carried out to improve the interfacial bonding with PBS matrix in the composite. The mechanical and thermal properties of jute fabric/PBS biodegradable composites were studied. The effect of alkali and silane treatment on the composite properties was investigated. The morphologies of jute

surfaces and composite-fractured surfaces were observed by scanning electron microscope (SEM) providing the information to evaluate the interfacial fiber-matrix adhesion.

2. Experimental set-up

2.1. Materials

The unidirectional plain jute fabric which was supplied by Hungyen Jute and Garment JSC (Hung Yen, Vietnam) made of the jute spun yarns in the warp direction and cotton thread in the weft direction (Figure 1). The areal density of the jute fabric is about 213 g/m^2. The biodegradable PBS pellets (#1001) with melting temperature of 115 °C were supplied by Showa High Polymers, Ltd. (Tokyo, Japan). The density of PBS resin is 1.26 g/cm^3.

2.2. Surface treatment of jute fabrics

Chemical treatments which were applied to modify the surface of untreated (UT) jute fabrics before composite fabrication include (i) alkali treatment (AT) and (ii) combined alkali-silane treatment (AST). For AT method, at first, the UT jute fabric was treated with 2% sodium hydroxide solution in a glass beaker for soaking time of 3 h at room temperature (RT). Next, the treated jute fabric was taken out of the solution. After that, the treated jute fabric was washed several times with fresh water and subsequently with distilled water until pH of the solution was equal to 7. Lastly, the AT jute fabric was air dried for two days.

For AST method, the AT jute fabric was treated with the silane-coupling agent by an aqueous solution of γ-Methacryloxypropyltrimethoxysinale (γ-MPS) which was supplied by Shin-Etsu Chemical Co., Ltd. (Tokyo, Japan) for 1 h at RT. To begin with, the aqueous solution was prepared by dissolving liquid 0.3 wt.% 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 for 10 min. Then, the AT jute fabrics were immersed in the solution for 1 h at RT. Finally, the AST jute fabrics were air dried for two days.

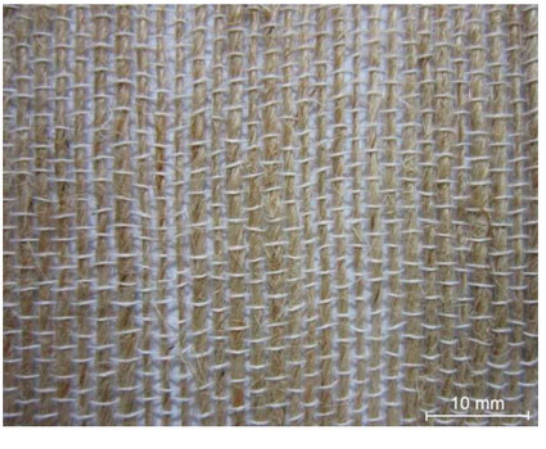

Figure 1. Structure of plain jute spun yarn fabric used in the present study.

2.3. Composite processing

The laminated composite plates made of the PBS sheets and different UT, AT, and AST jute fabric layers were fabricated by compression molding method. First, the PBS sheets of 0.1 (±0.01) mm thickness were prepared by compression molding at 150 °C in a hot press (Imoto Corp., Kyoto, Japan). Next, the jute fabrics were dried at 80 °C in a vacuum oven until obtaining constant weight. After that, the UT and different treated jute fabric/PBS biodegradable composites were prepared by sandwiching n pre-weighted fabric layers (4–7 for UT and 4–6 for AT and AST) between *m* layers of pre-weighted PBS sheets (5–9 for UT and 5–8 for AT and AST). Lastly, the prepared sandwich was pressed in a stainless steel mold with a thickness of 2 mm under 12 MPa pressure for 10 min at 150 °C and then quickly cooled by fan. The processing of the laminated composite plates based on the PBS sheets and jute fabric layers was shown in Figure 2. For all composites, the weight fraction of the jute fabric was calculated from the pre-weighted PBS sheets and the total weight of the composite.

2.4. Tensile test

The tensile specimens of 120 mm × 10 mm × 2 (+0.2/−0.05) mm were cut out from the composite plate using a cutting machine AC-300CF (MARUTO Testing Machine Co., Tokyo, Japan). After that, they were kept in desiccator at 25 °C and 35% relative humidity before testing. The both clamped ends of the specimens were glued by two glass fiber-reinforced plastic tabs. The strain gauges 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 each composite were obtained from seven specimens.

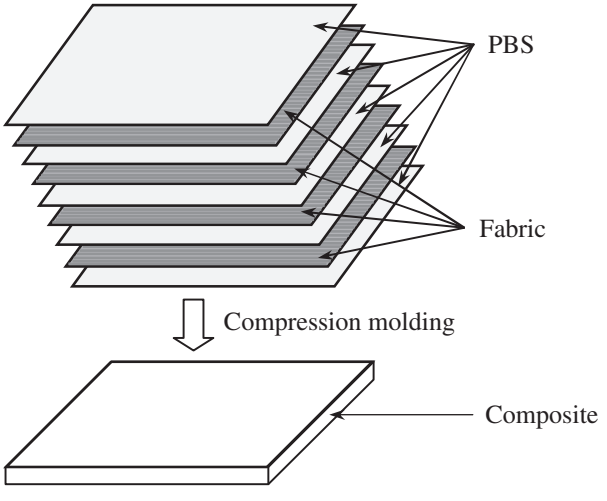

Figure 2. The fabricated procedure of jute fabric/PBS biodegradable laminated composite.

