



Structure and properties of poly (lactic acid)/*Sterculia urens* uniaxial fabric biocomposites



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ABSTRACT

Uniaxial cellulose fabric *Sterculia urens* reinforced poly (lactic acid) (PLA) matrix biocomposites were prepared by a two-roll mill. In order to assess the suitability of *Sterculia* fabric as reinforcement for PLA matrix, the PLA/*Sterculia* fabric biocomposites were prepared. Tensile parameters, such as maximum stress, Young's modulus and elongation-at-break, were determined using the Universal Testing Machine. The effect of alkali treatment and silane-coupling agent on the tensile properties of PLA-based biocomposites was studied. The results of thermogravimetric analysis show that uniaxial treatment of the fabric can improve the degradation temperature of the biocomposites. Moreover, morphological studies by scanning electron microscopy confirmed that better adhesion between the uniaxial fabric and the matrix was achieved. It was established that standard PLA resins are suitable for the manufacture of *S. urens* uniaxial fabric reinforced biocomposites with excellent engineering properties, useful for food packaging.

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1. Introduction

Nowadays, there has been a dramatic increase in environmentally friendly products and technologies. Biocomposites, where either the reinforcing cellulose fibre or matrix (or ideally both), are derived from natural and renewable resources, have a significant role to play in the next generation of sustainable materials. These materials help reduce fossil CO₂ emission, preserve non-renewable resources, increase sustainability and in most cases, can be biodegradable. Due to their outstanding properties, such as low cost, ease of processing, light weight, low maintenance cost etc., plastics in general and polymer composites in particular, find immense applications in fields such as automotive, aerospace, transportation, sports, agriculture etc. Unfortunately, plastics and composites made from synthetic and non-degradable polymers, pose severe environmental problems in their disposal. Hence,

polymer scientists and engineers, all over the world, now focus on the development of environmentally friendly plastics and biocomposites with balanced properties (Cheung, Ho, Lau, Cardona, & Hui, 2009; Mohanty, Misra, & Drzal, 2002; Mukherjee & Kao, 2011; Omar Faruka, Andrzej, Bledzka, Hans-Peter Fink, & Mohini, 2012; Satyanarayana, Arizaga, & Wypych, 2009).

In recent years, because of their outstanding properties, the usage of biodegradable polymer-based biocomposites is increasing rapidly. The performance of a polymer biocomposite depends not only on the selection of their components, but also on the interfacial interactions between matrix and filler. In order to meet specific needs, it is sometimes necessary to modify the matrix and the reinforcement. Reinforcement of biopolymer, with natural fibres, is the future of “green composites” that will address many sustainability issues. Amongst the available biopolymer, PLA is the most extensively used natural resource-based polymer produced at a large scale of over 140,000 tonnes per year (Shen, Worrell, & Patel, 2009). PLA, a biodegradable thermoplastic polymer with high strength and modulus, is a perfect eco-friendly material for many studies during the past decade. This is because of its potential in renewable engineering materials and food packaging (Nampoothiri, Rajendran Nair, & John, 2010). PLA provides good aesthetics and easy processability in most equipment (Lim, Auras, & Rubino, 2008). In

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processing and manufacturing, the price and inherent brittleness are the limitations for its wide practical applications. In the last decade, natural fibre reinforced PLA-based biocomposites have been widely investigated by the polymer scientists in an effort to make them to compete with non-renewable petroleum-based products. The type of fabric/fibres used plays an important role in the fibre/matrix adhesion and thereby affects the mechanical performance of the biocomposites. In order to make the composites environmentally friendly, natural fibres/fabrics can be used as the reinforcement (Huda, Drzal, Mohanty, & Misra, 2007; Jayaramudu, Jeevan Prasad Reddy, Babu Rao, & Varada Rajulu, 2009; Ku, Wang, Pattarachaiyakooop, & Trada, 2011; Lee & Wang, 2005; Oksman, Skrifvars, & Selin, 2003; Petersen et al., 1999; Rowell, 2001; Tokoro et al., 2008; Xu, Wang, Teng, & Yu, 2008; Zhong, Li, Yu, & Tan, 2011). David, Tom, Walther, and Lotte (2003) determined the mechanical properties of the PLA/jute fibre composites and reported that the tensile strength of composites was significantly higher than those of neat PLA. But the elongation-at-break of the composites is still very low (at about 2%). Masud, Lawrence, Amar, and Misra (2008) studied the effect of fibre surface-treatment on the properties of laminated PLA/kenaf fibre composites. The results show that silane-treated fibres reinforced composite and alkali-treated fibre reinforced composite offered superior mechanical properties when compared to that without fibre treatment. Hu and Lim (2007) reported that alkali treatment significantly improved the tensile properties of hemp fibre reinforced PLA when compared to those untreated. The composites with 40% volume fraction of alkali-treated fibre have the best tensile properties. The tensile strength and tensile modulus of the composites with 40% treated fibre are 54.6 MPa and 85 GPa respectively, which are much higher than neat PLA, especially for the tensile modulus which is more than twice that of neat PLA (35 GPa). Researchers have attempted various surface treatments (Gassan & Bledzki, 1999) of which alkali treatment using sodium hydroxide (NaOH) has been found to be the most feasible (Ray, Sarkar, Basak, & Rana, 2004). Huda, Drzal, Mohanty, and Misra (2008) treated kenaf fibre with NaOH in order to produce kenaf fibre reinforced PLA composites, using film stacking. They reported a 34% increase in flexural strength, 69% increase of flexural modulus and a 50% increase in impact strength (IS) when compared to the composites with untreated kenaf fibre. Khan and Hassan (2006) studied the effect of amino-based silane coupling agent on the performance of jute polycarbonate composites. Using Fourier transform infrared (FTIR) spectra, they confirmed the formation of Si–OH bonds between the fibres and the coupling agent.

