Effect of silane coupling agent on properties of biocomposites based on poly(lactic acid) and durian rind cellulose

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Abstract. Durian rind cellulose reinforced poly(lactic acid) (PLA) biocomposites were prepared using Brabender internal mixer followed by hot compression molding technique. Cellulose was previously treated by 3-aminopropyltriethoxysilane for improving the compatibility with PLA matrix. The silane-grafting of cellulose was confirmed via Fourier transform infrared spectroscopy (FTIR) with the presence of Si-O-Si, Si-C, and Si-O-C bonds. The silane-treated cellulose was subsequently introduced into PLA matrix, and the effects of cellulose surface modification on mechanical, thermal and morphological properties, and water absorption of biocomposites were studied. It was found that silane-treated cellulose reinforced biocomposites have superior mechanical properties compared with untreated cellulose reinforced biocomposites. The lowest crystallization temperature of silane-treated biocomposites was confirmed via Differential scanning calorimetry (DSC). Scanning electron microscopy (SEM) investigation also showed that adhesion of cellulose and PLA matrix was improved by modification of cellulose surface using 3-aminopropyltriethoxysilane which can result in less water absorption into biocomposites.

1. Introduction

Biocomposite material is defined as the combination of natural filler and a matrix. A matrix is obtained from petroleum-based polymer, such as polyethylene, polypropylene, and biopolymer, such as polylactic acid (PLA) and polyhydroxyalkanoate (PHA) [1, 2]. However, biocomposite materials made from petroleum-based polymers cannot break down completely; only the natural filler will decompose whereas the matrix remains and affects the environment negatively. Therefore, the development of a fully biodegradable polymer using natural fiber as a reinforcement or filler in biopolymer has been attracting extra attention home and abroad in recent years [3]. Compared with conventional reinforcement materials produced from inorganic fiber, natural fiber has various advantages, including low density, durability and...
comparable strength, and most importantly, renewability and biodegradability [4]. However, high costs due to low volume production affect the widespread usage of these biocomposite materials for the manufacturing scale. Thus, they are suitable for the packaging industry [5, 6] and minor strength requirement applications including short-life-cycle products and indoor used products for example, device using in household, food packaging and disposable utensils [2, 7].

In the current study, the matrix phase was poly (lactic acid), or PLA, and the filler phase was cellulose from durian rinds. The combination of the two components is aimed at developing a composite suitable for light and short-term packaging applications. PLA is considered as a fully biodegradable polymer for commercial use. It is made from the fermentation of renewable agricultural raw materials to produce lactic acid. It has high performance in mechanical properties and good processability compared with other biodegradable materials [8,9]. Moreover, its packaging performance characteristics are similar to polyethylene terephthalate (PET) [10, 11]. Therefore, it has become one of the most promising biodegradable polymers to replace conventional synthetic polymers for industrial plastic applications [12].

Durian fruit called as “king of fruits” has been well-known and most consumed in Southeast Asia region. However, two-thirds of the fruit; the seeds and peels are not edible making large-scale of biological wastes [13, 14]. In common practice, the wastes are incinerated or dumped in landfills without awareness of their negative impact [15]. In long-term, the conversion of these wastes into added value product should be an alternative way. Durian rinds have been found to be a good source of cellulose [16]. Meanwhile, the study of using cellulose from durian rinds as the reinforcing material in PLA has not been explored deeply.

Although natural fibers provide varying advantages for biocomposites, the using of natural fiber as a reinforced material is also restricted. Amiscibility gap due to incompatibility between surface of the fiber and the matrix becomes a main bottleneck [8, 17]. The fiber performs high hydrophilic behavior owing to its rich in polar of hydroxyl and also other polar groups while the matrix is, generally, non-polar and demonstrates hydrophobicity [18]. Thus, the surface adhesion of two components is generally somewhat poor [17, 19, 20]. This weakness affects the reduction of stress transfer ability from the matrix phase to filler phase and resulting decrease of strength [21]. Therefore, an enhancement of the compatibility of natural fillers and biodegradable polymers is needed.

Various developments of the adherence between the cellulose and PLA surfaces have been done. The surface modification of cellulose via esterification or graft copolymerization onto cellulose substrates, and the use of compatibilizers, such as maleated polylactide and isocyanate, has been studied [8, 22].