2.5. Flexural test

The flexural properties of jute fabric/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 specimens of 50 mm × 25 mm × 2 (+0.2/−0.05) mm were cut out from the composite plate by the cutting machine AC-300CF. These specimens were then put in desiccator at 25 °C and 35% relative humidity before testing. The flexural test was carried out at RT with a cross-head speed of 2 mm/min. 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:

$$\sigma_f = \frac{3FL}{2bh^2} \tag{1}$$

$$E_f = \frac{L^3 m}{4bh^3} \tag{2}$$

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

2.6. Thermogravimetric analysis

The thermal stability of jute fabric, PBS resin, and each jute fabric/PBS biodegradable composite was analyzed up to 500 °C under a purging nitrogen gas with a flow rate of 100 ml/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. Morphological characterization

The morphologies of jute surfaces and composite-fractured surfaces were examined using SEM (VE-7800, Keyence Inc., Osaka, Japan).

3. Results and discussion

3.1. Mechanical properties of UT jute fabric/PBS biodegradable composites

The mechanical properties of PBS resin and UT jute fabric/PBS biodegradable composites were given in Table 1. As Table 1 shows, the reinforcement of jute fabric layers improved considerably the mechanical properties of PBS resin. In addition, the mechanical strength and modulus of the composites gradually increased with increasing the number of jute fabric layers from four to six, but they decreased at seven fabric layers. The increase in the mechanical strength and modulus of the composites is due to the reinforcement of jute fabrics in PBS matrix, because the strength and modulus of the jute fiber are higher than those of PBS resin.[11] At low fiber content (e.g. four fabric layers), poor fiber population causes low load transfer capacity among the fibers. The decrease in the mechanical strength and flexural modulus of the composites reinforced

Table 1. Mechanical properties of PBS resin and UT jute fabric/PBS biodegradable composites.

Fabric layers	Fiber content (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Fracture strain (%)	Flexural strength (MPa)	Flexural modulus (GPa)
0[a]	0	37.5 ± 1.13	1.0 ± 0.06	9.0 ± 0.91	49.4 ± 2.42	0.86 ± 0.18
4	32.6	57.68 ± 2.75	8.17 ± 1.49	1.27 ± 0.21	73.29 ± 3.51	4.64 ± 0.44
5	41.3	64.05 ± 2.73	9.50 ± 1.51	1.25 ± 0.15	78.95 ± 2.79	5.27 ± 0.36
6	47.9	78.06 ± 4.54	11.89 ± 1.24	1.21 ± 0.17	82.04 ± 3.24	5.53 ± 0.35
7	54.0	76.46 ± 4.52	11.52 ± 1.14	1.20 ± 0.15	76.21 ± 3.65	5.41 ± 0.38

[a]PBS resin.

with seven jute fabric layers (54 wt.%) probably results from incomplete fabric wetting, because PBS resin content is not sufficient to wet all jute fabrics. The highest mechanical strength of the composite reinforced with six jute fabric layers (47.9 wt.%) might be due to adequate fabric content in the composites, resulting in greater wetting.

In this study, the PBS biodegradable composite reinforced with six jute fabric layers achieved the best mechanical properties. The PBS biodegradable composite reinforced by six UT jute fabric layers showed an increase in tensile strength by 108%, tensile modulus by 1088%, flexural strength by 66.1%, and flexural modulus by 543.8% compared with those of pure PBS. However, contrast to the mechanical strength and moduli, the addition of jute fabric strongly reduced the fracture strain of PBS resin. For instance, the fracture strain of PBS biodegradable composite reinforced with four UT jute fabric layers reduced approximately 86% compared with that of pure PBS. This is explainable that the fracture strain of PBS resin (Table 1) is much higher than that of jute fabrics.[16] As a result, ductile nature of PBS resin rapidly decreases with the incorporation of jute fabric. Moreover, the fracture strain of UT jute fabric/PBS biodegradable composites slightly decreased with increasing the jute fabric layer, because increasing the amount of reinforced fillers leads to the decrease in the amount of polymeric matrix available for the fracture strain.

