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"Green" composites from recycled cellulose and poly(lactic acid): Physico-mechanical and morphological properties evaluation

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"*Green*"/biobased composites were prepared from poly(lactic acid) (PLA) and recycled cellulose fibers (from newsprint) by extrusion followed by injection molding processing. The physico-mechanical and morphological properties of the composites were investigated as a function of varying amounts of cellulose fibers. Compared to the neat resin, the tensile and flexural moduli of the composites were significantly higher. This is due to higher modulus of the reinforcement added to the PLA matrix. Dynamic mechanical analysis (DMA) results also confirmed that the storage modulus of PLA increased on reinforcements with cellulose fibers indicating the stress transfers from the matrix resin to cellulose fiber. Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) showed that the presence of cellulose fibers did not significantly affect the crystallinity, or the thermal decomposition of PLA matrix up to 30 wt% cellulose fiber content. Overall it was concluded that recycled cellulose fibers from newsprint could be a potential reinforcement for the high performance biodegradable polymer composites.

1. Introduction

In recent years, the use of natural/bio-fiber reinforced composites has rapidly expanded due to the availability of natural/bio-fibers derived from annually renewable resources, for use as reinforcing fibers in both thermoplastic and thermosetting matrix composites as well as for the positive environmental benefits gained by such materials [1–3]. In addition, the low cost and high specific properties of these natural fibers provide significant performance with advantages and economic benefits when combined with commodity biodegradable polymers [2, 3]. Recently, poly(lactic acid) (PLA) has attracted increasing interest because this polymer can be manufactured from renewable resources, e.g. corn starch, to yield articles for use in textiles, industrial packaging or for biocompatible/bioabsorbable medical devices [2, 4]. PLA, the matrix resin in this study, is a thermoplastic that has high strength and modulus [5]. Research is also being conducted on the potential of cellulose-based fibers as reinforcement for polymers, because cellulose materials offer many potential advantages [6–8], e.g., abundant renewable source, inexpensive, low cost, flexibility during processing, desirable fiber aspect ratio, low density, high specific stiffness and strength, biodegradability, reduced wear of the processing machinery, etc. Moreover, considering the extensive intermolecular hydrogen-bonding potential resulting from the surface rich in hydroxyl groups, cellulose has the potential for string interaction with polar thermoplastics polymers [9]. However, as a result of their low thermal stability during processing, and poor dispersion in the

polymer melt, use of cellulose fibers in thermoplastics in general has not been extensive [10]. The mechanical properties of a composite material depend primarily on the strength and modulus of the fibers, the strength and the chemical stability of the matrix and the effectiveness of the bonding between matrix and fibers in transferring stress across the interface. Generally, the utilization of natural fibers as reinforcing materials in thermoplastics requires strong adhesion between the fiber and the polymer matrix. Cellulose has a strong hydrophilic character due to three hydroxyl groups per monomeric unit, but biopolymers like PLA and poly(hydroxy butyrate) (PHB) are generally hydrophobic.

Wood and cellulose fibers are attractive reinforcing fillers for thermoplastics, which can reduce material costs and improve certain properties [11, 12]. A review of the literature shows that a comparative study of thermal and mechanical properties of thermoplastic composites containing different wood species in the form of various pulps is limited [13–15]. Moreover, use of recycling paper can have a positive benefit on the environment. Newspaper is the most commonly recycled paper in residential programs. Each week the average household in the developed world generates more than 2 kg of newspaper, which can be a source for natural fibers as a basis for materials for construction, or to make new newsprint or paperboard packaging, or for manufacturing of insulation or other building materials [12, 13, 16]. More than 60% of the newspapers in the USA are recovered for recycling. All virgin paper is made from highly compressed and heated cellulose fibers from soft woods, specifically those grown and harvested as "paper pulp trees". It is now possible to purchase recycled newspaper as fiber forms. The purpose of this study is to investigate the properties and processing of recycled newspaper fiber forms as a possible reinforcement for "*green*" composites.

2. Experimental

2.1. Materials

Poly(lactic acid) (PLA; molecular weight, M_w = 20 kDa; M_n = 10.1 kDa) was obtained from Biomer, Krailling, Germany (product name—Biomer L 9000). CreaFill Fibers Corp., Chestertown, MD, supplied the cellulose fibers (CreaMix TC 1004, R 0083 and R 0084 fibers). R 0083 and R 0084 were made with the TC 1004 fibers (gray product). The only difference with TC 1004 fibers was the use of processing aid (e.g., polyester based lubricant) to make the R 0083 and R 0084 fibers. The fibers are reclaimed from newspaper/magazine or kraft paper stock. The average length of fibers is about 850 microns and the average width of the fibers is 20 microns. The high cellulose content (75% minimum) indicates that this is an alpha cellulose with maximum ash content of 23%. "Ash" is a combination of the carbon that is left after burning and other organics/non-organics (any clays, inks, lignin, tannins, extractives etc.) that are not volatilized after ignition. Moisture content of these fibers was less than 5%.