To enhance the property of fiber reinforced composites, a commercially coupling agent named silane is widely used for fiber treating [17]. Generally speaking, coupling agent represents two functional groups. The available hydroxyl (-OH) groups on the surface of natural fiber react with the former one by hydrogen bonding whereas the latter improves mechanical properties by covalent bonding between the fiber phase and the matrix phase [14, 17]. Hydrolization of ethoxy group from 3-aminopropyltriethoxysilane (APS) in water and solvent produces a silanol. Then, the stable covalent bonding is formed by reaction between silanol and -OH group on the surface of fiber, as shown in figure 1 [22, 23]. On the contrary, COO-sites on the hydrolyzed PLA structure form hydrogen bonding with amino groups were given by APS [22, 23, 24]. The aim of the present research is to investigate the effects of surface modification of cellulose derived from durian rinds on the mechanical and thermal properties, morphological feature, and water absorption behavior of biocomposites.
2. Materials and methods

2.1. Materials and chemicals
Durian rinds (cultivar D159 Monthong) were collected from Phatthalung Province, Thailand. Poly(lactic acid) (PLA) resin (Ingeo biopolymer 2003Dfood packaging-grade with >98% purity; specific gravity of 1.24 and 6 g/10 min melting flow index at 210 °C) was purchased from Natureworks (USA). Reagent-grade of glacial acetic acid (\(\text{CH}_3\text{COOH}\)) and sodium hydroxide (NaOH), and technical-grade of 80% purity sodium chlorite (NaClO₂) and 99% purity 3-aminopropyltriethoxysilane (APS) were supplied by Fisher Chemicals Sdn Bhd. (Malaysia).

2.2. Extraction of cellulose from durian rinds
The durian rinds were first sun-dried and then ground. Two-step process according to [14, 25, 26] with slight modifications was used to produce cellulose. First, bleaching was used to remove lignin. Twenty grams of the sample was rinsed with tap water for preliminary cleaning. The sample was subsequently soaked in 640 ml of hot distilled water contained in a 1,000-ml beaker. The sample was then immersed in a water bath at 70 °C. Then, 8 g of NaClO₂ and 4 ml of CH₃COOH were added into the sample. At an hour interval, these two chemicals were continuously added to the beaker for 5 h until the color of sample became white which indicated that lignin was completely removed from the sample. The bleached sample is known as holo cellulose. Finally, holo cellulose was then filtered, and tap water was used to remove a residue chlorine dioxide and yellow color. Mercerization is the second step in which cellulose was derived from converting of holo cellulose using alkali solution. Alkali solution, 80 ml of 17.5 % w/v NaOH was poured into holo cellulose. A glass rod was used to stir the mixture and was then left for 5 min at ambient temperature. Every 5 min, another 40 ml of 17.5% w/v NaOH was added for three times and left for another 30 min. Next, the sample was mixed with 240 ml of distilled and left 1h before filtering. Then, 800 ml of 8.3% w/v NaOH was added and left for 5 min. The sample was then rinsed with water. The adding 120 ml of 10% v/v acetic acid for 5 min purposed to neutralize sample. Next, the sample was filtered and residue acetic acid was removed using distilled water. The sample was then dried to remove moisture in oven for overnight at 80°C. Finally, the cellulose was ground using a grinder and then passed through a sieve. The ground cellulose was collected with size between sieves 250μm and 125 μm and kept in an airtight box at room temperature.

2.3. Preparation of silane- treated cellulose
The cellulose was treated with silane coupling agent according to [22] with slight modification. First, the solution was prepared. Distilled water was mixed with ethanol with a ratio of 40:60 w/w for 600 ml. Next, 5 wt.% of APS (compared with the weight of cellulose) was hydrolysed in water-ethanol solution. The solution was adjusted pH to between 3.5 and 4 using an acetic acid and was then stirred 1 h. The APS solution was poured onto10 g of dried cellulose and left for 3 h. Then, the treated cellulose was filtered and dried in ambient condition for 3 days. Finally, residue moisture was then expelled using an oven at 80°C for 12 h.
2.4. Preparation of biocomposites
An internal mixer (Brabender Plastograph EC, Germany) at 165 °C was used to prepare a mixture of PLA-cellulose with ratio of 65:35 w/w. The blending was processed for 15 min using screw speed of 50 rpm. Before further step, the biocomposites were oven dried to avoid pores’ formation at 80 °C for 16 h. Then, the molded sheets were then compressed at 160 °C using a hot press machine as shown in figure 2. For tensile testing specimens with 1-mm thickness, the process was conducted at 5 min of preheating, 5 min of compressing, and 2 min of cooling.