3.2. Effect of surface treatment on mechanical properties of jute fabric/PBS biodegradable composites

For fiber-reinforced polymer composites, the improved interfacial bond between the fiber and polymer matrix results in greater composite strength and longer service life. The surface microstructures of the jute filaments extracted from UT, AT, and AST jute fabric were shown in Figure 3. As seen in Figure 3, the jute filaments have changed their surface structure after chemical treatment. The UT jute surface (Figure 3(a)) has several impurities and other substances like wax, pectin, or lignin. As observed in Figure 3(b), a number of lumpy strips existed on the AT jute surface. This proved that the impurities and also other substances on the jute surface were removed after AT, which caused the jute surface cleaner and rougher than before (Figure 3(b)). A rougher surface morphology was typical for the AT jutes because of the removal of lignin and hemicelluloses, resulting in improvement of fiber crystallinity.[22] Surface modification by alkalization might 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.[23] Moreover, AT of the jute could lead to a decrease in the spiral angle and increase in molecule orientation.[24] In addition,

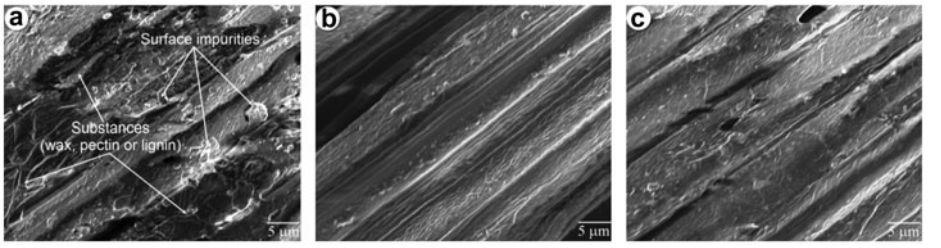

Figure 3. SEM micrographs of jute filament surface extracted from jute fabrics: (a) UT, (b) AT, and (c) AST.

AT improved the interfacial fiber–matrix adhesion because of the change in the arrangement of units in the cellulose macromolecule.[25] The hydrophilic behavior of the jute after AT induced by predominance of –OH groups will thus be weakened, and its compatibility with hydrophobic polymer will accordingly be improved.[16] After AT, the amount of cellulose exposed on the fiber surface leading to better mechanical interlocking with polymer matrix. Therefore, the development of a rough surface tomography and enhancement in aspect ratio offer better fiber–matrix interfacial bond, resulting in increasing the mechanical properties of the composites.

Furthermore, silane coupling agent was applied to modify the surface structure of AT jute so as to provide a stable bond between the jute fabric and incompatible PBS matrix (Figure 3(c)). Silane coupling agent is actually a chemical compound which functions at the interface to create a chemical bridge between the reinforcement and matrix. AT jutes were treated with hydrolyzed silane solution to allow silane penetrates into the fiber lumina and further diffuse into the cell walls. As seen in Figure 3(c), silane was effectively deposited on the jute surface as an adhesion promoter. This is explainable 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.[26–28] The silanols of γ-MPS form a monolayer on the fiber at first, and then are further adsorbed resulting in the formation of a rigid polysiloxane layer on the fiber surface. 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.[29] This is one characteristic that makes silane coupling agent useful for improving the mechanical strength and modulus of composite materials. Generally, the improvement in the mechanical properties of the composites might be mainly attributed to the improving wettability, forming chemical bonds and increasing compatibility between reinforced fibers and thermoplastic matrices. As a result, the main defects of jute as reinforcing material, such as high moisture absorption, poor wettability, and incompatibility with hydrophobic polymer, can be improved by the surface modification.

The mechanical properties of AT and AST jute fabric/PBS biodegradable composites were given in Tables 2 and 3, respectively. The effects of surface treatments on the tensile and flexural properties of the PBS biodegradable composites reinforced with different jute fabric layers were presented in Figures 4 and 5. Results show that the mechanical properties of AT and AST jute fabric/PBS biodegradable composites were significantly higher than those of untreated ones. Compared with UT jute fabric, AT and AST jute fabric/PBS biodegradable composite with six fiber layers exhibited an increase in tensile strength by 13.5 and 16.4%, in tensile modulus by 8.4 and 10.8%,

Table 2. Mechanical properties of AT jute fabric/PBS biodegradable composites.

Fabric layers	Fiber content (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Fracture strain (%)	Flexural strength (MPa)	Flexural modulus (GPa)
4	31.9	72.06 ± 3.40	9.14 ± 0.79	1.46 ± 0.15	81.62 ± 4.46	4.71 ± 0.35
5	40.1	82.45 ± 4.11	10.91 ± 1.40	1.40 ± 0.15	90.25 ± 4.12	5.57 ± 0.43
6	46.9	88.61 ± 4.42	12.88 ± 1.40	1.37 ± 0.16	99.10 ± 5.07	6.31 ± 0.49

Table 3. Mechanical properties of AST jute fabric/PBS biodegradable composites.