The mechanical properties of composites depend on the orientation of the reinforcement towards the stress direction. If the reinforcement is composed of uniaxial fibres, then their orientation can be achieved accurately with ease. Rarely, some natural fabrics exist with the fibres arranged in a uniaxial fashion. Recently, new uniaxial fabrics *Grewia tilifolia* (Jayaramudu, Guduri, & Varada Rajulu, 2010), *Cordia dichotoma* (Jayaramudu et al., 2011) and *Polyalthia cerasoides* (Jayaramudu, Jeevan Prasad Reddy, Guduri, & Varada Rajulu, 2009) are some of the examples of uniaxial fabrics that were identified. Of recent, new uniaxial fabric, *Sterculia urens* was identified and its properties studied (Jayaramudu, Guduri, & Varada Rajulu, 2009).

The aim of the present work is to report the preparation and characterization of new types of environmentally friendly biocomposites. In the present paper, PLA, a biodegradable polymer was used as the matrix material and biodegradable uniaxial natural fabric *S. urens* as reinforcing material. The uniaxial fabric *S. urens* reinforced PLA biocomposites were prepared by a two-roll mill. The effect of surface-treatment method on untreated, alkali-treated and silane-coupling agent-treated indicated significant changes in the morphological characteristics of the biocomposites. The effect

of fabric surface-treatments on the mechanical and thermal properties of the biocomposites were studied, in order to ascertain whether the PLA and *S. urens* fabric system could effectively be used for making biocomposites suitable for packaging applications.

2. Experimental

2.1. Materials

The PLA used in the study is a commercial grade (PLA 2002D), obtained from Nature works LLC, USA. It had a D-isomer content of about 4%; weight average molecular weight, $M_w = 235$ kg/mol; density = 1.24 g/cm³; glass transition temperature, $T_g \approx 60^\circ\text{C}$; and melting temperature, $T_m \approx 153^\circ\text{C}$. Before use, PLA was dried at 60°C under a vacuum for 36 h. The chemicals: sodium hydroxide (NaOH) and coupling agent ((3-aminopropyl)triethoxysilane) (CA) were obtained from Merck. Natural fabric *S. urens* with a thickness of 0.16 mm was used as the reinforcement. In order to protect the tree, the fabric was separated from its branches only as described elsewhere (Jayaramudu, Guduri, et al., 2009). The extracted fabric was washed thoroughly in distilled water, and allowed to dry for seven days in the sun. This fabric was dried for one day in a vacuum oven, prior to using it as reinforcement.

2.2. Surface treatment with alkali and silane-coupling agent (SCA)

Uniaxial fabric *S. urens* was treated with 5% aq sodium hydroxide (NaOH) solution at room temperature, maintaining a liquor ratio of 20:1. Fabrics were immersed in the alkali solution for 30 min in order to remove the hemicellulose and other greasy materials. Then, the fabrics were washed repeatedly with water in order to neutralize NaOH. Finally, the fabrics were washed with distilled water and dried (Jayaramudu, Guduri, et al., 2009). A portion of the dried alkali-treated uniaxial fabric *S. urens* was treated with silane-coupling agent solution (diluted to 1% using acetone). Fabrics were dipped in the silane-coupling agent for 6 h and dried in hot air oven at 60°C for 1 h. Then, uniaxial fabric *S. urens* was cut to an average length of 5 ± 3 mm. Then two types of fabrics surface-treatment methods were used to improve the interfacial bonding properties between *S. urens* fabric and PLA matrix.

2.3. Composites preparation

The uniaxial fabric *S. urens* and PLA were blended using a two-roll plastics mill at 160°C for 10 min and the volume percent of the uniaxial fabric *S. urens* was 20%. The composites obtained were then moulded into sheets by hot pressing at 180°C and 20 MPa for 4 min, followed by cooling to room temperature at 5 MPa (Tao, Yan, & Jie, 2009). The sheets were prepared for structural characterization thermal and tensile properties measurements.

2.4. FTIR spectral analysis

Untreated, alkali-treated and silane-coupling agent treated fabric samples were cryogenically cooled and powdered. This powder was mixed with KBr and pellets were prepared by employing a hydraulic press. The FTIR spectra of these samples were recorded in the $4000\text{--}500$ cm^{−1} region, on a Perkin Elmer 16PC FT-IR instrument with 32 scans in each case, at a resolution of 4 cm^{−1}.