![Figure 2](image)

2.5. Characterization

**Fourier transform infrared spectroscopy (FTIR).** The functional groups of untreated and APS-treated cellulose were identified via the FTIR Perkin Elmer, Spectrum One FT-IR Spectrometer (USA). The test was carried out in the wavenumber range of 4000 to 650 cm⁻¹ with the transmittance mode at resolution of 4 cm⁻¹. All spectra were recorded.

**Tensile measurement.** Tensile properties were measured according to ASTM 1882L using an instron universal testing machine Model 5566 (USA). Three important properties; tensile strength, modulus of Young and elongation at break were recorded. The rectangular specimen with 63 mm length, 12.7 mm width, and 1 mm thickness was prepared. The test was run at a cross speed of 5 mm/min. At least five replications of each treatment were tested.

**Differential scanning calorimetry (DSC).** Thermal properties were analyzed via the DSC model Perkin Elmer-DSC7 (USA). Ten mg of specimen was packed in aluminum cup and sealed. The sealed sample was then placed into heating chamber of the DSC. The DSC was heated with 5 °C/min from 30 to 200 °C. Nitrogen gas was flushed with flow rate 10 ml/min.

**Morphological characterization.** Characterization of the cracked surfaces of biocomposites was carried out using the SEM model S-3400N, Hitachi (Japan). All surfaces were then scanned at 15 kV after the first coated with a thin layer of gold in a vacuum.
Water absorption behaviour. Five specimens of each treatment with 30 mm length, 12.7 mm width, and 3 mm thickness were prepared. The rectangular specimens were then oven dried overnight at 80°C and were then cooled in a desiccator. The weight and the thickness of each specimen were recorded. The specimens were soaked in distilled water at ambient temperature. At 24 h intervals, each specimen was then removed from water. The tissue paper was used to wipe off the surface water and subsequently weighted by electronic balance to a precision of 0.0001 g. The measurement was terminated at 50 days. The percentage of water uptake ($Mt$) was computed as formulation (1) [27].

$$Mt \% = \left( \frac{W_t - W_o}{W_o} \right) \times 100\%$$ (1)

Where $W_t$ is the weight of specimen at the given test time and $W_o$ is the weight of the specimen after oven dried. All data from five replications were averaged [28].

The diffusion of water was performed theoretically by Fick’s law as formulation (2) [28].

$$\log \left( \frac{Mt}{M_o} \right) = \log (k) + n \log (t)$$ (2)

Where $M_t$ is the percentage of water uptake at given test time and $M_o$ is the percentage of water uptake at the equilibrium point. The coefficient $k$ and $n$ were computed from slope of curve and intercept of the log plot of $\left( \frac{Mt}{M_o} \right)$ against time (h) [29].

Typically, at early stages $M_t/M_o \leq 0.5$, the diffusion phenomenon was demonstrated as formulation (3) [29].

$$\frac{Mt}{M_o} = 4 \left( \frac{Dt}{\pi L^2} \right)^{1/2}$$ (3)

Where $L$ is the specimen thickness and $D$ is the diffusion coefficient.

3. Results and Discussion

3.1. Chemical modification of durian rind-derived cellulose

The untreated and treated cellulose show a considerable difference in IR spectra as seen in figure 3. Both samples presented the hydrogen bonded -OH stretching at 4000-3200 cm$^{-1}$, the -CH stretching at 3000-2850 cm$^{-1}$, and the -OH bending at 1638 cm$^{-1}$ attributed to adsorbed water [19, 30]. These bands related to cellulose characteristic [19, 30]. However, the result was found that the peak related to –OH group of APS-treated cellulose was lower in intensity since available-OH groups from cellulose bonded with APS. The comparison of obtained spectra gave a clear confirmation of the effectiveness of surfaces modification using APS. The amine groups were found in the APS-treated cellulose at the peak of 1562 cm$^{-1}$ due to hydroxyl groups of both silanol and cellulose being bonded with these amine groups from APS [12, 19, 31]. After cellulose was modified with APS, the highest peak at 1030 cm$^{-1}$ was observed which indicated the Si-O-Si or Si-O-cellulose was investigated [32]. These linkages are generally present in the 1200-1000 cm$^{-1}$ region [6, 12, 19, 33]. The peak showed more apparent because of the Si-O-Si bond lapped with the C-O stretching from cellulose structure [17]. The additional peaks at 852 cm$^{-1}$ and 802 cm$^{-1}$ were observed after the modification of cellulose using APS. These peaks are indicated to the Si-O-C bond [6] and Si-C stretching [8] respectively. In a comparison of intensity of peak at 1638 cm$^{-1}$, which is related to absorbed water [19], it was found that the intensity decreased after APS treatment. This result concluded that APS grafting onto the surface of cellulose is advantageous as APS is completely combined with the hydroxyl group of cellulose and resulting in reducing water uptake into cellulose.
3.2. Mechanical properties of biocomposites