Fabric layers	Fiber content (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Fracture strain (%)	Flexural strength (MPa)	Flexural modulus (GPa)
4	32.4	75.42 ± 4.67	10.61 ± 1.05	1.48 ± 0.11	85.98 ± 4.00	4.84 ± 0.46
5	40.7	85.11 ± 3.93	11.64 ± 1.07	1.45 ± 0.20	94.88 ± 2.78	5.90 ± 0.34
6	47.5	90.88 ± 4.51	13.17 ± 1.40	1.40 ± 0.15	101.88 ± 4.35	6.75 ± 0.54

in fracture strain by 13.5 and 16.0%, in flexural strength by 20.8 and 24.2%, and in flexural modulus by 14.1 and 21.9%, respectively. In addition, the tensile strength and modulus of surface-treated jute fabric/PBS biodegradable composites increased with increasing the fiber layers from four to six. However, the fracture strain of jute fabric/PBS composites decreased slightly (Tables 2 and 3). This result is attributed to the fact that the fracture strain of jute fabric reported in [16] is much lower than that of PBS resin (see Table 1). It is interesting to note that with increasing the fabric layer, the flexural strength has the same trend as the tensile strength. Moreover, the mechanical properties of AST jute fabric/PBS biodegradable composites were higher than those of AT ones. Nevertheless, the enhanced mechanical properties of AST jute fabric/PBS biodegradable composites are not so much (see Tables 2 and 3). In general, surface modification of the jute fabric by AT and AST improved considerably the mechanical properties of jute fabric/PBS biodegradable composites. This reflects the contribution of alkali or silane in terms of increasing the interfacial fiber–matrix adhesion.

The increase in the mechanical properties of surface-treated jute fabric/PBS composites is attributable to greater fiber–matrix interfacial and physical bonding. As seen in Figure 6(a), the pulled-out fibers can be found on the fractured surface of the UT jute fabric/PBS composite, suggesting poor interfacial fiber-matrix adhesion. The UT jute can be easily pulled out from the interfacial region with poor compatibility, resulting in rapid partial collapse of the PBS composite.[11] For the AT (Figure 6(b)) and AST jute fabric/PBS composite (Figure 6(c)), lots of pulled-out fibers disappeared proving good compatibility being formed in PBS biodegradable composites. As shown above, AT by sodium hydroxide removed the surface impurities of the jute, thereby increasing fibers surface roughness leading to better mechanical interlocking with PBS matrix. Nevertheless, the matrix cracking and debonding of several AT jutes might be seen in Figure 6(b), because AT actually removes only certain amount of wax, pectin, lignin, and other substances covering the external surface of the fibers cell wall.[22,23] In the case of AST, a covalent bond may form between AT jute and γ-MPS after silane coupling agent.[29] The surface of AST jutes is covered by a thin layer as an adhesion

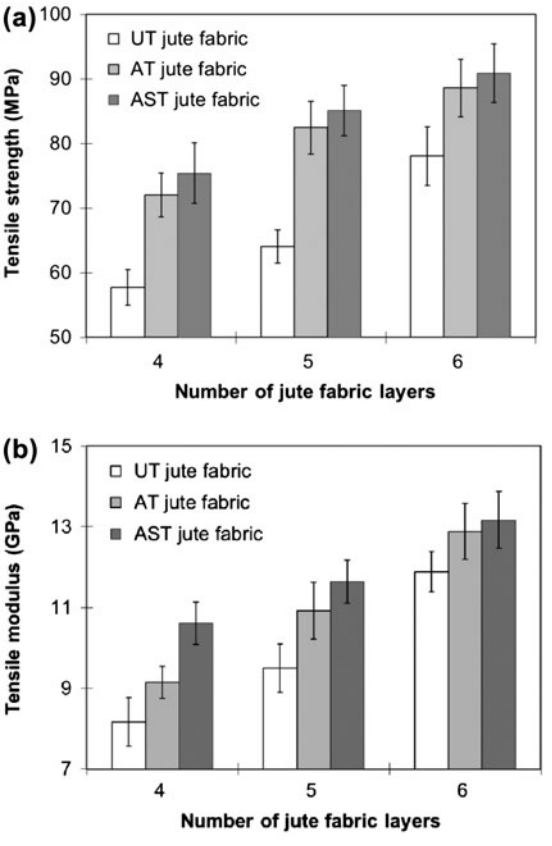

Figure 4. Effect of surface treatment on tensile strength and modulus of jute fabric/PBS biodegradable composites.

promoter making a good interfacial adhesion with PBS resin. Therefore, the composites prepared with AST jute fabrics had a predominant transverse fracture with the minimum pulled-out fibers (Figure 6(c)). This is attributable to strong interface between AST jutes and PBS matrix, leading to lowering of the critical fiber length for effective stress transfer. It is noted that the fibers failed by tearing, but that no complete interfacial failure occurred in the case of AST jute fabric/PBS biodegradable composite. This proved that the interfacial adhesion between AST jute and PBS matrix was strong enough. As a result, AST showed better improvement of jute surface properties and strengthened the fiber-matrix interaction than AT. Generally, surface-treated jute having a good adhesion with PBS matrix can effectively disperse and transfer stress. In short, surface modification of the jute is necessary to improve the mechanical properties of jute fabric/PBS biodegradable composites.