2.5. Microscopic analysis

Surface morphologies of the fabric and the tensile fabric pull-out from the biocomposites were examined using a Scanning Electron Microscope (SEM) Model JSM 820 microscope (Akishima, Japan).

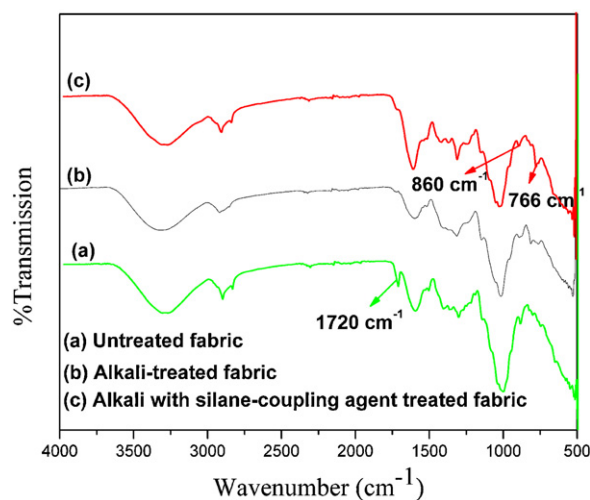


Fig. 1. FTIR spectra of the composites with (a) untreated, (b) alkali-treated and (c) alkali with silane-coupling agent treated. (For interpretation of the references to color in figure legend, the reader is referred to the web version of the article.)

The SEM fractured surfaces were prepared in a cryo-break fracture (the “dog-bone” test specimens were stored in liquid nitrogen for at least 10 min, then removed and immediately broken). The samples were gold-coated by an electro deposition technique in order to impart electrical conduction before SEM studies. The optical micrographs of the fabrics were recorded using a Leica DMLP polarized optical microscope.

2.6. Thermogravimetric analysis (TGA)

Thermogravimetric analysis (TGA) was performed on Q100 thermogravimetric analyzer (Tainstsh, USA) at a heating rate of 20 °C/min. Samples were examined under nitrogen (80 ml/min), over a temperature range from ambient to 600 °C.

2.7. Tensile properties

Tensile stress and tensile modulus were determined using Instron 3369 Model UTM. The crosshead speed for tensile test was maintained at 5 mm/min. In each case, 10 samples were tested and the average is reported.

3. Results and discussion

3.1. Functional group analysis

To investigate the structural changes in the *S. urens* fabric of the untreated, alkali treated and treated with silane-coupling agent samples were characterized by FTIR spectrometer. The spectra of

untreated fabric (neat) and alkali-treated fabric in the presence of silane-coupling agent, show similar bands (corresponding to hemicellulose, lignin and cellulose) except for the disappearance of the band at 1720 cm⁻¹ when the fabric was treated with alkali. This band corresponds to the C=O stretching of the hemicellulose present in the untreated fabric. The disappearance of C=O stretching indicates that the hemicellulose has been removed after alkali treatment of the fabric. Furthermore, no appreciable changes in the spectra of the fabric in the presence of coupling agent were noticed, however the silane-coupling agent treated *S. urens* fabrics reveal absorption peaks at around 766 cm⁻¹ (refer to Fig. 1), which is attributed to a Si–C stretching bond (Ishida & Koeing, 1976). Again, the band around 860 cm⁻¹ corresponds to the Si–OH bond which confirms the fact that –OH group of silane reacts with cellulose. Similar observation was made by Khan and Hassan (2006) in the case of jute fibres. The bands at 860 cm⁻¹ appeared very weak due to the fact that the spectra were taken for bulk samples, whereas the silane coupling agent used was only 1% and it reacts only with groups on the surface of the fabric.

3.2. Surface analysis of the fabric

3.2.1. Morphological analysis

The SEM images and optical photographs of the untreated, alkali treated and silane-coupling agent treated uniaxial fabric *S. urens* are shown in Figs. 2 and 3 respectively. From Fig. 2(a)–(c), it can be observed from the micrographs that the fibres in the fabric became thin due to the removal of alkali soluble components and many shallow grooves were formed on the surface when silane-coupling agent was used. Jayaramudu, Guduri, et al. (2009) previously reported that hemicellulose of *S. urens* fabric was eliminated, following alkali treatment. Rout, Tripathy, Nayak, Misra, and Mohanty (2001) reported SEM studies of the alkali-treated coir fibres. They reported the progressive changes in the surface of coir fibres by SEM due to the removal of greasy material from the surface, as a result of alkali treatment. This resulted in the fabric surface becoming rough following on alkali treatment. In addition, structural changes in the fabric due to, alkali and silane-coupling agent treatments, is confirmed by the FTIR spectra (refer to Fig. 1).