In our previous research, we found that the optimal condition considering the mechanical properties was found to be at cellulose loading, mixing temperature, and mixing time of 35 wt%, 165 °C, and 15 mins with a desirability of 94.16%. At this optimal condition, the tensile strength was found to be 46.207 MPa, which was lower than PLA [34] because of the difference of the surface between cellulose and PLA matrix [17, 19, 20]. To improve mechanical properties of biocomposites, this research used 3-aminopropyltriethoxysilane (APS) to modify the surface of cellulose. The results are presented in table 1.

Table 1. Tensile properties of PLA and biocomposites.

| Sample     | Tensile strength (MPa) | Tensile strength change (%) | Modulus of Young (GPa) | Modulus of Young change (%) | Elongation at break (%) | Elongation at break changes (%) |
|------------|------------------------|-----------------------------|------------------------|----------------------------|-------------------------|--------------------------------|
| PLA        | 52.42±1.05             |                             | 3.61±0.05              |                            | 3.65±0.37               |                                |
| PLA-UC     | 47.04±2.02             | -10.26                      | 4.84±0.43              | +25.41                     | 2.58±0.40               | -29.32                         |
| PLA-TC     | 55.37±0.94             | +5.63                       | 6.40±0.22              | +77.29                     | 2.44±0.19               | -33.15                         |

*UC means untreated cellulose and TC means APS-treated cellulose.

Changes of properties were compared with neat PLA.

The neat PLA had tensile strength of 52.42 MPa. With the addition of 35 wt% untreated cellulose, it was shown that tensile strength decreased to 47.04 MPa. The biocomposites reinforced with 35 wt% treated cellulose exhibited an increase of tensile strength to 55.37 MPa, or 5.63%, compared with neat PLA, which indicated that the treatment led to a better load transfer and homogenization of biocomposites microstructure, providing for better continuity and a stronger interface between the matrix and filler. The similar results in PLA/flax composites were found by Le Moigne et al. [33].
Modulus of Young for PLA is 3.61 GPa. The Modulus of Young of biocomposites reinforced increased to 4.84 GPa and 6.40 GPa, respectively. Way et al. [35] have suggested that the modulus of Young of biocomposites increased due to the modulus of fiber being much higher than PLA. The use of 3-aminopropyltriethoxysilane (APS) provides the hydrophobicity of the cellulose fiber surface by the presences of a high polar amine group. Its polar amine group toward the fiber surface forms strong hydrogen bonds, and thus, the configuration leaves the hydrophobic methylene group exposed at the surface [17]. Therefore, adhesion between the hydrophobic matrix surface and hydrophilic fiber surface can be improved via APS treatment. The improved adhesion reduces the deformation of elastic zone of matrix, then resulting in increase of the modulus of Young [31, 35].

Comparison with neat PLA, the elongation at break reduced with continuous adding cellulose. However, the results present that elongation at break remained unaffected by silane treatment. Some researchers have obtained the similar results; composites based on PLA and flax [36], PLA and jute fiber [37], and PLA reinforced with lysine-diisocyanate treated by bamboo fiber [38]. Therefore, two main factors affecting the elongation at break are homogenization of fiber phase and matrix phase [37] and fiber dispersion ability [38]. Basically, the elongation at break of material related to the strain property. Therefore, the decrease of elongation at break by adding of cellulose indicated that the strain of biocomposites was lower than neat PLA [35]. Generally, the incorporation of higher amount of fiber makes fiber agglomeration and then restricts the mobility of macromolecules resulting in lower resistance to breaking [35, 39]. Therefore, the elongation at break of fiber reinforced PLA biocomposites decreases.

3.3. Thermal Properties

The DSC curves of PLA and both biocomposites (PLA-untreated cellulose and PLA-treated cellulose) are shown in figure 4. The detailed data; the glass transition temperature ($T_g$), crystallization temperature ($T_c$), melting temperature ($T_m$), enthalpy of fusion ($\Delta H_m$), enthalpy of crystallization ($\Delta H_c$) and degree of crystallinity ($X_c$) are summarized in table 2. $T_g$ was reported as the temperature at the midpoint while $T_c$ and $T_m$ were reported as the maximum peak.