The mechanical strength and moduli of jute fabric/PBS biodegradable composites usually achieve a maximum values at a certain optimum fiber content. At the optimum fiber content, the reinforced fibers mixed homogeneously within the matrix resulting in greater wetting, thereby stress is uniformly distributed among the fibers leading to maximal properties of the composites. Several researches have reported the optimum fiber content of jute-reinforced polymer composites.[11,30,31] As shown in our

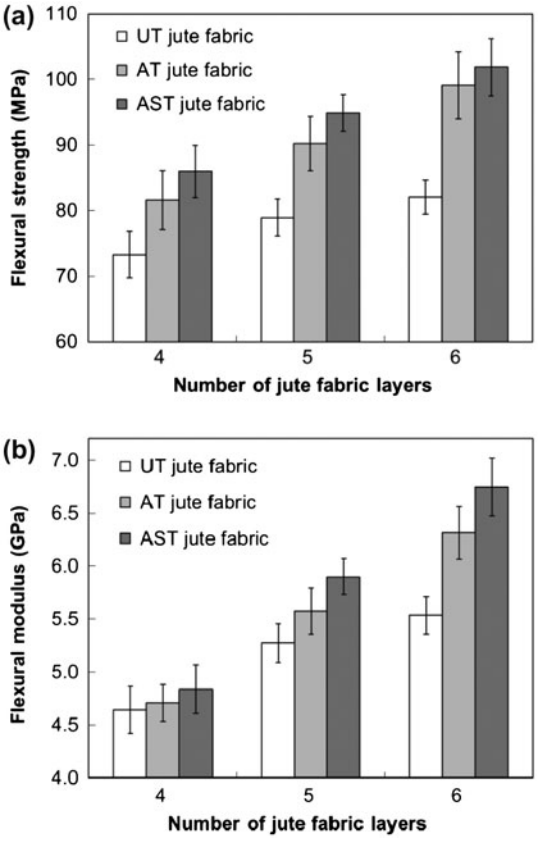

Figure 5. Effect of surface treatment on flexural strength and modulus of jute fabric/PBS biodegradable composites.

previous research,[11] the optimum fiber content of the jute fiber/PBS biodegradable composites was 50 wt.%. In this study, the best mechanical properties of jute fabric/PBS biodegradable composites were obtained with six fabric layers corresponding to approximate jute loading of 48 wt.%. Overall, the results of mechanical properties again pointed out the importance by using the right amount of jute as the reinforcement in the PBS composites.

3.3. Comparison of mechanical properties between jute fiber and fabric/PBS composites

Unlike our previous research [11] using the jute fiber as the reinforcement in the composites, in present work, we utilized the jute fabric reinforced in PBS biodegradable composites. The results in Tables 1 and 4 showed that UT jute fiber-reinforced PBS biodegradable composites have higher mechanical strength and moduli, but lower fracture strain compared with UT jute fabric/PBS biodegradable composites. This is explainable that the jute fiber has stiffer characteristics compared to the jute fabric. As reported by Gowda et al. [16], the ultimate tensile strength and elastic modulus of the jute fabric (85 MPa and 0.8 GPa) were lower than those of the jute fiber (120 MPa and

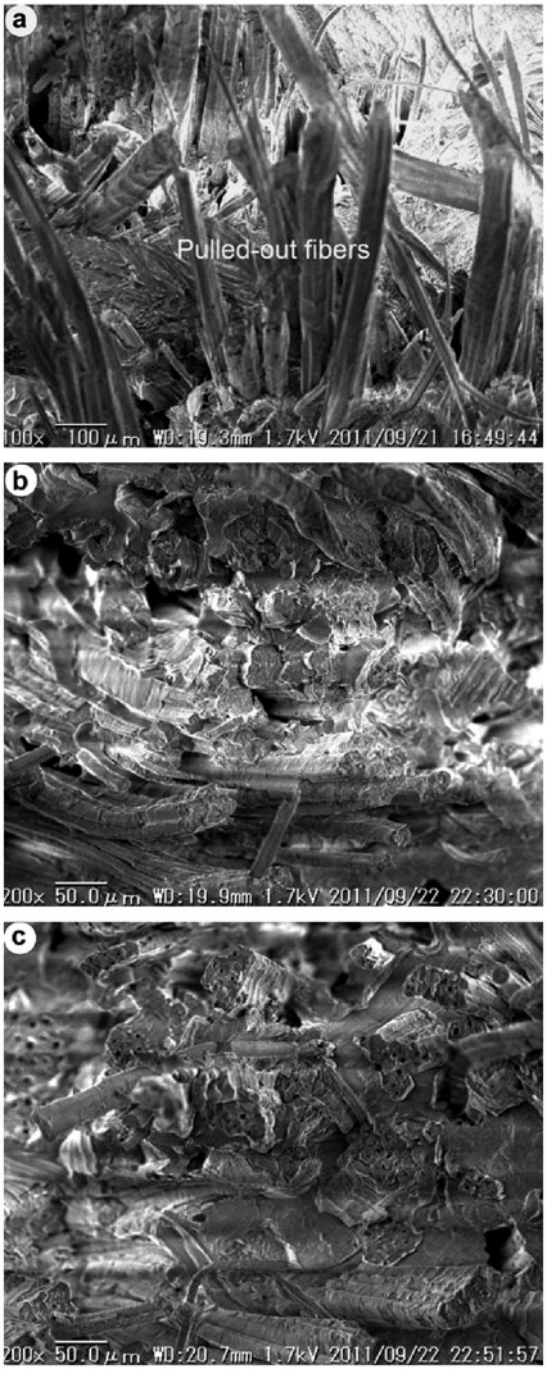

Figure 6. SEM micrograph of fractured surface of PBS biodegradable composite reinforced with six fabric layers of: (a) UT jute, (b) AT jute, and (c) AST jute.

