

This article was downloaded by: [Siauliu University Library]

On: 17 February 2013, At: 06:47

Publisher: Taylor & Francis

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



## Advanced Composite Materials

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/tacm20>

### Interfacial, Mechanical and Thermal Properties of Coir Fiber-Reinforced Poly(Lactic Acid) Biodegradable Composites

Tran Huu Nam<sup>a</sup>, Shinji Ogihara<sup>a</sup> & Satoshi Kobayashi<sup>b</sup>

<sup>a</sup> Department of Mechanical Engineering, Tokyo University of Science, 2641 Yamazaki, Noda, Chiba 278-8510, Japan

<sup>b</sup> Department of Mechanical Engineering, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo 192-0397, Japan

Version of record first published: 17 Jul 2012.

To cite this article: Tran Huu Nam, Shinji Ogihara & Satoshi Kobayashi (2012): Interfacial, Mechanical and Thermal Properties of Coir Fiber-Reinforced Poly(Lactic Acid) Biodegradable Composites, *Advanced Composite Materials*, 21:1, 103-122

To link to this article: <http://dx.doi.org/10.1163/156855112X629540>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.tandfonline.com/page/terms-and-conditions>

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae, and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand, or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

# Interfacial, Mechanical and Thermal Properties of Coir Fiber-Reinforced Poly(Lactic Acid) Biodegradable Composites

Tran Huu Nam <sup>a,\*</sup>, Shinji Ogihara <sup>a</sup> and Satoshi Kobayashi <sup>b</sup>

<sup>a</sup> Department of Mechanical Engineering, Tokyo University of Science, 2641 Yamazaki, Noda, Chiba 278–8510, Japan

<sup>b</sup> Department of Mechanical Engineering, Tokyo Metropolitan University, 1–1 Minami–Osawa, Hachioji, Tokyo 192–0397, Japan

Received 20 May 2011; accepted 13 January 2012

## Abstract

The poly(lactic acid) (PLA) biodegradable composites reinforced with short coir fibers were developed and studied. The effect of fiber mass content varying from 10 to 50% on mechanical and thermal properties of coir/PLA composites have been studied in terms of tensile properties, thermal stability and thermal expansion. The effect of alkali treatment on interfacial shear strength (IFSS), mechanical and thermal properties of coir/PLA composites was investigated. The coir fibers which are soaked in 5% sodium hydroxide solution at room temperature for 72 h showed the highest IFSS at 72.8% higher than untreated coir fibers. Alkali treatment of coir fibers enhanced the tensile properties of coir/PLA composites. The best tensile strength of coir/PLA composite was achieved at fiber mass content of 20% in this study. Below about 370°C, the coir/PLA biodegradable composites are found to have much higher thermal stability compared to coir fiber, but lower than that of PLA resin. Alkali-treated coir/PLA composites having improved fiber–matrix adhesion resulted in stable composites with better thermal stability than untreated coir/PLA composites. The thermomechanical stability of PLA resin is significantly improved by the addition of reinforcing coir fibers. The fractured surface morphology of the composite specimens exhibited an improvement of interfacial fiber–matrix adhesion in the composites reinforced with alkali-treated coir fibers.

## Keywords

Natural fiber biodegradable composites, interface, mechanical properties, thermal properties, thermomechanical properties

\* To whom correspondence should be addressed. E-mail: trannam@rs.noda.tus.ac.jp;  
thnam.hut@gmail.com  
Edited by the JSCM

## 1. Introduction

In recent past decades, natural fiber composites based on petroleum-based thermoplastics or thermosets matrices have been used in various industrial sectors, especially in the automobile industry for components such as door panels, seat backs, headliners, package trays, dashboards and interior parts [1, 2], and even in the building sector [3]. However, most commercially available petroleum-based polymers are non-biodegradable, so the composites derived from them are not fully environmentally friendly and are still a burden to the environment [4].

As a result, many researches are currently focusing on fully biodegradable composites, so-called ‘green composites’ or biocomposites, which are produced by the combination of biodegradable polymers and natural fibers [5]. Green composites have attracted great interest in recent years due to their potential applications in biomedical, bioengineering and environmental fields [6]. Natural fiber biodegradable composites have some major advantages over conventional composite materials, such as eco-friendliness, low volumetric cost, lightweight, high specific mechanical properties and biodegradable characteristics [7–9]. Nevertheless, they have disadvantages such as poor interfacial bonding between natural fiber and polymer matrix: this demands fiber surface modification to improve interfacial adhesion [10].

The commercial natural fibers such as hemp, henequen, jute, kenaf, sisal, flax, bamboo, coir, banana, palm, silk, cotton and wood are renewable resources in many developing countries. They are considered as strong candidates to replace the conventional glass fibers due to their eco-friendliness, low cost, low density, low pollutant emissions, acceptable specific properties, renewable resources and biodegradability [4, 7]. Among natural fibers, plant fibers which contain strongly polarized hydroxyl groups are hydrophilic in nature [11]. These fibers are inherently incompatible with hydrophobic thermoplastics, because the presence of pendant hydroxyl and polar groups in various constituents of the fibers. Moisture absorption of fibers is very high and 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 chemical modification [12–17].

Among the plant fibers, coir fibers are nowadays extensively used in several industrial applications. Coir is a versatile lignocellulosic fiber extracted from the tissues surrounding the seed of coconut palm (*Cocos nucifera*). Coir consists of cellulosic fibers with hemicellulose and lignin as the bonding materials for the fibers. Coir fiber has low cellulose (36–43%) and hemicellulose (0.2%), high lignin content (41–45%) and high microfibrillar angle (30–45°) compared with other natural fibers [17]. Consequently, the tensile strength and modulus of coir fiber are lower than those of other plant fibers. Moreover, due to the high microfibrillar angle [18] the elongation at break of coir is the highest among typical natural fibers [19]. This property of coir fiber is certainly useful in cushion applications, such as seat cushions for automobiles [20]. The lignin content in coir fiber is quite high and responsible for other useful properties, such as weather, fungal and bacterial

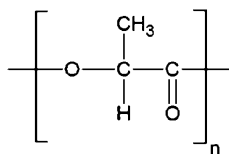
resistance [19]. Apart from typical applications of coir, such as floor-furnishing materials, yarn, rope and so on, coir has found a place in applications for automobiles, construction and other areas [3, 20].