Polarized optical photographs (POM) of *S. urens* fabrics: untreated, alkali treated and silane-coupling agent-treated is respectively presented in Fig. 3(a)–(c). POM image of the untreated fabric (Fig. 3(a)) was found to be diffuse. This may be the result of the presence of amorphous hemicellulose layer on the surface of the fabric. However, the photographs (Fig. 3(b)) of the alkali-treated fabric are found to be sharp. This is because of the elimination of the hemicellulose content upon alkali treatment. From the photographs (Fig. 3(c)), it is clear that fabrics treated with silane-coupling agent showed very thin and rough surfaces. Similarly, the bonding was strong when alkali treatment with coupling agent was employed. The results of FTIR (refer to Fig. 1) prove that chemical bonding does exist, thereby improving the surface

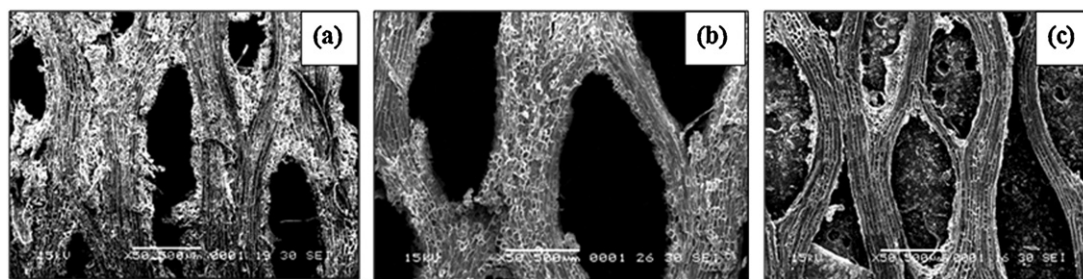


Fig. 2. Scanning electron micrographs of: (a) untreated, (b) alkali-treated and (c) alkali with silane-coupling agent treated used natural fabrics of *Sterculia urens* at same magnification.

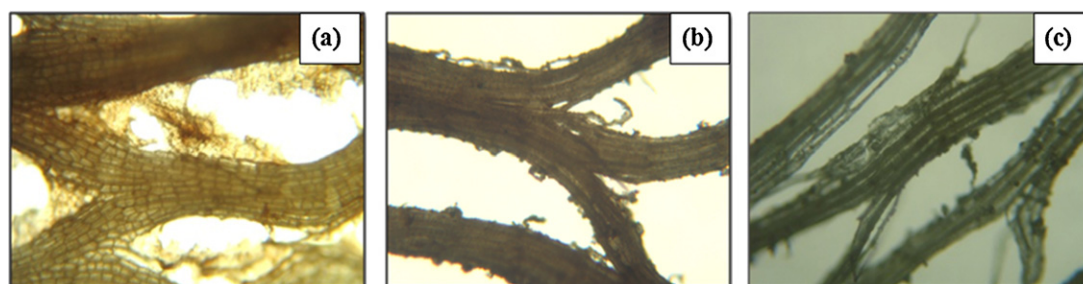


Fig. 3. Polarized optical photographs of *Sterculia urens* natural fabric: (a) untreated, (b) alkali treated, (c) alkali with silane-coupling agent treated.

interfacial bond strength between silane-treated fabric and the matrix (PLA).

3.3. Tensile properties

The tensile modulus, strength and elongation-at-break for different fabric contents of the PLA/*S. urens* fabric biocomposites, before and after alkali treatment and in the absence or presence of the silane-coupling agent, are presented in Table 1. For comparison, these values for the untreated and alkali-treated fabric are also presented in Table 1. In order to probe the effect of coupling agent on the properties of the neat and alkali treated fabrics, their tensile properties were also studied. From Table 1, it is evident that for the neat and alkali-treated fabrics, the silane-coupling agent had little effect on the maximum stress (it increased only from 10 to 11 and 18.9 to 20.34 MPa, respectively). This may be due to the fact that changes were taking place only on the surface and not in the bulk. Furthermore, for the neat and alkali-treated fabrics, the modulus was found to increase considerably, whereas the elongation-at-break decreased marginally when silane-coupling agent was used. This is due to the fact that as modulus is an initial property within the elastic limit, the slight increase in rigidity might have enhanced it. From Table 1, it is further evident that the tensile strength of the alkali-treated fabrics increased from 10.0 MPa (untreated) to 18.9 MPa on alkali treatment. Similarly, the Young's modulus increased from 640.7 MPa to 2018.6 MPa on alkali treatment. This increment in tensile properties is attributed to the elimination of the amorphous hemicellulose content on alkali treatment.