3.75 GPa), whereas the fracture strain of the jute fabric was higher than that of the jute fiber. Evidently, the mechanical strength and moduli of fiber-reinforced composite

Table 4. Mechanical properties of UT jute fiber/PBS biodegradable composites [11].

Fiber content (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Fracture strain (%)	Flexural strength (MPa)	Flexural modulus (GPa)
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

material are mainly dependent on the mechanical strength and modulus of the fibers, as well as the strength and stability of the matrix. Moreover, the composite strength increases with decreasing fiber diameter, because small fiber can obtain higher mechanical resistance by larger specific contact surface with matrix. In addition, dry fabric can lead to lower bonding between yarns and resin, resulting in delamination and lower composite properties.[32,33] Therefore, the jute fabric/PBS composites showed lower mechanical strength and moduli than those of the jute fiber/PBS composites. The higher fracture strain can be explained by the fact that the jute fabric is more stretching than the nature of the fiber, because the jute yarns in the fabric have initial twisting.

Furthermore, the performance of fiber-reinforced polymer composites are governed by the interfacial fiber–matrix bonding strength in transferring stress. Hence, the interfacial bonding strength between the matrix and the fiber plays an important role in determining the strength of the composite. In this study, surface modification of the jute by AT and AST improved the interfacial bonding strength between the jute and PBS matrix, thereby increasing the mechanical strength and moduli of the composites. However, the mechanical strength and moduli of surface-treated jute fabric/PBS composites obtained were lower than those of same surface-treated jute fiber-reinforced PBS composites. The higher mechanical strength and moduli of the jute fiber against jute fabric-reinforced PBS composites can be due to better wetting of the jute fibers in PBS matrix compared with the jute fabric. As shown in [18], there are two kinds of interfaces that should be taken into account in the case of jute fabric/PBS composites: one between the yarns in the jute fabric and PBS matrix and the other between the jute strands in the yarn. After AT or AST of jute fabric, outer strands of the jute yarn in the fabric are surface modified resulting in good interfacial bonding strength with PBS matrix. Nevertheless, interfacial bonding between inner strands in the yarns of the fabric might be weak. As a result, inner strands of the yarns in the jute fabric could first break at low stress leading to lower mechanical strength and moduli of jute fabric/PBS composites compared with jute fiber-reinforced PBS composites.

To evaluate the effectiveness of surface modification on the jute fiber and fabric, the normalized mechanical strength and moduli of AST and UT jute fiber and fabric-reinforced PBS composites were calculated by the following formulae:

$$\sigma_{ef} = \frac{\sigma^*_{AST}}{\sigma^*_{UT}} \tag{3}$$

$$E_{ef} = \frac{E^*_{AST}}{E^*_{UT}} \tag{4}$$

where σ_{ef} and E_{ef} are the normalized strength and modulus. σ^*_{AST} and σ^*_{UT} are the mean strength of AST and UT jute(fiber or fabric)-reinforced PBS composites, respectively.

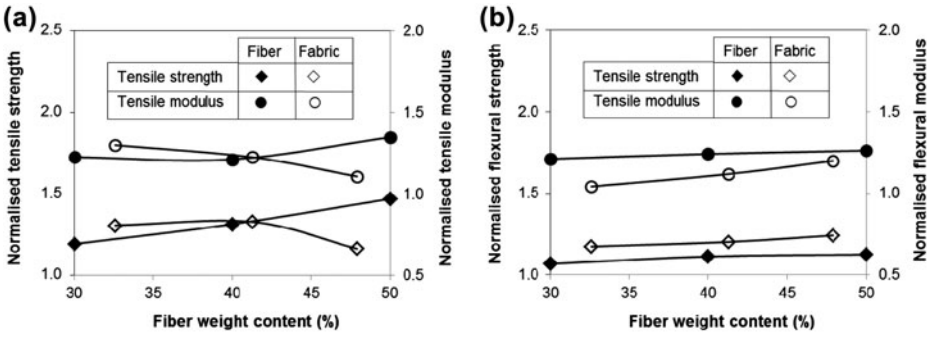

Figure 7. Comparison of mechanical strength and moduli between AST jute fiber/PBS composites [11] and AST jute fabric/PBS composites.

E^*_{AST} and E^*_{UT} are the mean elastic moduli of AST and UT jute(fiber or fabric)-reinforced PBS composites, respectively.