![Figure 4. DSC curves of PLA and biocomposites.](image_url)

The $T_g$ and $T_m$ of neat PLA were 56.96 °C and 156.26 °C, respectively. The addition of 35 wt.% untreated and treated cellulose affecting the $T_g$ decreased slightly to 56.20 °C and 55.97 °C, respectively, whereas the $T_m$ increased slightly to 157.59 °C and 158.14 °C, respectively. The similar results were
demonstrated by González et al. [24] and Lee et al. [40]. They found that adding kenaf fiber reduced the \( T_g \) and increased the \( T_m \) with comparison with neat PLA.

Table 2. DSC results of PLA and biocomposites.

| Specimens | \( T_g \) (°C) | \( T_c \) (°C) | \( T_m \) (°C) | \( \Delta H_m \) (J/g) | \( \Delta H_c \) (J/g) | \( X_c \) (%) |
|-----------|---------------|---------------|---------------|----------------------|----------------------|------------|
| PLA       | 56.96         | 109.74        | 156.26        | 18.01                | 14.63                | 15.73      |
| PLA-UC    | 56.20         | 101.24        | 157.59        | 20.31                | 20.86                | 22.43      |
| PLA-TC    | 55.97         | 96.75         | 158.14        | 23.34                | 15.94                | 17.14      |

*UC means untreated cellulose and TC means APS-treated cellulose.

The PLA had a \( T_c \) of 109.74 °C. The \( T_c \) of untreated and treated cellulose reinforce PLA biocomposites decreased to 101.24 °C and 96.75 °C, respectively. Based on infinite size, PLA crystal has melting enthalpy of 93 J/g, therefore \( \Delta H_c / 93 \) was used to calculate \( X_c \). The both biocomposites showed higher \( X_c \) than neat PLA as seen in table 2. The \( X_c \) of PLA was 15.73% whereas the \( X_c \) of untreated and treated cellulose reinforced PLA biocomposites increased to 22.43% and 17.14%, respectively. The decrease in \( T_c \) and increase in crystallinity confirmed the previous observations related to the role of cellulose as nucleating agents [2, 40, 42]. In terms of the crystallization behavior, the results showed significant effects of APS treatment on cellulose as a reinforcing material. The DSC curves demonstrated that both biocomposites showed dual melting peaks. This phenomenon may be attributed to lamellae rearrangement, resulting in imperfect crystal formation on melting endotherms of PLA [35]. Generally, the polymer is always melted to a slightly above \( T_g \). Therefore, the drop in \( T_g \) could be an advantage for packaging industry that involves at lower temperature for sheet thermoforming to avoid thermal degradation [6].

3.4. Fracture morphologies of biocomposites

Figure 5. SEM of cellulose-PLA biocomposites (a) untreated cellulose and (b) APS-treated cellulose.

The coupling agent is needed to improve the surface of cellulosic filler and enhance chemical reaction with the polymer matrix. Moreover, it was found that coupling agent provides well-disperse of the natural fiber in the matrix phase[40]. Figure 5 shows the scanned fractured surfaces of biocomposites by SEM. Many holes were observed after the cellulose was pulled out from the PLA matrix as shown in figure 5(a). Meanwhile, APS-treated cellulose-PLA biocomposites showing better adhesion exist at interphase as
APS-treated cellulose still embedded in the matrix as seen in figure 5(b). Besides, fewer cavities present on the surface and the pull out of filler from PLA matrix decreased. Similar results were obtained by other authors; silane-modified bamboo fiber and natural rubber composites [39], maleic anhydride (MA) grafted PLA and wood composited [43].

3.5. Water absorption behavior

The major disadvantage of the natural fiber reinforced materials is their hygroscopic behavior which is extremely responsive to water, affecting their mechanical properties which in return makes them perform poorly [17, 26]. Figure 6 represents the percentage of water uptake against soaking time. The result demonstrated that the water uptake by PLA was less than 1% with the maximum of 0.52%. Both biocomposites absorbed water rapidly in the first 15 days of soaking. After that, the curves showed a gradual increase and finally reached an equilibrium point, where the weight of biocomposites remained constant. However, treatment of the 35 wt.% biocomposites with APS displayed reduced water absorption compared with the untreated cellulose biocomposites. The maximum water absorptions of untreated and APS-treated cellulose-PLA biocomposites were 5.91% and 4.05%, respectively.