Among the biodegradable polymers which have been frequently studied as the matrices in biodegradable composites, poly(lactic acid) (PLA) is the most widely used biodegradable polymer with increasing commercial interest [21]. PLA is a thermoplastic aliphatic polyester produced from a lactic acid monomer through fermentation of renewable resources, such as corn starch, sugar cane and sugar beet [22]. PLA is produced either by direct condensation polymerization of lactic acid or by ring opening polymerization of cyclic lactide dimer [23]. PLA has good stiffness, high strength and low elongation at break. PLA has been widely used in several applications, such as medical applications [24, 25], automotive parts [26], food packaging, water and milk bottles, degradable plastic bags, etc. [27]. However, the application of PLA polymer has been limited due to its brittle physical properties [28]. As a result, it is necessary to improve the mechanical and thermal properties of PLA by the addition of fibers or filler materials. In the present work, biodegradable composites based on PLA resin and short coir fibers were fabricated by injection molding method. The effect of alkali treatment on the interfacial shear strength (IFSS) of coir/PLA system was evaluated by single fiber pull-out test. The effect of alkali treatment and fiber mass content on mechanical and thermal properties of coir/PLA composites was studied. Fractured surface morphology of coir/PLA composite specimens were investigated by a scanning electron microscope (SEM) providing the information for the evaluation of interfacial fiber–matrix adhesion.

## 2. Experimental Procedures

### 2.1. Materials

The golden brown coir fibers in the present work were supplied from Betrimex JSC (Ben Tre, Viet Nam). Several physical, chemical and mechanical properties of coir fiber compared with other typical natural fibers such as flax, hemp, jute, ramie and sisal were reported in [17]. Biodegradable PLA pellets (Teramac TE–7000, Unitika Ltd, Tokyo, Japan) have low density and high heat resistance. The melting temperature of PLA is about 170°C, and the density is 1.27 g/cm<sup>3</sup>. Figure 1 depicts the chemical structure of PLA used in this study.



**Figure 1.** Chemical structure of PLA used in the present study.

## 2.2. Alkali Treatment of Coir Fibers

To begin with, coir fibers were treated with 5% NaOH solution in a glass beaker for different soaking times (24, 48, 72 and 96 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. Lastly, the coir fibers were air-dried for more than 2 days. The mean IFSS of 5% alkali-treated coir fibers for 72 h which will be shown in the next section is higher than that of untreated and other alkali-treated coir fibers. Therefore, another series of experiments with the same procedure were followed except that the coir fibers were soaked in 3 and 7% concentration of NaOH solution for 72 h in order to investigate the effect of alkali concentration on the IFSS. The series are designated by 3NX, 5NX and 7NX in which 3N, 5N and 7N correspond to the soaking in 3, 5 and 7% NaOH solution, respectively and X corresponds to the soaking time in hours. The reaction of sodium hydroxide with coir fiber is described as follows:

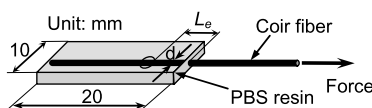


## 2.3. Single Fiber Pull-out Test

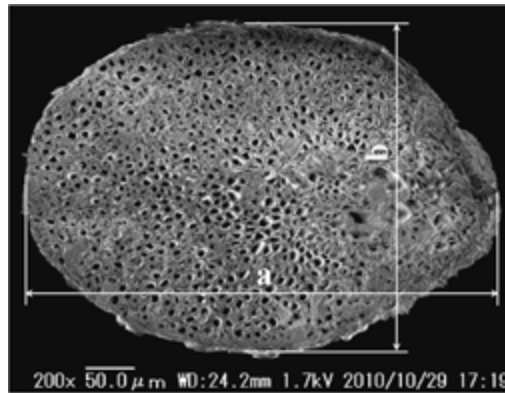
A single fiber pull-out test was used to measure the IFSS of both untreated and alkali-treated coir/PLA system. The untreated and alkali-treated coir fibers having length over 120 mm were used to make pull-out test specimens by pressing the fibers between two PLA sheets using a hot press equipment (Imoto Corp., Kyoto, Japan). The fibers were kept straight and oriented by fixing both ends, extending outside the PLA sheets, on the mold using glue as described in [17]. Specimens with a thickness of 1 mm were removed from the mold after quickly cooling by fan. The fiber embedded length in the PLA matrix was obtained by cutting the fiber by punching a hole through the specimen. The schematic representation of single fiber pull-out test is shown in Fig. 2, in which  $d$  is the mean diameter of coir fiber and  $L_e$  is the embedded length. A single fiber pull-out test was performed by a universal testing machine Instron 4442 (Instron Corp., Canton, MA, USA) with a load cell of 50 N and a crosshead speed of 1 mm/min. A force is applied to the free end of fiber to pull it out of the matrix while the force is continuously monitored and recorded. IFSS value of coir/PLA system was estimated from the maximum debonding force ( $F_d$ ) as follows:

$$\tau = \frac{F_d}{A_e}, \quad (2)$$

where  $A_e$  is the area embedded between the fiber and the matrix.



**Figure 2.** Schematic representation of single fiber pull-out test.



**Figure 3.** Typical cross-section of coir fiber.

The embedded area is calculated as follows:

$$A_e = \pi \times d \times L_e. \quad (3)$$

The diameter ( $d$ ) of coir fiber is calculated approximately as follows:

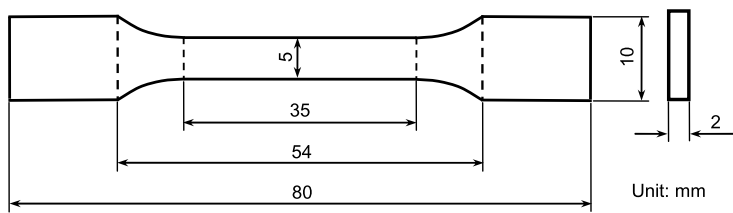
$$d = \sqrt{ab}, \quad (4)$$

in which the dimensions of  $a$  and  $b$  as shown in Fig. 3 are measured by an optical microscope MX-7575CS (Hirox Co. Ltd, Tokyo, Japan) at the intersection between coir fiber and PLA matrix.

The single fiber pull-out test was carried out for untreated, and 5N24, 5N48, 5N72, 5N96, 3N72 and 7N72 treated coir fibers to investigate the effect of soaking time and concentration of sodium hydroxide on the IFSS of coir/PLA system in order to make the optimal choice of alkali treatment. The mean IFSS between coir fibers and PLA matrix was obtained from at least twenty pull-out test specimens.

#### 2.4. Composite Fabrication

Firstly, both untreated and alkali-treated (5N72) coir fibers were dried at 80°C in the vacuum oven for 24 h. The dried coir fibers were cut into the short fibers from 10 to 20 mm long and mixed thoroughly with melted PLA resin in the mixer W50EHT (Brabender GmbH and Co. KG, Germany) at a constant temperature of 180°C. The mixed composites were immediately laminated and cut into about 5 mm long pieces. All the pieces were cut into the smaller pellets using a Strand Pelletizer (LZ 120, Sweden). The composite specimens made from PLA reinforced with untreated, and alkali-treated coir fiber at different fiber mass content (10, 20, 30, 40 and 50%) were molded from the pellets by injection. Before molding the pellets were dried at 70°C for 12 h. The dumbbell-shaped specimens, as shown in Fig. 4, were injection molded with a small size injection molded machine (NP7 Real Mini, Nissei Plastic Industrial Co. Ltd, Japan) and kept in desiccator at 25°C and 35% relative humidity before testing. The injection molding condition is listed in Table 1. Two specimens



**Figure 4.** Dumbbell-shaped specimen for tensile test.

**Table 1.**  
Injection molding condition

Mold clamping force (kN)	69
Injection pressure (MPa)	136
Fusion temperature (°C)	130
Mold temperature (°C)	35
Cooling time (s)	30

were molded for each shot. In the mold a single gate was located at the end of each specimen.