The comparison of the mechanical strength and moduli between AST jute fiber/PBS composites [11] and AST jute fabric/PBS composites is shown in Figure 7. As seen in Figure 7, the normalized mechanical strength and moduli are evidently higher than 1. This proved that the mechanical strength and moduli of AST jute fiber and fabric-reinforced PBS composites are higher than those of UT ones. The normalized tensile strength and modulus (Figure 7(a)) range from 1.19 to 1.47 and 1.21 to 1.35 for AST jute fiber/PBS composites, and from 1.16 to 1.33 and 1.11 to 1.30 for AST jute fabric/PBS composites, respectively. Obviously, the effectiveness of surface treatment on the tensile strength and modulus of jute fiber/PBS composite is higher than that of jute fabric-reinforced PBS composites. Moreover, the effectiveness of surface modification for jute fiber/PBS composites increased with increasing the fiber content, whereas it decreased slightly for jute fabric-reinforced PBS composites. Especially, at higher fiber weight content than 40%, the effectiveness of surface treatment raised rapidly for jute fiber-reinforced PBS composites, although it reduced significantly for jute fabric/PBS composites. This is explainable that at near-optimal fiber content, the jute fibers and PBS resin were intimately mixed leading to greater wetting, but the jute fabric was not mixed homogeneously within PBS matrix, because inner strands in the yarns of the jute fabric could not be impregnated thoroughly with PBS resin. In addition, the normalized flexural strength and modulus (Figure 7(b)) changed slightly at different fiber weight contents. As a result, the effectiveness of surface treatment is less on the flexural properties compared with the tensile properties. This can be explained by the fact that flexural failure mode usually shows little or no fiber pull-out [23].

3.4. TGA analysis

TGA is an analytical technique for measuring changes in the mass of a material that occur in response to programed temperature changes. Thermogravimetric (TG) measurements can be used to predict the thermal stability of a composite material. 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.[34] TG and DTG curves of the UT jute fabric, PBS resin, and UT jute fabric/PBS biodegradable composites were depicted

in Figure 8. The peak temperatures and weight loss percentage of PBS resin and UT jute fabric/PBS composites obtained from DTG curves are given in Table 5. As observed in Figure 8(a), the weight loss of PBS resin occurred in a one-step degradation process from 350 °C to less than 500 °C. This is confirmable by the presence of only one peak in DTG curve at temperature of 403.6 °C (Table 5 and Figure 8(b)). The weight loss of PBS resin starts near 300 °C and continues to be extremely slow at temperature below 370 °C. Above 370 °C, the weight loss occurs rapidly, thereby the quantity of PBS residue is very low due to further breakdown of it into gaseous products at higher temperature. About 56% weight loss of PBS resin is observed at the peak of temperature and 99.8% weight loss occurs at 500 °C (Table 5).

DTG thermogram in Figure 8(b) showed that the decomposition of the jute fabric occurs as a three-step process. The first step process below about 250 °C corresponds to the loss of adsorbed moisture from the fabrics. The weight loss of UT jute fabric by absorbed moisture at this stage is about 3.5%. The initial weight loss around 57 °C can be due to the evaporation of water in the jute fabric. Therefore, there is no significant weight loss in the temperature range of 100–250 °C similar to the decomposition behavior of other natural fibers, such as coir [35,36], kenaf [37], silk [38], and wood.[39] The decomposition of cellulose and hemicellulose of the jute fabrics occurs at 250 and 380 °C in second step process. In this stage, the thermal degradation rate is slow below 300 °C, but above 300 °C, the weight loss becomes fast. About 53% of the

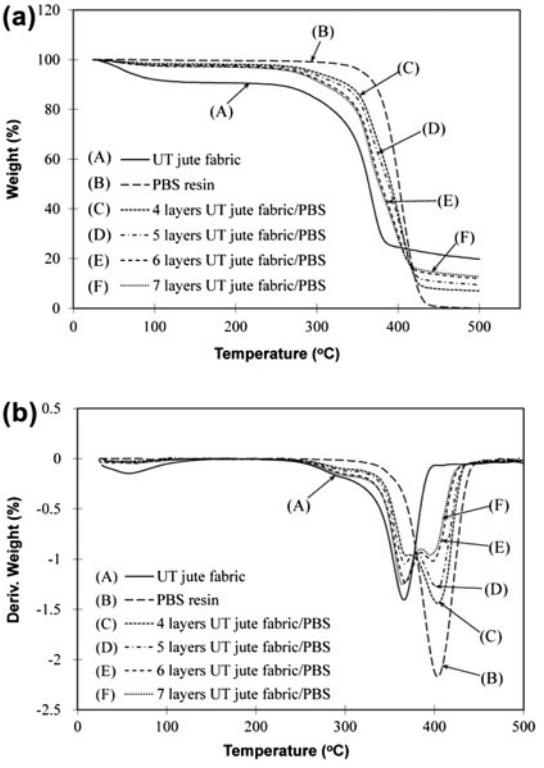

Figure 8. TG (top) and DTG (bottom) curves showing the thermal properties of UT jute fabric, PBS resin, and UT jute fabric/PBS biodegradable composites.