In the case of the PLA-biocomposites (untreated and alkali-treated fabrics), the tensile strength and the modulus also increased appreciably. For these biocomposites, alkali treatment improves the fabric surface adhesion characteristics by removing natural and artificial impurities. This is understandable as the weak amorphous hemi cellulose and lignin contents decrease on alkali treatment. Jayaramudu, Guduri, et al. (2009) already reported that the surface of these fabrics became rough and thin on alkali treatment, due to the elimination of hemicellulose and soluble lignin. The improvement of tensile strength and modulus is due to the improved fabric–matrix adhesion by the surface treatments. Natural and artificial impurities can be removed (the weak amorphous hemi

cellulose and lignin contents) and a rough surface morphology is obtained by surface treatments (refer to Fig. 4(b)). On the other hand, for the PLA-based biocomposites, when the silane-coupling agent was used, the modulus increased remarkably. Khan, Mina, and Drzal (2000) made a similar observation in the case of jute-polycarbonate composites. They reported that the tensile strength and modulus of jute-polycarbonate composites increased by 28% and 70%, respectively, when amino based silane-coupling agent was used. In the present study, a similar enhancement of 13% and 40% was observed when a silane-coupling agent was used. However, no significant change in elongation-at-break was observed for the PLA-based biocomposites. The improved bonding between the fabrics and PLA matrix by the silane-coupling agent may be responsible for this behaviour. PLA-biocomposites morphology plays an important role in improving the mechanical properties of the PLA biocomposites.

3.4. Morphology of the composites

The morphologies of uniaxial fabric *S. urens* PLA-based biocomposites (untreated, alkali treated and silane-coupling agent treated natural fabric *S. urens* PLA biocomposites) were studied. Interfacial studies were carried out in order to investigate the interaction between uniaxial fabric *S. urens*/matrix (PLA) by SEM studies. In general, there is clear pull out, detachment and debonding of some of the *S. urens*, which indicates a poor interfacial adhesion of the untreated *S. urens* with the PLA matrix.

It can be seen in Fig. 4(a) that the fabrics are detached from the matrix surface due to the poor interfacial bonding, splitting in the inner fabrics, fabrics breakage and void formation on the resin surface. Such an observation proves that there is a poor interaction between fabrics and matrix. According to Mohanty, Misra, and Drzal (2001), such waxy substance contributed to the ineffective fibre–matrix bonding and poor surface wet out. This is due to the presence of polar groups in the matrix and the cellulosic fabrics. It is an indication of crack deflection, where the crack path is changed by the fabrics and directed along the fabric surface. This leads to fabrics debonding, which is an indication of matrix separation around the fabrics as crack front intersects the PLA/*S. urens* fabrics interface. This clearly indicates that bonding is poor. In addition, empty

Table 1

Maximum stress, Young's modulus, and elongation-at-break of untreated, alkali-treated and coupling agent treated PLA/*Sterculia urens* natural fabric biocomposites.

<i>Sterculia urens</i> fabric	Maximum stress (MPa)		Young's modulus (MPa)		Elongation-at-break (%)	
	Without SCA	With SCA	Without SCA	With SCA	Without SCA	With SCA
Untreated	10.0	11.0	640.7	870.04	2.0	1.82
Alkali treated	18.9	20.34	2018.6	2693.3	2.4	2.29
PLA/untreated biocomposites	58.2	62.1	3180.9	4120.6	3.7	4.2
PLA/biocomposites treated by alkali	69.1	78.4	4207.5	5904.7	5.0	5.4

SCA = silane-coupling agent.

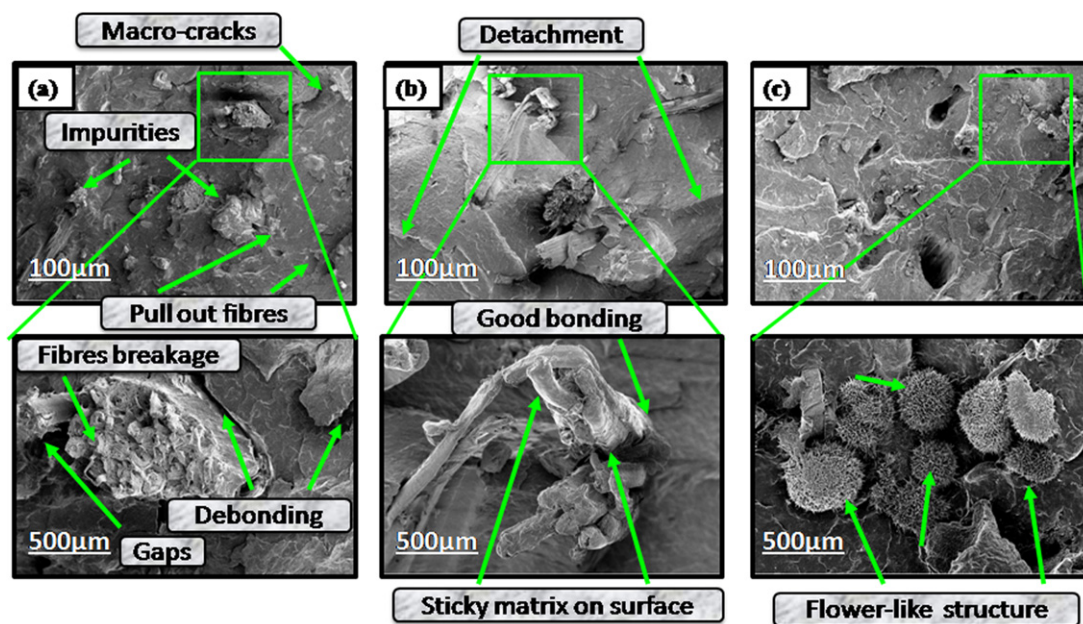


Fig. 4. Scanning electron micrograph of fractured surfaces (cross-section) of PLA/*Sterculia urens* composites: (a) PLA/untreated biocomposites; (b) PLA/alkali-treated biocomposites and (c) PLA/alkali with silane-coupling agent fabric biocomposites at different magnifications.

fabric where the matrix was not able to penetrate in the envelope of the fabric is observed, resulting in low mechanical properties (refer to Table 1). This confirms the explanation advanced earlier.