2.2. Composites processing

The polymer and cellulose fiber were dried in a vacuum oven at 80°C for 24 h before processing and mixed mechanically at 100 rpm with micro-compounding molding equipment (DSM Micro 15 $\times$ 10^{-6} m^3 compounding system, DSM research, The Netherlands) at 183°C for 10 min. The extruder is equipped with a screw of length 150 mm, L/D of 18. In order to obtain the desired specimen samples for various measurements and analysis, the molten composite samples were transferred after extrusion through a preheated cylinder to a mini injection molder.

2.3. Measurements

2.3.1. Mechanical testing

A mechanical testing machine, United Calibration Corp SFM 20, was used to measure the tensile properties according to ASTM D 638 standard, and the flexural properties according to ASTM D 790. System control and data analysis were preformed using Datum software. The Notched Izod impact strength was measured with a Testing Machines Inc. (TMI) 43–02–01 Monitor/Impact machine according to ASTM D256. All results presented are the average values of five measurements.

2.3.2. Differential scanning calorimeter (DSC)

The melting and crystallization behavior of the composites were studied using a differential scanning calorimeter (TA instruments 2920) equipped with a cooling attachment, under a nitrogen atmosphere. The data were collected by heating the composite specimen from 25°C to 200°C at a constant heating rate of 5°C/min.

2.3.3. Dynamic mechanical analysis (DMA)

The storage modulus, loss modulus, and loss factor (Tan Delta) of the composite specimen were measured as function of temperature (25 to 120°C) using a TA instruments 2980 Dynamic Mechanical Analyzer (DMA) at a frequency of 1 Hz and at constant heating rate of 5°C/min.

2.3.4. Heat defection temperature (HDT)

HDT measurements were obtained on flex bars of blends at 4.6 $\times$ 10^{-1} MPa load according to ASTM Standard D 648 deflection test using a TA instruments 2980 Dynamic Mechanical Analyzer (DMA) at a heating rate of 2°C/min.

2.3.5. Thermo gravimetric analysis (TGA)

Thermo gravimetric analysis was carried out by a TA thermo gravimetric analyzer (TA instruments 2950). The samples were scanned from 30 to 600°C at a heating rate of 20°C/min in the presence of nitrogen.

2.3.6. Scanning electron microscopy (SEM)

The morphology of impact fracture surfaces of the composites was observed by scanning electron microscope (SEM) at room temperature. A JEOL (model JSM-6300F) SEM with field emission gun and accelerating voltage of 10 kV was used to collect SEM images for the composite specimen. A gold coating of a few nanometers in thickness was coated on impact fracture

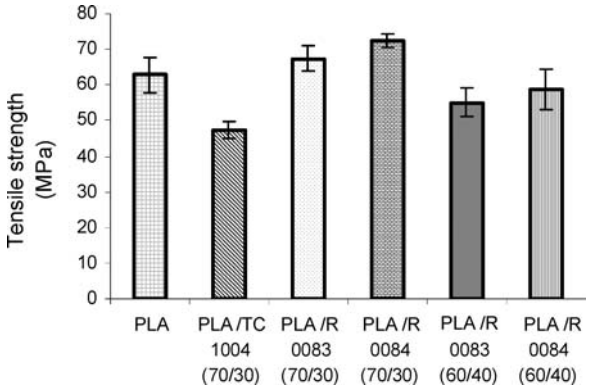

Figure 1 Tensile strength of the composites.

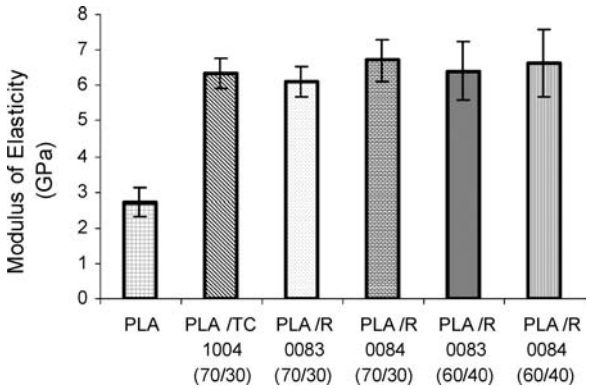

Figure 2 Tensile modulus of the composites.