2.5. *Tensile Test*

The tensile properties of both untreated and 5N72 treated coir/PLA composites with different fiber mass content were measured according to JIS K7113 using a universal testing machine (JT Tohsi Inc., Tokyo, Japan) with a load cell of 5 kN. All the tensile tests were carried out at RT with strain rate of 0.5 mm/min. The tensile specimens were chosen carefully before testing. The mean values of tensile properties of each composite were obtained from at least seven test specimens.

2.6. *Thermogravimetric Analysis (TGA)*

The thermal stability of coir fiber, PLA and each untreated and 5N72 treated coir/PLA 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 20°C/min. The TGA and differential thermal analysis (DTA) curves were recorded.

2.7. *Thermomechanical Analysis (TMA)*

The thermomechanical stability and the coefficient of thermal expansion (CTE) were examined from RT to 100°C under a purging nitrogen gas with a flow rate of 100 ml/min using a thermomechanical analyzer TMA-60 (Shimadzu Corp., Japan). A heating rate of 10°C/min was used. The dimensions of untreated and 5N72 treated coir/PLA composite specimens were about 10 × 2 × 2 mm<sup>3</sup>.

## 2.8. Morphological Characterization

The fractured surface morphology of the composite specimens after tensile tests was examined using SEM (VE–7800, Keyence Inc., Osaka, Japan).

## 3. Results and Discussion

### 3.1. Interfacial Characterization

The single fiber pull-out test used in this study provides a simple and effective way to measure the IFSS of the coir/PLA system. The plots of the pull-out force *versus* embedded area for both untreated and 5N72 treated coir/PLA are shown in Fig. 5. The embedded area, which means the contacting area between the fiber and the matrix, was used instead of the conventional embedded length. Park *et al.* [29] reported that critical embedded area can be obtained by the intersection of two linear regression lines in which one is approach to modeling the fiber pulled-out while the other is beyond the point where fiber breakages occurred. Figure 6 shows the relation between IFSS and embedded area for both untreated and 5N72 treated coir/PLA. In general, IFSS between coir fiber and PLA matrix decreased with increasing embedded area (Fig. 6).

Table 2 shows the IFSS of both untreated and alkali-treated coir/PLA. The mean IFSS value of untreated coir/PLA system obtained is low (4.56 MPa) compared with other natural fibers, such as jute [29] and pineapple [5]. The IFSS depends mainly on two factors: mechanical interlocking and chemical bonding [5]. In the case of untreated coir/PLA, hydrogen bonding is possible between the ester group on the resin and –OH group on the fiber. However, due to the hydrophobicity of methyl groups in the resin, the probability of hydrogen bonding is low. The low IFSS value could be due to the existence of a cuticle layer and globular particles on the coir fiber surface, as seen in Fig. 7, which results in less mechanical interaction between the fiber and the matrix. Moreover, the IFSS of alkali-treated coir/PLA increases

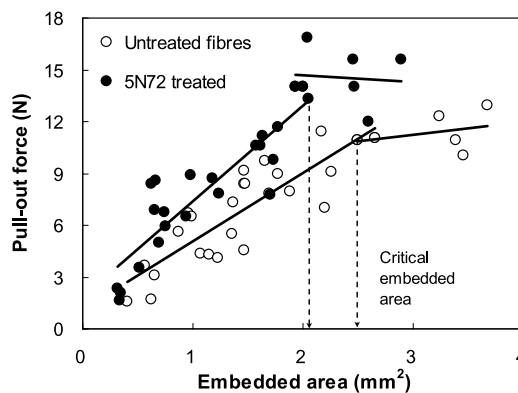
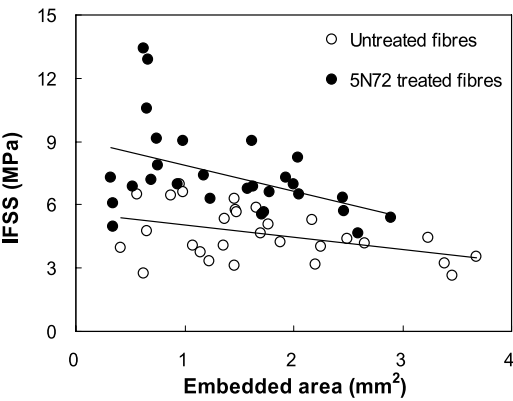


Figure 5. The pull-out force *versus* embedded area.

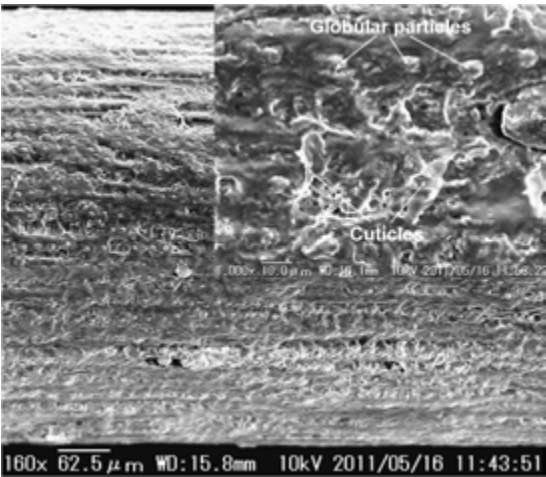




**Figure 6.** The interfacial shear strength of untreated and 5N72 treated coir/PLA *versus* embedded area.

**Table 2.**  
IFSS of coir/PLA with different fiber alkali treatment using pull-out test

Sample	Mean diameter (mm)	Embedded length (mm)	Embedded area (mm <sup>2</sup> )	Pull-out force (N)	IFSS (MPa)
Untreated	0.34 ± 0.08	1.39 ± 0.65	1.60 ± 1.04	6.74 ± 3.62	4.56 ± 1.39
5N24 treated	0.33 ± 0.07	1.44 ± 0.38	1.53 ± 0.63	8.38 ± 3.58	5.60 ± 1.03
5N48 treated	0.30 ± 0.07	1.35 ± 0.57	1.20 ± 0.87	7.10 ± 2.11	6.58 ± 2.02
5N72 treated	0.29 ± 0.08	1.08 ± 0.43	1.05 ± 0.59	7.94 ± 4.44	7.88 ± 2.26
5N96 treated	0.27 ± 0.06	1.23 ± 0.50	1.23 ± 0.60	7.66 ± 3.59	6.77 ± 2.21
3N72 treated	0.30 ± 0.05	1.54 ± 0.44	1.49 ± 0.64	10.26 ± 3.82	7.25 ± 1.83
7N72 treated	0.27 ± 0.04	1.52 ± 0.45	1.31 ± 0.55	9.92 ± 4.21	7.67 ± 1.57