Furthermore, Fig. 4(b) shows the alkali-treated fabric biocomposites with almost no signs of fabric debonding, detachment or pull-out as the alkali-treated surface of the fabric became rough. This result suggests that interfacial adhesion between the *S. urens* fabric and PLA matrix has become highly favourable and strong, following treatment of fabrics with alkali (NaOH). These observations indicate that the bonding between the reinforcement and the matrix had partially improved when compared with untreated biocomposites. The elimination of the greasy material (waxy layer and impurities) in the *S. urens* fabric by alkali treatment led to increased roughness of fabric surface, as the hemi-cellulose and some part of the lignin were removed, as a result, the fabric became thin and then created a good interlocking mechanism with the surface of PLA matrix.

In Fig. 4(c), a better dispersion is observed in PLA/alkali-treated with silane-coupling agent fabric biocomposites, prepared by using the treated fabric with silane-coupling agents, respectively. The predominantly polar silane-coupling agent makes good dispersion with the polar fabric, due to the dipole–dipole attraction and hydrogen bonding, resulting in effective Van der Waals forces. More interestingly, fabric skin formation was observed on the surface of the matrix. These observations indicate that the bonding between the fabric and the matrix is good and it shows flower-like structure in some parts of the PLA/*S. urens* fabric composites surface, when pressed during sliding. This type of structure is hereby reported for the first time in natural fabric composites as far as we know. Similarly, the bonding was strong when alkali treatment with silane-coupling agent was employed. The predominantly polar nature of the silane-coupling agent is believed to be responsible for the improvement of the interfacial bonding in the composites. Furthermore, matrix skin formation was observed on the surface of the fabrics. These observations indicate that the bonding between the fabric and the matrix is good. This confirms the findings of [Colom, Carrasco, Pagcs, and Canavate \(2003\)](#) who studied the effect of different chemical modifications on the interfacial characteristics of aspen fibre-reinforced high-density polyethylene composites. They observed that silane was a better coupling agent than epolene. This

was attributed to the fact that in composites modified with epolene, adhesion occurs due to multiple mechanisms of adsorption and inter diffusion, whereas the composites treated with silane showed a chemical mechanism of adhesion with the formation of covalent bonds, in addition to hydrogen bridges which would lead to greater adhesion between the fibres and the matrix. Similarly, the bonding was strong when alkali treatment with silane-coupling agent was employed. The predominantly polar nature of the silane-coupling agent is believed to be responsible for the improvement of the interfacial bonding in composites. This observation gives direct evidence about the adhesion improvement at the interface in the presence of reactive silane-coupling agent. These arguments are reflected in all the mechanical properties investigated. In good agreement with SEM study, the mechanical properties of PLA/*S. urens* fabric biocomposites prepared using silane-coupling agents are significantly higher than the control, untreated *S. urens* fabric PLA matrix biocomposites (refer to Table 1).

3.5. Thermogravimetric analysis

The thermal stabilities of neat PLA and PLA-based biocomposites were investigated for the weight loss as a function of temperature by thermogravimetric analysis under nitrogen flow. The TGA curves shown in Fig. 5 reveal the thermal stability of neat PLA, *S. urens* uniaxial fabric reinforced PLA biocomposites. From the TGA thermograms of neat PLA, PLA biocomposites and its fabric treated composites (as seen in Fig. 5), indicated that neat PLA degraded at around 300 °C. Generally, *S. urens* fabrics degrade through three main stages: the first stage below 150 °C is mainly due to the removal of the moisture absorbed, the second; thermal degradation occurs between 190 and 350 °C and is mainly related to the degradation of cellulosic substances such as: hemicellulose and cellulose and the third stage (350–500 °C) is attributed to the degradation of non-cellulosic materials in the fabric ([Huda et al., 2007](#)). As seen in Fig. 5, the biocomposites with neat PLA show lower degradation temperature than the untreated fabric biocomposites. This is due to the decrease in molecular weight of PLA when the composites are processed on a two-roll mill. The incorporation of *S. urens* fabrics in PLA the matrix has also affected the thermal degradation temperature. Thermal degradation temperature of the biocomposites