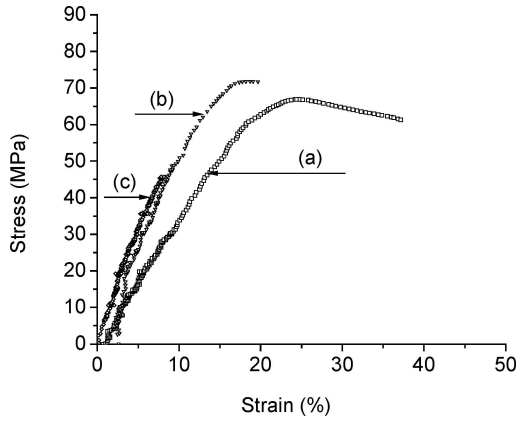

Figure 3 Stress-strain curves: (a) PLA/Cellulose (100/0) (∀), (b) PLA/R 8400 (70/30) (X), and (c) PLA/TC 1004 (70/30) (−).

surfaces. The samples were viewed perpendicular to the fractured surface.

3. Results and discussions

3.1. Mechanical properties

The tensile properties of the PLA matrix and PLA-cellulose based composites are shown in Figs 1 and 2. Comparison between the averaged tensile stress-strain curves of the PLA/TC 1004 and PLA/R 0084 (with 30 wt% fiber content) composites along with the PLA matrix alone is informative (Fig. 3). The pure PLA has a tensile strength of 62 MPa and a modulus of 2.7 GPa. As presented in Table I, the addition of cellulose fibers improved the modulus of PLA, which indicates that the stress transfers from the PLA polymer matrix to the stiffer fiber occurred [17]. Though the addition of TC 1004 fibers increased the modulus, the tensile strength of this PLA composite did not show any improvement. It can also be seen in Table I that the addition of R 0083 and R 0084 fibers (at 30% cellulose fiber content) improved both tensile strength and modulus of PLA. However at higher fiber content the tensile strength did not improve in either composites of R 0083 and R 0084. A possible explanation of this can be due to cellulose fiber agglomeration.

TABLE I Tensile properties of the PLA and PLA based composites

Polymer/cellulose (wt%)	Tensile strength (MPa)	Modulus of elasticity (GPa)	Improvement (modulus) (%)
PLA/Cellulose (100/0)	62.9 ± 4.9	2.7 ± 0.4	–
PLA/TC 1004 (70/30)	47.3 ± 2.5	6.3 ± 0.4	132
PLA/R 0083 (70/30)	67.4 ± 3.5	6.1 ± 0.4	124
PLA/R 0084 (70/30)	72.5 ± 2.0	6.7 ± 0.6	148
PLA/R 0083 (60/40)	55.0 ± 4.2	6.4 ± 0.8	136
PLA/R 0084 (60/40)	58.6 ± 5.8	6.6 ± 0.9	144

In Fig. 4, the dependence of tensile strength, tensile modulus, and impact strength on the composition of PLA-fiber composites is presented. The three parameters vary linearly with the fiber content in these composites [16]. As the amount of fibers in the composite increases, the modulus of the PLA increased proportionately [16]. The tensile strength decreases when the fiber content increases (Table II). This could be the result of less than adequate adhesion between the cellulose fibers and the PLA matrix. In addition, the pure PLA has an impact strength of 25 J/m and the impact strength of the composite decreased with the presence of cellulose content, as seen in Fig. 4. Generally, the addition of high fiber content increases the probability of fiber agglomeration [18], which creates regions of stress concentrations that require less energy to elongate the crack propagation. The impact strength reflects the ability of the material to resist impact, namely the toughness. Optimizing the interface between the fibers and the matrix through the use of compatibilizers or coupling agents can improve the toughness of these composites.