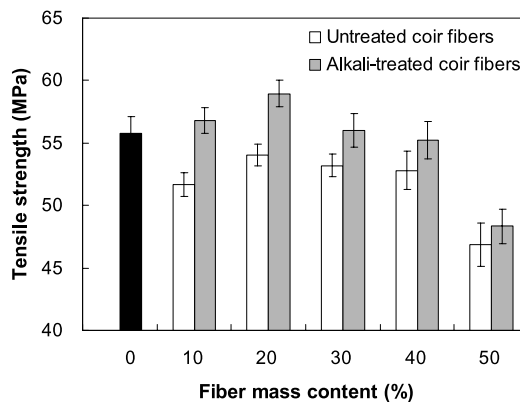


**Figure 7.** SEM micrograph of untreated coir fiber surface.

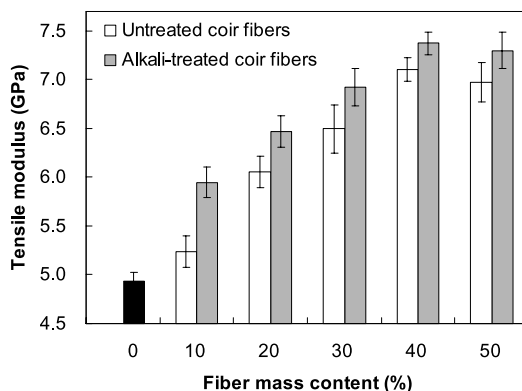
with increasing soaking time from 24 to 72 h (Table 2). This was explained due to the removal of cuticle layer and globular particles on the fiber surface during alkali treatment resulting in the increase of interfacial fiber–matrix adhesion in [17]. However, the IFSS of 5N96 treated coir/PLA is lower than that of 5N72 treated fiber. It was explained that the surface of 5N96 treated coir fiber is smoother than that of 5N72 leading to the lower mechanical interlocking between the fiber and PLA resin [17]. As seen in Table 2, the mean IFSS of 5N72 treated coir fibers is higher than that of untreated and other alkali-treated coir fibers. As a result, the effect of sodium hydroxide concentration on the IFSS between alkali-treated coir and PLA matrix was investigated and shown in Table 2. The mean IFSS of 3N72 and 7N72 treated coir/PLA is lower than that of 5N72 treated fiber. This could be due to less interfacial fiber–matrix adhesion and mechanical interaction between the fiber and the matrix [17]. In short, the best alkali treatment of coir fiber in present study is 5N72 in which coir fibers were soaked in 5% sodium hydroxide for 72 h.

### 3.2. Tensile Properties

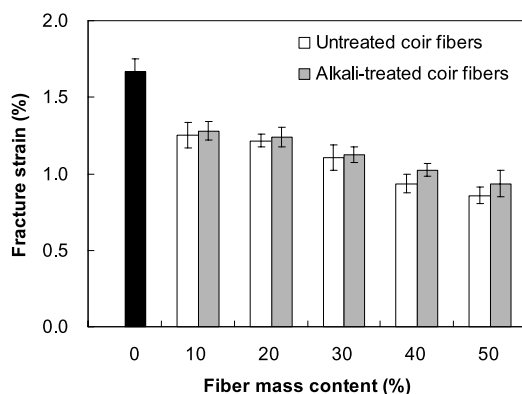
Tensile properties of both untreated and 5N72 treated coir/PLA biodegradable composites with different fiber mass content from 0 to 50% are shown in Figs 8–10. It is realized that tensile strength of the composites gradually increased with increasing fiber mass content up to 20%, but decreased with higher fiber contents (Fig. 8). The increase in the tensile strength is due to increased wetting of the fiber with the matrix. The high tensile strength at the fiber mass content of 20% might also be due to adequate fiber content in composites, which leads to greater wetting. In addition, the decrease of tensile strength of the composites with over 20% fiber mass content probably resulted from the poor fiber wetting leading to a weak interface, because the PLA resin content is not sufficient to wet all the fiber surfaces resulting in poor interfacial adhesion. In principle, lack of proper wetting between the short fiber and the matrix should lead to the formation of debondings at the fiber–matrix interface.



**Figure 8.** Tensile strength of untreated and 5N72 treated coir/PLA biodegradable composites (mean value and standard deviation).



**Figure 9.** Tensile modulus of untreated and 5N72 treated coir/PLA biodegradable composites (mean value and standard deviation).



**Figure 10.** Fracture strain of untreated and 5N72 treated coir/PLA biodegradable composites (mean value and standard deviation).

The debonding results in void formation, which lowers the tensile strength because cracks can easily propagate through regions containing voids as shown in [30]. As seen in Fig. 9, the incorporation of coir fibers improved the tensile modulus of PLA, indicating that stress transferred from the polymer matrix to the stiffer fiber. Tensile modulus of the composites gradually increased with increasing fiber mass content up to 40%, but slightly decreased with 50% fiber mass content.

Regarding the untreated coir fiber, the tensile modulus of coir/PLA biodegradable composites at 10, 20, 30, 40 and 50% fiber mass content were 6.1, 22.6, 31.6, 43.9 and 41.2% higher than that of PLA resin, respectively. This can be explained by the presence of coir fibers in the PLA matrix, which contributes effectively to enhance the tensile modulus of PLA resin, because tensile modulus of a fiber-reinforced composite depends on the modulus of the fiber and the matrix, and the fiber content. However, the tensile strength of untreated coir/PLA composites was lower than that of PLA (as seen in Fig. 8). This could be attributed to the less