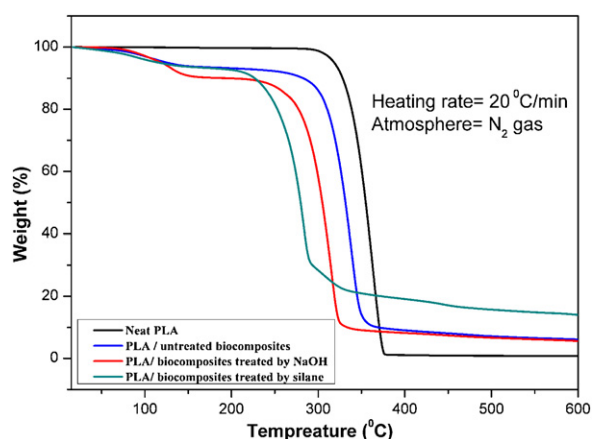


Fig. 5. Thermogravimetric curves of neat PLA and PLA-biocomposites.

with surface treatment increased when compared to that of the biocomposites without surface treatment (untreated fabric). The chemical bond between *S. urens* fabrics and PLA matrix enhanced the interfacial adhesion and consequently the thermal degradation temperature increased. Conversely, the thermal degradation temperature of the biocomposites with silane-coupling agent treated fabric is lower than that of the biocomposites with alkali-treated *S. urens* fabric. The result indicated that the stability of the interface bond between silane-coupling agent treated *S. urens* fabric and PLA matrix is worse than that of the interface bond between NaOH treated *S. urens* fabric and PLA matrix.

4. Conclusions

Environmentally friendly biocomposites of alkali and silane-coupling agent treated uniaxial fabric *S. urens* reinforced PLA matrix biocomposites were prepared on a two-roll mill. Results showed that surface treatment improves the compatibility between the PLA matrix and the reinforcing *S. urens* fabric. It is also observed that when the alkali-treated fabric and silane-coupling agent were used, the mechanical properties increased. The mechanical and thermal properties of the alkali-treated and treated with silane-coupling agent biocomposites are better than that of untreated fabric biocomposites and the neat PLA matrix. The thermal behaviours of PLA and PLA-based biocomposites, consequently, confirm the improvement in the thermal properties because of the improved adhesion between the fabrics and the agents (alkali and silane-coupling agent). SEM micrographs indicate that the bonding between the *S. urens* fabric and the matrix was enhanced by alkali treatment and silane-coupling agent. Alkali treatment in the presence of a silane-coupling agent, gives rise to matrix skin formation, flower-like structure on the surface of the fabric, indicating good bonding between the reinforcement and the matrix.

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References

Cheung, H. Y., Ho, M. P., Lau, K. T., Cardona, F., & Hui, D. (2009). Natural fibre-reinforced composites for bioengineering and environmental engineering applications. *Composites Part B: Engineering*, 40, 655–663.