Table 5. Peak temperature and percent weight loss of PBS resin and UT jute fabric/PBS biodegradable composites.

Fabric layers	1st peak temperature (°C)	% 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
4	369.1	27.4	403.7	66.9	93.0
5	367.0	29.6	403.2	68.1	90.5
6	367.3	37.0	398.1	68.4	88.0
7	365.6	37.5	396.8	68.7	87.1

thermal degradation was obtained at the peak temperature between 300 °C and 380 °C. The third stage occurs above 380 °C in which the jute fabric starts to decompose with a low rate of weight loss. In the third stage, all the volatile materials are driven off from the sample resulting in the residual char.

As seen in Figure 8(a), the thermal stability of jute fabric/PBS biodegradable composites is remarkably to be intermediate between the PBS matrix and the jute fabric depending on the number of fabric layers. The thermal degradation of jute fabric/PBS biodegradable composites occurred in a two-step degradation process (Figure 8(b)). This is provable by the presence of two peaks from DTG curve: the first peak corresponds mainly to the jute degradation and the second peak relates to degradation of PBS resin. Below about 415 °C, the thermal degradation of the composites increases with increasing the fabric content, whereas it decreases above 415 °C. Moreover, the weight loss percentage of jute fabric/PBS biodegradable composites at 500 °C decreases with increasing the fabric layer (Table 5). Consequently, at low temperature region, the thermal stability of the composites decreases with increasing the jute fabric content as the same results reported in [35,36] for coir/PBS biodegradable composites. Basically, the less thermal stability of jute fabric 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 compensated by the presence of jute fabric. As a result, the reinforcing jute fabric in the PBS can act as barriers for better heat insulation and hinder the permeation of volatile degradation products into the composites.

The peak temperatures and weight loss percentage of different surface-treated jute fabric/PBS biodegradable composites are presented in Table 6. Surface-treated jute fabric/PBS composites showed a slight decrease in the weight loss percentage (Table 6) compared with that of UT ones (Table 5). This proved that the thermal stability of surface-treated composites was enhanced. Evidently, the PBS resin is well penetrated

Table 6. Peak temperature and percent weight loss of surface-treated jute fabric/PBS biodegradable composites.

	1st peak temperature (°C)		% Weight loss at 1st peak		2nd peak temperature (°C)		% Weight loss at 2nd peak	
Fabric layers	AT	AST	AT	AST	AT	AST	AT	AST
4	369.7	366.2	25.5	25.2	403.2	403.3	66.2	65.5
5	367.3	370.4	28.7	28.0	401.8	403.5	67.3	66.7
6	367.9	367.1	31.4	30.5	401.6	401.8	67.6	67.4

into the AT jute thereby resulting in a strong fiber–matrix interface. In the case of AST, the γ-MPS was first hydrolyzed in distilled water to form silanols. Then, silanol groups react with hydroxyl groups that exist on the surface of the jute because of cellulose structure in order to form strong covalent bonds or H^- bonds with OH group of cellulose.[40] As a result, the γ-MPS can form a bridge at the interface as shown in [41]. This leads to a strong interface between the treated jute and PBS resin, thereby increasing the thermal stability of the composites. In short, surface treatment of jute fabric by AT and AST increased interfacial adhesion between the jute and PBS matrix, resulting in improving the thermal stability of the jute fabric/PBS biodegradable composites.

4. Conclusions

Biodegradable laminated composites based on PBS resin and jute fabrics have been developed. Effects of jute surface modification by AT and AST on the mechanical and thermal properties of jute fabric/PBS biodegradable composites have been investigated. The following conclusions can be drawn from this study:

(1) AT improved the interfacial fiber–matrix bonding of jute fabric/PBS biodegradable composites. AST also increased the interfacial adhesion between the jute fabrics and incompatible PBS resin in the composites through an adhesion promoter deposited on the jute surface.
(2) Mechanical properties of AT and AST jute fabric/PBS biodegradable composites enhanced significantly in comparison with UT ones. The mechanical properties of AST jute fabric/PBS biodegradable composites found to be better than those of AT ones.
(3) Mechanical properties of jute fabric/PBS biodegradable composites increased with increasing the number of fabric layers from four to six, but they decreased at seven fabric layers. In this study, the best mechanical properties of AST jute fabric/PBS biodegradable composite were achieved with six jute fabric layers corresponding to fiber weight content of 47.5%.
(4) The jute fabric-reinforced PBS biodegradable composites have shown a higher fracture strain and lower mechanical strength and moduli than those of the jute fiber/PBS biodegradable composites.
(5) The less thermal stability of the PBS at high temperature region was compensated by the presence of jute fabrics. Moreover, surface treatments of the jute fabric increased the thermal stability of the composites.
(6) The present results suggest that the composite with acceptable specific properties can be formed from jute spun yarn fabric and PBS resin.

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