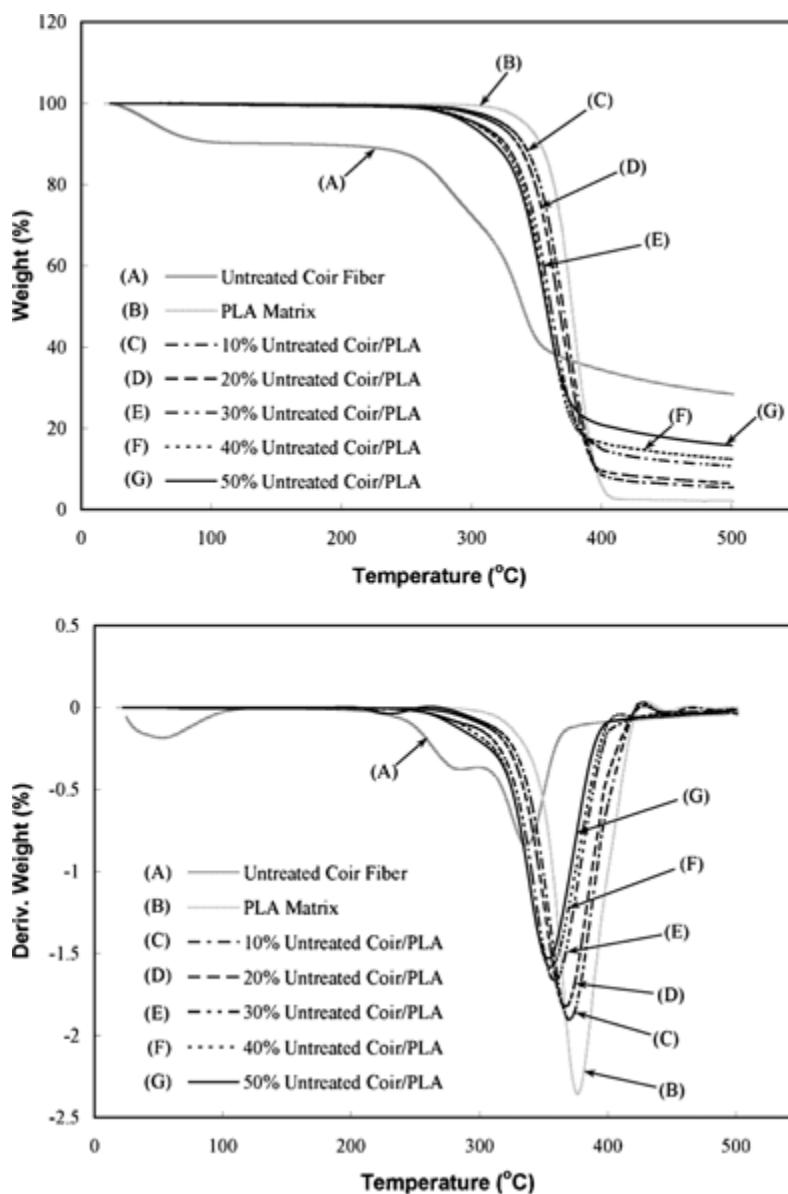
adequate adhesion between untreated coir fiber and PLA resin resulting in a poor interfacial interaction between the fiber and the matrix. Moreover, the increase in the fiber content enhances the interfacial area and weakens the fiber–matrix interaction leading to a decreasing trend of the tensile strength [15]. In general, the polymer matrix composites reinforced with high fiber content of low modulus fiber like coir which are made by applying pressure would result in a highly porous structure and their strength will be much lower than that of the matrix [16].

The addition of high fiber content can reduce fracture strain of the composite, because increasing the amount of filler will lead to the decrease in the amount of polymeric matrix available for the elongation. Figure 10 shows that the fracture strain evidently reduces compared with pure PLA. The decrease in fracture strain of coir/PLA composite is mainly due to the structural integrity of PLA being destroyed by the loading of coir fiber, and increasing fiber content implies poor interfacial fiber–matrix adhesion, leading to quicker fracture than PLA. The fracture strain of untreated coir/PLA composite at 10% fiber mass content significantly reduced by approximately 25% compared with that of PLA. Mathew *et al.* [31] reported that the lowering of fracture strain with the addition of fibers to polymers is a common trend observed in thermoplastic composites. In addition, the incorporation of higher fiber amount increases the possibility of fiber agglomeration and can lead to the formation of a stress concentrated region where less energy is required to elongate the crack propagation. Furthermore, the inefficient stress-transfer near to the stress concentrated region may result in the failure of specimens before the yield [30].

As seen in Figs 8–10, alkali treatment of coir fibers improved the tensile properties of short coir/PLA biodegradable composites. Alkali treatment of natural fibers is actually a process of surface activation leading to the formation of rough fiber surface. Alkali treatment cleans surface impurities and makes the roughness with many pits on the fiber surface [17]. The formation of the pits results in greater mechanical interlocking of the matrix on the fiber surfaces and makes the interfacial adhesion stronger. As a result, the mechanical properties of alkali-treated natural fiber-reinforced biocomposites are improved. The increase of tensile properties in the case of 5N72 treated coir/PLA composite may be due to higher fiber–matrix interfacial and physical bonding, because physical bonding also increases after alkali treatment because of the dipolar interactions between fiber and matrix [13]. The experimental results in this study showed that best tensile strength can be obtained at the fiber mass content of 20%.

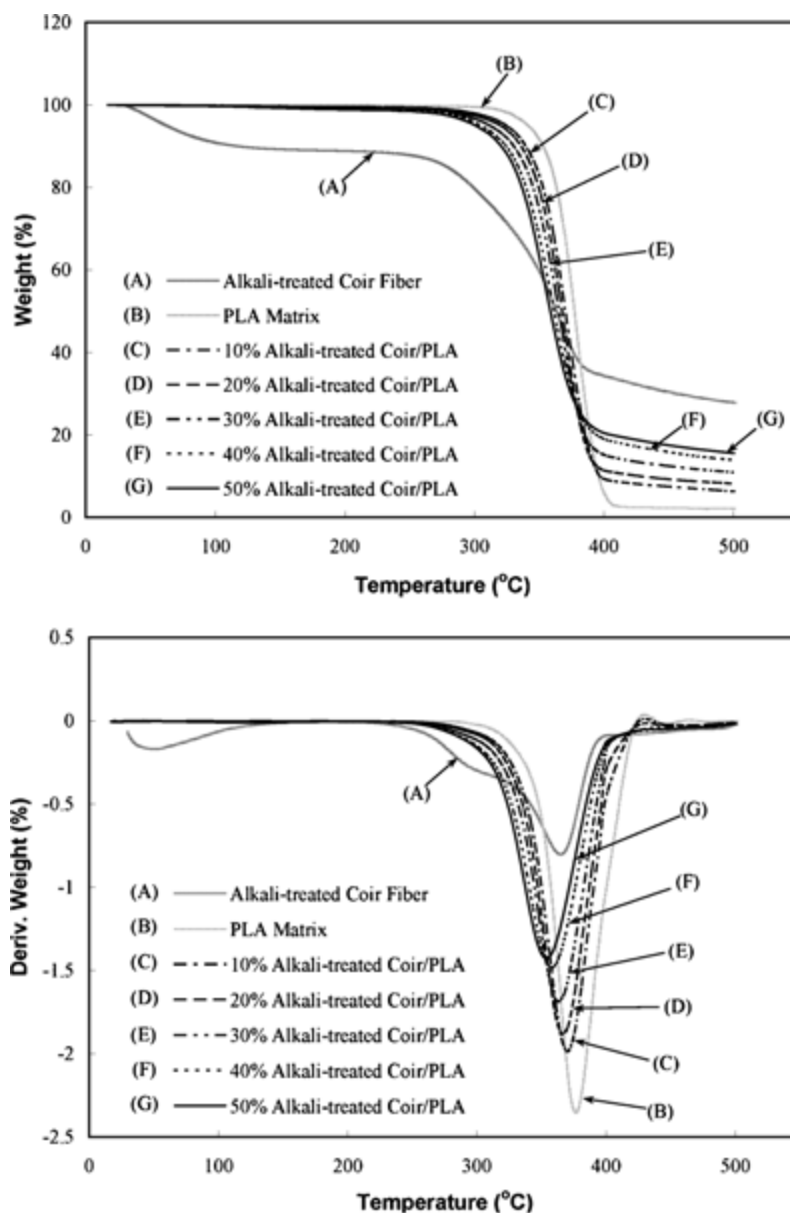
### 3.3. TGA Analysis

TGA is a useful method that is 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 [32]. Thermogravimetric (TG) and the DTG curves of un-