![Figure 6. Water absorption curves of PLA and biocomposites.](image)

Similar results have been observed by other authors who found that water was absorbed by PLA up to 1% [26, 38, 44]. Meanwhile, water absorbed by biocomposites increased with the addition of cellulose and their values were very high compared with pure PLA, due to the number of hydroxyl group in the biocomposites increasing [25, 26]. Our previous paper found that after mercerization with an alkali solution, durian cellulose demonstrated more –OH groups than untreated durian rind [14]. The alkali treatment increased the accessibility of free hydroxyl groups, resulting in their better hydrophilic characteristics [26, 45]. It then became the cause for water uptake in biocomposites affecting their physical, mechanical and thermal properties. Therefore, moisture uptake due to their hygroscopic property is the important factor when using natural fiber as filler in composite system. It is important that this problem be addressed so that natural fiber may be considered as a viable reinforcement in composite materials [46].

Silane is known as a commercial coupling agent available in large-scale within a large number of functional groups, such as the amine group forming hydrogen bonding to COO-site on PLA structure [22, 24]. Additionally, alkoxsilane groups react to hydroxyl groups in cellulosic material. Thus, adding treated cellulose with a coupling agent can reduce water uptake of biocomposites. Such results include the lysine-
based diisocyanate modified bamboo fiber as filler in composite based on poly (lactic acid) (PLA) and poly (butylene succinate) (PBS)[38], 3-glycidoxypropyl trimethoxysilane in biocomposites of kenaf fibers in polylactide [40], and silane treated oil palm – derived fiber reinforced PLA and polycaprolactone (PCL) composites [47].

Figure 7 presents the diffusion curves fitting of PLA and biocomposites. The coefficient k and n were computed as formulation (2) and were summarized in table 3.

![Figure 7. Diffusion curves fitting of PLA and biocomposites.](image)

In general, three known mechanisms of diffusion behavior are Fickian diffusion with n=0.5, relaxation controlled with n > 0.5, and non-Fickian or anomalous with 0.5 < n < 1[29, 48, 49]. All formulations had n-values less than 0.5. Thus, the diffusion behaviors of all formulations corresponded to the Fickian model. The diffusion coefficient is the most significant parameter of the Fickian model. This value indicates the penetration efficacy of water into the composite structures [28].

**Table 3.** Water absorption behavior parameters of PLA and biocomposites.

| Sample     | n     | k (h^{-1}) | Maximum water absorption (%) | Water diffusion coefficient; D (\times 10^{-13} \text{m}^2\text{s}^{-1}) |
|------------|-------|------------|-----------------------------|--------------------------------------------------------------------------------|
| PLA        | 0.4492 | 0.0510     | 0.52                        | 7.0269                                                                         |
| PLA-UC     | 0.5034 | 0.0472     | 5.91                        | 12.6607                                                                        |
| PLA-TC     | 0.4983 | 0.0497     | 4.05                        | 10.6790                                                                        |

*UC means untreated cellulose and TC means APS-treated cellulose.

Although many advantages of natural fiber have been reported; low density, innocuousness, high stiffness, renewable and biodegradable [4], however, both biocomposites displayed disadvantages of higher water diffusion coefficient than neat PLA. The immiscibility due to its poor interaction of the natural fiber and the matrix phase is a major limitation [17]. This limitation makes water penetrate into the gaps at the interfaces [50, 51]. However, the results also demonstrated that the biocomposites prepared from APS-treated cellulose had a lower water diffusion coefficient compared with untreated cellulose biocomposites. The similar result was also found in modified empty fruit bund oil palm fiber reinforced
polypropylene (PP) biocomposites [53]. Thusly, it can be concluded that the gaps due to its poor interaction between cellulose and PLA were successfully narrowed using the silane coupling agent.

4. Conclusion

Cellulose from durian rinds was successfully modified with 3-aminopropyltriethoxysilane (APS). The surface modification was confirmed via FTIR. From the above experimental results and analysis, it could be realized that the tensile strength and the modulus of Young of cellulose reinforced PLA biocomposites were improved. The improvement was attributed to better interfacial between filler and matrix as given morphological feature leading to lower water uptake into biocomposites. In addition, it could be summarized that cellulose showed the role of nucleating agent in PLA to generate crystallization with lower temperature as investigated in DSC curves.

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