The flexural strength and modulus of neat PLA and PLA-cellulose based composites are summarized in

TABLE II Tensile properties of the PLA/TC 1004 composites

Polymer/cellulose (wt%)	Tensile strength (MPa)	Modulus of elasticity (GPa)	Improvement (modulus) (%)
PLA/Cellulose (100/0)	62.9 ± 4.9	2.7 ± 0.4	–
PLA/TC 1004 (90/10)	61.3 ± 2.6	3.2 ± 0.5	16
PLA/TC 1004 (80/20)	53.7 ± 7.8	4.5 ± 1.1	65
PLA/TC 1004 (70/30)	47.3 ± 2.5	6.3 ± 0.4	132
PLA/TC 1004 (60/40)	39.4 ± 5.3	6.8 ± 0.3	151

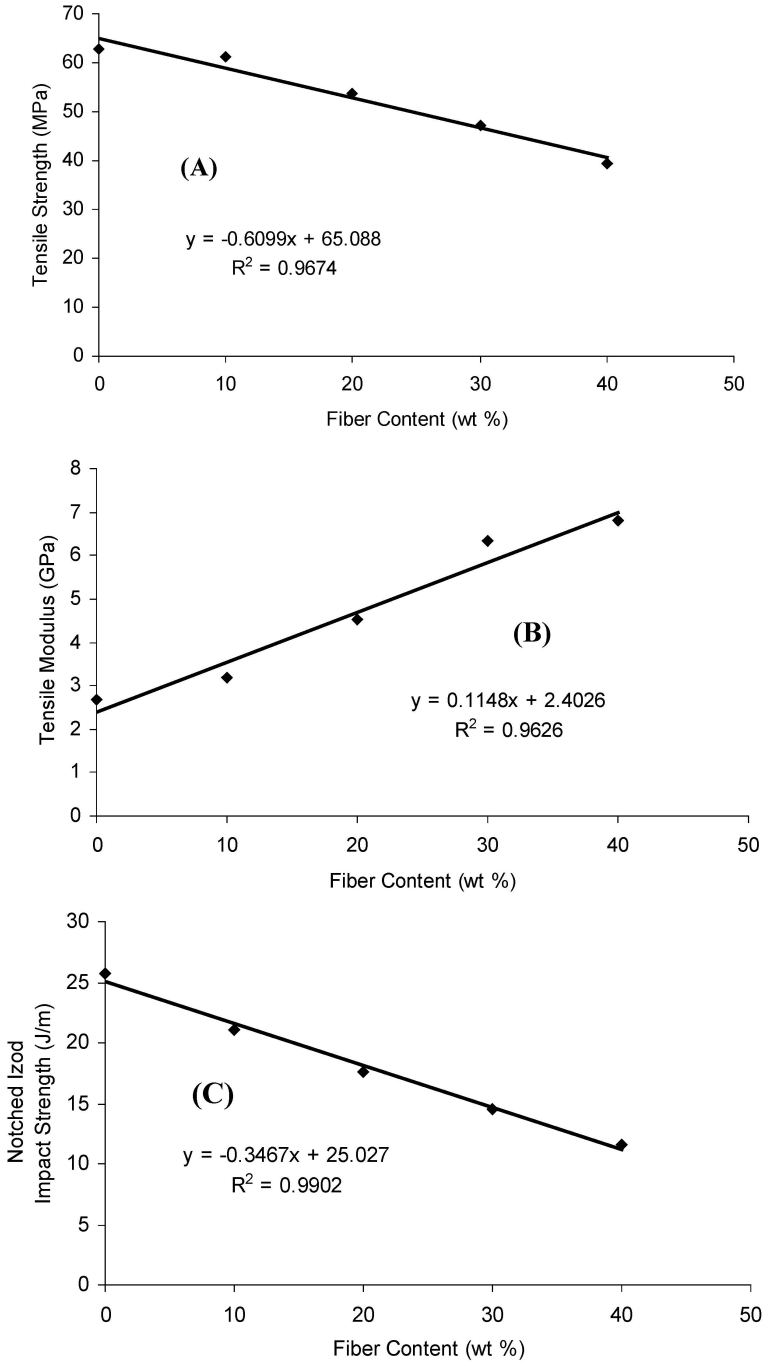

Figure 4 Dependence of (A) tensile strength, (B) tensile modulus, and (C) impact strength on the content for PLA/TC 1004 composites.

Table III. There is a significant increase in flexural modulus in the cases of PLA-cellulose based composites compared to that of neat PLA. As seen in Table III, the trend of the flexural modulus was identical to the trend in the tensile modulus described earlier. Generally, the tension and flexure moduli measurements can differ significantly when the material is inhomogeneous and anisotropic. A flexural test is strongly influenced by the properties of the specimen closest to the top and bottom surfaces, whereas a simple tension test reflects the average property through the thickness [19]. The stresses in a tensile test are uniform throughout the specimen cross-section, whereas the stresses in flexure vary from zero in the middle to maximum in the top and bottom surfaces [19].