**Figure 11.** TG (top) and DTG (bottom) thermograms showing the thermal properties of untreated coir fiber, PLA and untreated coir/PLA biodegradable composites.

treated coir fiber, PLA and untreated coir/PLA composites are given in Fig. 11. The thermal stability of 5N72 treated coir fiber, PLA and 5N72 treated coir/PLA composites are shown in Fig. 12. The peak temperatures and the percentage of weight loss of untreated and 5N72 treated fibers obtained from DTG curves are given in Table 3. The peak temperatures and the percentage of weight losses of PLA and coir/PLA composites with different fiber mass content are shown in Table 4.



**Figure 12.** TG (top) and DTG (bottom) thermograms showing the thermal properties of 5N72 treated coir fiber, PLA and 5N72 treated coir/PLA biodegradable composites.

As observed in Figs 11 and 12, untreated and 5N72 treated coir fiber degrades through three main stages. The first stage, from 30°C to about 200°C, is attributed to the release of absorbed moisture in the coir fiber. Table 3 shows that approximately 3–5% weight loss was observed in the first stage because of absorbed moisture. The initial weight loss around 50°C can be due to the evaporation of water in the

**Table 3.**  
Peak temperature and the percent weight loss of untreated and 5N72 treated coir fibers

Sample	Peak temperature (°C)	% Weight loss at peak	% Weight loss at 500°C
Untreated coirs	53.0	4.4	71.6
	285.2	22.2	
	336.3	47.4	
5N72 treated coirs	50.5	3.2	72.2
	364.5	51.0	

**Table 4.**  
Peak temperature and percent weight loss of PLA and coir/PLA composites

Fiber mass content (%)	Peak temperature (°C)		% Weight loss at peak		% Weight loss at 500°C	
	Untreated coir fibers	5N72 treated coir fibers	Untreated coir fibers	5N72 treated coir fibers	Untreated coir fibers	5N72 treated coir fibers
0	376.4		48.3		97.8	
10	370.0	370.1	49.4	49.1	94.5	93.7
20	366.9	366.6	49.3	48.1	93.5	91.8
30	358.7	362.7	45.9	45.7	89.3	88.9
40	355.9	357.7	45.6	43.9	87.6	86.1
50	353.7	354.0	43.5	42.4	84.2	84.0

coir fiber; therefore, it is not significant weight loss in the range of 100–200°C approximately similarly with the decomposition behavior of other natural fibers, such as kenaf [30], jute [33], silk [34] and wood [35].

The decrease of percentage moisture loss in the 5N72 treated fibers could be the effect of increased crystallinity [33]. The second transition occurs from 200 to 370°C, where coir fiber undergoes thermal degradation of cellulose and hemicellulose substances. In this stage, the thermal degradation rate is slow below 250°C, but above 250°C the weight loss becomes fast. In untreated coir fiber, the main degradation peak occurs between 280 and 350°C, where about 50% of the degradation occurs. In the alkali-treated fiber, the main degradation peak temperature shifted to higher temperature. The main decomposition temperature increased from 336.3 to 364.5°C and a higher percentage of residue, which show better thermal stability of alkali-treated fibers similarly as reported in [36]. The third stage occurs above 370°C, and coir 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.

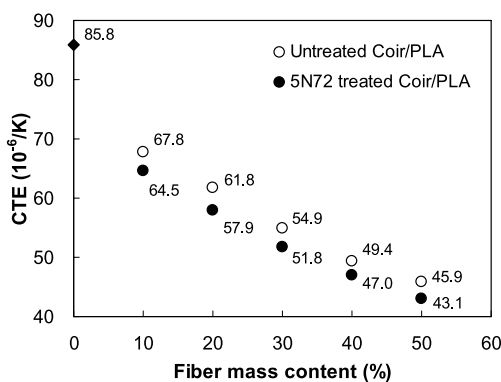
As seen in Figs 11 and 12, the thermal degradation of pure PLA starts near 300°C and the fastest weight loss occurs with a peak of temperature at 376.4°C. About 97.8% weight loss of PLA occurs at 500°C (Table 4). The thermal stability of coir/PLA composites is likely to be intermediate between the PLA matrix and the coir fiber depending on the fiber mass content. Below about 370°C, the thermal degradation of the composites increases with increasing the fiber content, but it decreases above 370°C. As a result, at low temperature region, the thermal stability of the composites decreases with increasing coir fiber content, similar to the results reported in [37] for banana fiber-reinforced phenol formaldehyde composites. The percentage weight loss for coir/PLA composites decreases with increasing fiber mass content at 500°C. In general, in the coir/PLA composites, the less thermal stability of coir fiber in the low temperature region can be compensated by the PLA, while the less thermal stability of the PLA in the high temperature region can also be compensated by the presence of coir fibers. As seen in Table 4, alkali treatment decreased the percentage weight loss and enhanced percentage residue resulting in increasing thermal stability of the composites. In addition, the resin is well penetrated into the alkali-treated fiber thereby resulting in a strong fiber–matrix interface leading to increase the stability of the composite [37]. Overall, alkali-treated coir/PLA composites having good fiber–matrix adhesion resulted in stable composites with better thermal stability than untreated coir/PLA composites.

### 3.4. TMA Analysis

The dimensional stability is a property that is a great necessary for composite materials in many applications. All materials change their size when subjected to a temperature change as long as the pressure is held constant. Dimensional changes in materials are caused by their CTE. The CTE is useful for understanding dimensional change as well as thermal stresses caused by thermal variation. Generally speaking, the CTE of a fiber-reinforced polymer matrix composite may be lower than that of a pure polymer material. A low CTE is desirable in minimizing thermo-dimensional changes in composites exposed to temperature changes during composite fabrication or use [38]. The measurement of CTE was obtained from a TMA test. The linear CTE of each specimen was determined in the temperature range of 30–80°C, where there was almost no weight loss observed by TGA. The CTE values of PLA, untreated and 5N72 treated coir/PLA composites with different fiber mass content are shown in Fig. 13.