- Colom, X., Carrasco, F., Pagcs, P., & Canavac, J. (2003). Effect of different treatments on the interface of HDPE/lignocellulosic fiber composites. *Composites Science and Technology*, 63, 161–169.
- David, P., Tom, L. A., Walther, B. P., & Lotte, N. (2003). Biodegradable composites based on L-poly(lactide) and jute fibres. *Composites Science and Technology*, 63, 1287–1296.
- Gassan, J., & Bledzki, A. K. (1999). Alkali treatment of jute fibers: Relationship between structure and mechanical properties. *Journal of Applied Polymer Science*, 71, 623–629.
- Hu, R., & Lim, J. K. (2007). Fabrication and mechanical properties of completely biodegradable hemp fibre reinforced poly(lactic acid) composites. *Journal of Composite Materials*, 41, 1655–1669.
- Huda, M. S., Drzal, L. T., Mohanty, A. K., & Misra, M. (2007). The effect of silane treated- and untreated talc on the mechanical and physicomechanical properties of poly(lactic acid)/newspring fibers/talc hybrid composites. *Composites Part B: Engineering*, 38, 367–379.
- Huda, M. S., Drzal, L. T., Mohanty, A. K., & Misra, M. (2008). Effect of fiber surface-treatments on the properties of laminated biocomposites from poly(lactic acid) (PLA) and kenaf fibers. *Composites Science and Technology*, 68, 424–432.
- Ishida, H., & Koeing, J. (1976). 31st annual tech. conf., Section 6-C Reinforced Plastic/Composites Institute, The Society of Plastic Industry Inc., (p. 17).
- Jayaramudu, J., Guduri, B. R., & Varada Rajulu, A. (2009). Characterization of natural fabric *Sterculia urens*. *International Journal of Polymer Analysis and Characterization*, 14, 115–125.
- Jayaramudu, J., Guduri, B. R., & Varada Rajulu, A. (2010). Characterization of new natural cellulosic fabric *Grewia tilifolia*. *Carbohydrate Polymers*, 79, 847–851.
- Jayaramudu, J., Jeevan Prasad Reddy, D., Babu Rao, G., & Varada Rajulu, A. (2009). Tensile properties of polycarbonate coated natural fabric *sterculia urens*: Effect of coupling agent. *Iranian Polymer Journal*, 18, 693–701.
- Jayaramudu, J., Jeevan Prasad Reddy, D., Guduri, B. R., & Varada Rajulu, A. (2009). Properties of natural fabric *Polyalthia cerasoides*. *Fibers and Polymers*, 10, 338–342.
- Jayaramudu, J., Maity, A., Sadiku, E. R., Guduri, B. R., Varada Rajulu, A., Ramana, C. H. V. V., et al. (2011). Structure and properties of new natural cellulose fabrics from *cordia dichotoma*. *Carbohydrate Polymers*, 86, 1623–1629.
- Khan, M. A., & Hassan, M. M. (2006). Effect of γ -aminopropyl trimethoxy silane on the performance of jute-polycarbonate composites. *Journal of Applied Polymer Science*, 100, 142–145.
- Khan, M. A., Mina, F., & Drzal, L. T. (2000). Influence of silane coupling agents of different functionalities on the performance of jute-polycarbonate composite. In *Proc 3rd international wood and natural fiber composites symposium* September 19–20, Kassel, Germany, (pp. 5–1–5–8).
- Ku, H., Wang, H., Pattarachaiyakop, N., & Trada, M. (2011). A review on the tensile properties of natural fiber reinforced polymer composites. *Composites Part B: Engineering*, 42, 856–873.
- Lee, S., & Wang, S. (2005). Biodegradable polymers/bamboo fiber biocomposite with bio-based coupling agent. *Composites Part A: Applied Science and Manufacturing*, 37, 80–91.
- Lim, L. T., Auras, R., & Rubino, M. (2008). Processing technologies for poly(lactic acid). *Progress in Polymer Science*, 33, 820–852.
- Masud, S. H., Lawrence, T. D., Amar, K. M., & Misra, M. (2008). Effect of surface-treatments on the properties of laminated biocomposites from poly(lactic acid) (PLA) and kenaf fibers. *Composites Science and Technology*, 68, 424–432.
- Mohanty, A. K., Misra, M., & Drzal, L. T. (2001). Surface modifications of natural fibers and performance of the resulting biocomposites: An overview. *Composite Interfaces*, 8, 313–343.
- Mohanty, A. K., Misra, M., & Drza, L. T. (2002). Sustainable biocomposites from renewable resources: Opportunities and challenges in green materials world. *Journal of Polymers and the Environment*, 10, 19–26.
- Mukherjee, T., & Kao, N. (2011). PLA based biopolymer reinforced with natural fibre: A review. *Journal of Polymers and the Environment*, 19, 714–725.
- Nampoothiri, K. M., Rajendran Nair, N., & John, R. P. (2010). An overview of the recent developments in polylactide (PLA) research. *Bioresource Technology*, 101, 8493–8501.
- Oksman, K., Skrifvars, M., & Selin, J. F. (2003). Natural fibers as reinforcement in polylactic acid (PLA) composites. *Composites Science and Technology*, 63, 1317–1324.
- Omar Faruka, D., Andrzej, K., Bledzka, C., Hans-Peter Fink, B., & Mohini, S. (2012). Biocomposites reinforced with natural fibers: 2000–2010. *Progress in Polymer Science*, 37, 1552–1596.
- Petersen, K., Nielsen, P. K., Bertelsen, G., Lawther, M., Olsen, M. B., & Nilsson, N. H. (1999). Potential of biobased materials for food packaging. *Trends in Food Science and Technology*, 10, 52–68.
- Ray, D., Sarkar, B. K., Basak, R. K., & Rana, A. K. (2004). Thermal behavior of vinyl ester resin matrix composites reinforced with alkali-treated jute fibers. *Journal of Applied Polymer Science*, 94, 123–129.
- Rout, J., Tripathy, S. S., Nayak, S. K., Misra, M., & Mohanty, A. K. (2001). Scanning electron microscopy study of chemically modified coir fibers. *Journal of Applied Polymer Science*, 79, 1169–1177.
- Rowell, R. M. (2001, December). *Proceeding international conference on science and technology of composite materials (COMAT)* Mar del Plata, Argentina.
- Satyanarayana, K. G., Arizaga, G. G. C., & Wypych, F. (2009). Biodegradable composites based on lignocellulosic fibersean overview. *Progress in Polymer Science*, 34, 982–1021.
- Shen, L., Worrell, E., & Patel, M. (2009). Present and future development in plastics from biomass. *Biofuels, Bioproducts and Biorefining*, 4, 25–40.

- Tao, Y. U., Yan, L. I., & Jie, R. E. N. (2009). Preparation and properties of short natural fiber reinforced poly(lactic acid) composites. *Transactions of Nonferrous Metals Society of China*, 19, s651–s655.
- Tokoro, R., Vu, D. M., Okubo, K., Tanaka, T., Fujii, T., & Fujiura, T. (2008). How to improve mechanical properties of poly lactic acid with bamboo fibers. *Journal of Materials Science*, 43, 775–787.
- Xu, H., Wang, L., Teng, C., & Yu, M. (2008). Biodegradable composites: Ramie fibre reinforced PLLA–PCL composite prepared by in situ polymerization process. *Polymer Bulletin*, 61, 663–670.
- Zhong, J., Li, H., Yu, J., & Tan, T. (2011). Effects of natural fiber surface modification on mechanical properties of poly (lactic acid) (PLA)/sweet sorghum fiber composites. *Polymer-Plastics Technology and Engineering*, 50, 1583–1589.