TABLE III Flexural properties of the PLA and PLA based composites

Polymer/cellulose (wt%)	Flexural strength (MPa)	Modulus of elasticity (GPa)	Improvement (modulus) (%)
PLA/Cellulose (100/0)	98.8 ± 0.9	3.3 ± 0.1	–
PLA/TC 1004 (70/30)	77.7 ± 4.6	6.7 ± 0.2	103
PLA/R 0083 (70/30)	82.6 ± 3.8	6.2 ± 0.1	89
PLA/R 0084 (70/30)	76.3 ± 6.2	6.0 ± 0.1	83

3.2. Crystallization and melting behavior of the composites

The thermal properties such as the glass transition temperature (T_g), crystallization temperature (T_c), melting temperature (T_m), crystallization enthalpy (ΔH_c) and melting enthalpy (ΔH_m) obtained from DSC studies of the composites are summarized in Table IV. The degree

TABLE IV Thermal properties of the PLA and PLA based composites

PLA/cellulose (wt%)	T_g (°C)	T_c (°C)	ΔH_c (J/g)	ΔH_m (J/g)	χ (%)	T_m (°C)
PLA/Cellulose (100/0)	59	101	26.7	43.6	46.6	172
PLA/TC 1004 (70/30)	60	90	25.6	50.2	53.6	172
PLA/R 0083 (70/30)	55	81	18.7	41.1	43.8	168
PLA/R 0084 (70/30)	57	86	15.7	38.0	40.6	169
PLA/R 0083 (60/40)	51	81	5.7	26.6	28.4	167

TABLE V Thermal properties of PLA/TC 1004 composites

PLA/cellulose (wt%)	T_g (○C)	T_c (○C)	ΔH_c (J/g)	ΔH_m (J/g)	χ (%)	T_m (○C)
PLA/cellulose (100/0)	59	101	26.7	43.6	46.6	172
PLA/TC 1004 (80/20)	60	95	25.1	45.4	48.5	172
PLA/TC 1004 (70/30)	60	90	25.6	50.2	53.6	172
PLA/TC 1004 (60/40)	51	83	7.2	30.2	32.2	170

of crystallinity (χ %) of PLA in the composites was calculated using the expression: $\chi = \Delta H_m / \Delta H_m^0 \times 100$; where: ΔH_m = experimental melting enthalpy (J/g) and ΔH_m^0 = melting enthalpy of 100% crystalline PLA (93.7 J/g) [20]. The neat PLA shows 46.6% degree of crystallinity along with a glass transition at 59°C. This result is in accordance with the result of Park *et al.* [21]. As shown in Table IV, T_g and T_m of the composite did not change for PLA/TC 1004 (70/30) when compared with PLA/Cellulose (100/0), however R 0083 and R 0084 composites show only a marginal change in T_g and T_m. The thermal behavior of the PLA/R 0083 (60/40) showed that high cellulose contents did not affect the melting temperature and the glass transition temperature significantly when compared with PLA/R 0083 (70/30) composite. Crystallization enthalpy, crystallization temperature, and melting enthalpy changed with the addition of cellulose fibers.

It can be seen from Table V that T_g and T_m of the composite did not change significantly up to 30% fiber content. Though T_c for PLA matrix in the composites changed with increasing the cellulose content, ΔH_c remained nearly unchanged. These results suggest that cellulose fibers do not affect significantly the crystallization behaviors of PLA matrix up to 30% cellulose content.

3.3. Dynamic mechanical properties

Figs 5A and B show dynamic storage modulus, and tan delta of the PLA based composites, as a function of temperature, respectively. It is clear from Fig. 5A and Table VI that the modulus increases in the presence of fibers in the composites, i.e., the storage modulus of PLA based composite is higher than that of PLA matrix. The height of the tan delta peak decreased with the presence of cellulose content as shown in Fig. 5B. According to Liu *et al.* [22], one possible explanation of this phenomenon is that neat PLA shows a sharp and intense peak because there is no restriction to the chain motion, where as the presence of the cellulose surfaces hinder the chain mobility, resulting in the reduction of sharpness and height of the tan delta peak. Furthermore,