As seen in Fig. 13, the thermal expansion gradually decreases with increasing the coir fiber content in the PLA matrix. The CTE value of PLA is higher than that of both untreated and 5N72 treated coir/PLA composites. This may be due to the thermal expansion, which becomes much less with increased fiber loading, reflecting the thermal restraint of the PLA matrix by the coir in the composite. As a result, dimensional stability of PLA resin is improved by the incorporation of reinforcing coir into the matrix. Furthermore, the CTE values of 5N72 treated coir/PLA composites are lower than those of the untreated one. The decrease in the





**Figure 13.** The CTE of PLA, untreated and 5N72 treated coir/PLA biodegradable composites.

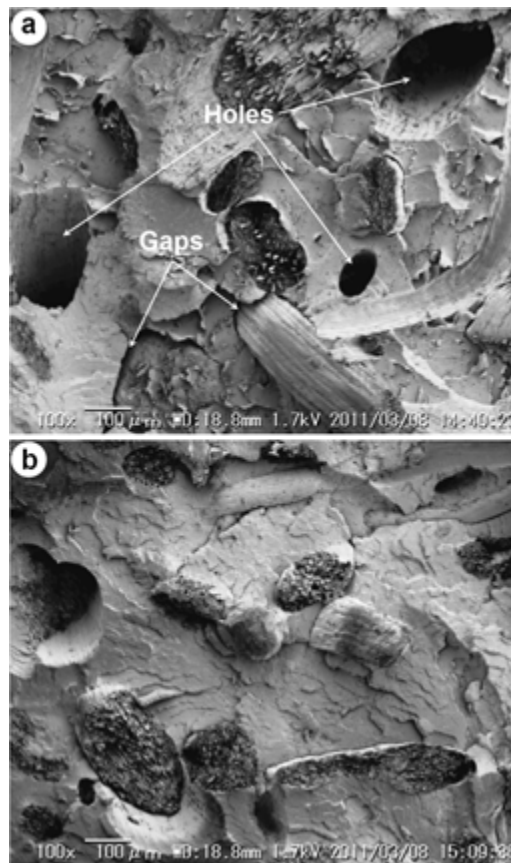
CTE of 5N72 treated coir/PLA composites is due to good fiber–matrix interfacial adhesion. The CTE value of 5N72 treated coir/PLA composite with 50% fiber mass content decreased about 33% compared with that of PLA resin.

### 3.5. Fracture Surface Morphology

The fracture surface morphologies of coir/PLA composite specimens with 20% fiber mass content are shown in Fig. 14. As can be seen in Fig. 14(a), several holes remained after the fibers were pulled-out from the matrix when stress was applied, for this led to a fractured surface occurring at the weak fiber–matrix interface. Several visible gaps can be found on the fracture surface, suggesting poor interfacial adhesion. This can be explained as fibers can easily be pulled out from the interfacial region with poor compatibility. Besides, the weak boundary layers probably caused by incomplete wettability or bonding between the coir fiber and PLA matrix may be created during composite fabrication. However, the gaps have almost disappeared in the case of 5N72 treated coir fiber (Fig. 14(b)), proving good compatibility being formed in PLA composites. Alkali-treated coir fiber having a good adhesion with PLA matrix can effectively disperse and transfer stress leading to the improvement in interfacial, thermal and mechanical properties of coir/PLA biodegradable composites. Consequently, alkali treatment of coir fiber is necessary to enhance the interfacial fiber–matrix adhesion prior to composite processing.

## 4. Conclusions

Biodegradable composites based on PLA resin and short coir fibers have been developed. The effect of alkali treatment on the interfacial, mechanical and thermal properties of coir/PLA biodegradable composites has been studied. The following conclusions can be drawn from this study:



**Figure 14.** SEM micrographs of tensile fracture surface of PLA biodegradable composite reinforced with 20% mass content of: (a) untreated coir fibers, (b) 5N72 treated coir fibers.

1. Alkali treatment of coir fiber enhanced the IFSS of coir/PLA composite. The highest IFSS between coir fiber and PLA matrix was obtained when coir fibers are soaked in 5% sodium hydroxide for 72 h.
2. Alkali treatment of coir fibers increased the interfacial bonding strength and the wettability of the fibers by PLA resin leading to the enhancement in mechanical properties of the composites.
3. Tensile strength of coir/PLA biodegradable composites increased with increasing fiber mass content up to 20%, but decreased over 20% fiber content. The tensile modulus gradually increased with increasing fiber mass content up to 40%, but slightly decreased with 50% fiber mass content.
4. Thermal stability of coir/PLA biodegradable composites is noted to be intermediate between the PLA resin and coir fiber depending on the fiber content.

Alkali treatment decreased the percentage weight loss resulting in increase in the thermal stability of the composites.

5. The thermal expansion of coir/PLA composites gradually decreased with increasing the fiber content. The thermomechanical stability of PLA matrix was improved by addition of reinforcing coir fibers.
6. The present results suggest that the coir/PLA biodegradable composites are capable of offering favorable mechanical and thermal properties for several applications where biocomposites with moderate strength may be desirable.

### Acknowledgements

The authors are grateful to Prof. Y. Kogo, Prof. A. Yasumori, Prof. K. Nishio, Prof. R. Tamura and Dr. H. Hyodo for the kind guidance to carry out thermal analysis. Authors are also thankful to Mr T. Izaki, Mr S. Mieno, Ms Y. Miyauchi and Dr. N. Venkatachalam for their assistance in the experimental work.

### References

1. Y. Chen, O. Chiparus, L. Sun, I. Negulescu, D. V. Parikh and T. A. Calamari, Natural fibers for automotive nonwoven composites, *J. Indust. Textiles* **35**, 47–62 (2005).
2. J. Holbery and D. Houston, Natural fiber-reinforced polymer composites in automotive applications, *JOM* **58**, 80–86 (2006).
3. J. Khedari, S. Charoemvai and J. Hiruanlabh, New insulating particle boards from durian peel and coconut coir, *Build. Environ.* **3**, 435–441 (2003).
4. M. Q. Zhang, M. Z. Rong and X. Lu, Fully biodegradable natural fiber composites from renewable resources: all plant fiber composites, *Compos. Sci. Technol.* **65**, 2514–2525 (2005).
5. S. Luo and A. N. Netravali, Interfacial and mechanical properties of environment-friendly ‘green’ composites made from pineapple fibres and poly(hydroxybutyrate-co-valerate) resin, *J. Mater. Sci.* **34**, 3709–3719 (1999).
6. H. Y. Cheung, M. P. Ho, K. T. Lau, F. Cardona and D. Hui, Natural fibre-reinforced composites for bioengineering and environmental engineering applications, *Composites Part B* **40**, 655–663 (2009).
7. S. N. Monteiro, F. P. D. Lopes, A. S. Ferreira and D. C. O. Nascimento, Natural-fiber polymer-matrix composites: cheaper, tougher, and environmentally friendly, *JOM* **61**, 17–22 (2009).
8. K. G. Satyanarayana, G. G. C. Arizaga and F. Wypych, Biodegradable composites based on lignocellulosic fibers: an overview, *Prog. Polym. Sci.* **34**, 982–1021 (2009).
9. A. K. Mohanty, M. Misra and G. Hinrichsen, Biofibres, biodegradable polymers and biocomposites: an overview, *Macromol. Mater. Eng.* **276/277**, 1–24 (2000).
10. J. George, M. S. Sreekala and S. Thomas, A review on interface modification and characterization of natural fiber-reinforced plastic composites, *Polym. Eng. Sci.* **41**, 1471–1485 (2001).
11. B. S. Westerlind and J. C. Berg, Surface energy of untreated and surface-modified cellulose fibres, *J. Appl. Polym. Sci.* **36**, 523–534 (1988).
12. M. J. John and R. D. Anandjiwala, Recent development in chemical modification and characterization of natural fiber-reinforced composites, *Polym. Compos.* **29**, 187–207 (2008).

13. J. Rout, S. S. Tripathy, S. K. Nayak, M. Misra and A. K. Mohanty, The influence of fiber surface modification on the mechanical properties of coir–polyester composites, *J. Appl. Polym. Sci.* **22**, 468–476 (2001).
14. M. M. Rahman and M. A. Khan, Surface treatment of coir (*Cocos nucifera*) fibers and its influence on the fibers' physico-mechanical properties, *Compos. Sci. Technol.* **67**, 2369–2376 (2007).
15. M. N. Islam, M. R. Rahman, M. M. Haque and M. M. Huque, Physico-mechanical properties of chemically treated coir reinforced polypropylene composites, *Composites Part A* **41**, 192–198 (2010).
16. S. V. Prasad, C. Pavithran and P. K. Rohatgi, Alkali treatment of coir fibers for coir–polyester composites, *J. Mater. Sci.* **18**, 1443–1454 (1983).
17. T. H. Nam, S. Ogihara, N. H. Tung and S. Kobayashi, Effect of alkali treatment on interfacial and mechanical properties of coir fiber-reinforced poly(butylene succinate) biodegradable composites, *Composites Part B* **42**, 1648–1656 (2011).
18. G. G. Silva, D. A. De Souza, J. C. Machado and D. J. Hourston, Mechanical and thermal characterization of native Brazilian coir fiber, *J. Appl. Polym. Sci.* **76**, 1197–1206 (2000).
19. K. G. Satyanarayana, A. G. Kulkarni and P. K. Rohatgi, Potential of natural fibres as a resource for industrial material in Kerala, *J. Sci. Ind. Res.* **40**, 222–237 (1981).
20. S. N. Monteiro, L. A. H. Terrones, F. P. D. Lopes and J. R. M. d'Almeida, Mechanical strength of polyester matrix composites reinforced with coconut fiber wastes, *Revista Matéria* **10**, 571–576 (2005).
21. S. H. Lee and A. Wang, Biodegradable polymers/bamboo fiber biocomposite with bio-based coupling agent, *Composites Part A* **37**, 80–91 (2006).
22. S. Jacobsen, P. H. Degee, H. G. Fritz, P. H. Dubois and R. Jerome, Polylactide (PLA) — a new way of production, *Polym. Eng. Sci.* **39**, 1311–1319 (1999).
23. D. A. Garlotta, Literature review of poly (lactic acid), *J. Polym. Environ.* **9**, 63–84 (2002).
24. S. Vainionpää, P. Rokkanen and P. Törmälä, Surgical applications of biodegradable polymers in human tissues, *Prog. Polym. Sci.* **14**, 679–716 (1989).
25. R. J. Kroeze, M. N. Helder, L. E. Govaert and T. H. Smit, Biodegradable polymers in bone tissue engineering, *Materials* **2**, 833–856 (2009).
26. T. Ino, Y. Kageyama, J. Koizumi and T. Mori, Applied investigation of bio-plastics, in: *Proc. JSAE Ann. Congr.*, pp. 11–13 (2003).
27. M. Jonoobi, J. Harun, A. P. Mathew and K. Oksman, Mechanical properties of cellulose nanofiber (CNF) reinforced polylactic acid (PLA) prepared by twin screw extrusion, *Compos. Sci. Technol.* **70**, 1742–1747 (2010).
28. D. Cohn and A. H. Salomon, Designing biodegradable multiblock PCL/PLA thermoplastic elastomers, *Biomaterials* **26**, 2297–2305 (2005).
29. J. M. Park, P. G. Kim, J. H. Jang, Z. Wang, B. S. Hwang and K. L. DeVries, Interfacial evaluation and durability of modified jute fibers/polypropylene (PP) composites using micromechanical test and acoustic emission, *Composites Part B* **39**, 1042–1061 (2008).
30. Z. Liang, P. Pan, B. Zhu, T. Dong and Y. Inoue, Mechanical and thermal properties of poly(butylene succinate)/plant fiber biodegradable composites, *J. Appl. Polym. Sci.* **115**, 3559–3567 (2010).
31. A. P. Mathew, K. Oksman and M. Sain, Mechanical properties of biodegradable composites from poly(lactic acid) (PLA) and microcrystalline cellulose (MCC), *J. Appl. Polym. Sci.* **97**, 2014–2025 (2005).
32. S. Renneckar, A. G. Zink-Sharp, T. C. Ward and W. G. Glasser, Compositional analysis of thermoplastic wood composites by TGA, *J. Appl. Polym. Sci.* **93**, 1484–1492 (2004).

33. D. Ray, B. K. Sarkar, R. K. Basak and A. K. Rana, Study of the thermal behavior of alkali-treated jute fiber, *J. Appl. Polym. Sci.* **85**, 2594–2599 (2002).
34. S. O. Han, S. M. Lee, W. H. Park and D. Cho, Mechanical and thermal properties of waste silk fiber-reinforced poly(butylene succinate) biocomposites, *J. Appl. Polym. Sci.* **100**, 4972–4980 (2006).
35. M. S. Huda, L. T. Drzal, M. Misra and A. K. Mohanty, Wood-fiber-reinforced poly(lactic acid) composites: evaluation of the physicommechanical and morphological properties, *J. Appl. Polym. Sci.* **102**, 4856–4869 (2006).
36. S. C. Saha, A. K. Ray, S. N. Pandey and K. Goswamy, IR and X-ray diffraction studies of raw and chemically treated pineapple leaf fiber (PALF), *J. Appl. Polym. Sci.* **42**, 2767–2772 (1991).
37. S. Joseph, M. S. Sreekala and S. Thomas, Effect of chemical modifications on the thermal stability and degradation of banana fiber and banana fiber-reinforced phenol formaldehyde composites, *J. Appl. Polym. Sci.* **110**, 2305–2314 (2008).
38. M. W. Lee, S. O. Han and Y. B. Seo, Red algae fibre/poly(butylene succinate) biocomposites: the effect of fibre content on their mechanical and thermal properties, *Compos. Sci. Technol.* **68**, 1266–1272 (2008).