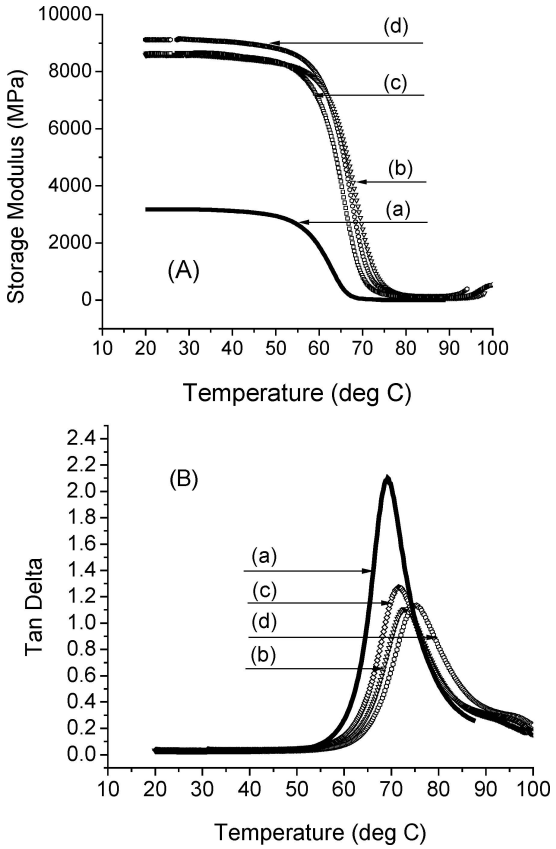

Figure 5 Temperature dependence of (A) storage modulus, and (B) Tan delta of PLA and PLA based composites: (a) PLA/Cellulose (100/0) (□) (b) PLA/R 0083 (70/30) (X), (c) PLA/TC 1004 (70/30) (∀), and (d) PLA/R 0084 (70/30) (υ).

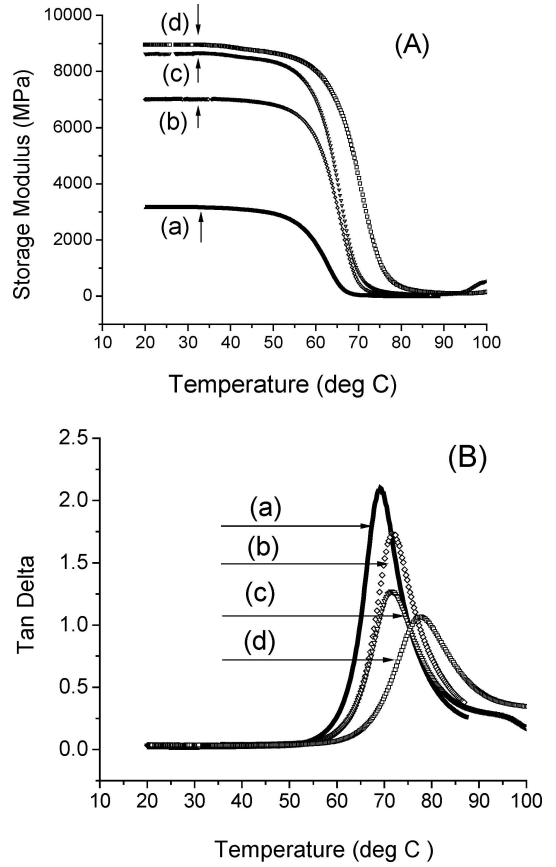

Figure 6 Temperature dependence of (A) storage modulus, and (B) Tan delta of PLA/TC 1004 composites: (a) PLA/TC 1004 (100/0) (□), (b) PLA/TC 1004 (80/20) (M), (c) PLA/TC 1004 (70/30) (X), and (d) PLA/TC 1004 (60/40) (∀).

TABLE VI The storage modulus of the PLA and PLA based composites

PLA/cellulose (wt%)	Storage modulus (GPa) at 25°C	Storage modulus (GPa) at 40°C	Storage modulus (GPa) at 60°C (near T_g of the composite)	Reinforcement imparted by the cellulose fibers at 25°C (modulus) (%)
PLA/cellulose (100/0)	3.2	3.1	1.8	–
PLA/TC 1004 (70/30)	8.6	8.5	7.0	168
PLA/R 0083 (70/30)	8.6	8.5	7.5	168
PLA/R 0084 (70/30)	9.1	9.0	7.8	184

TABLE VII The storage modulus of the PLA/TC 1004 composites

PLA/cellulose (wt%)	Storage modulus (GPa) at 25°C	Storage modulus (GPa) at 40°C	Storage modulus (GPa) at 60°C (near T_g of the composite)	Reinforcement imparted by the cellulose fibers at 25°C (modulus) (%)
PLA/TC 1004 (100/0)	3.2	3.1	1.8	–
PLA/TC 1004 (80/20)	7.0	6.9	5.5	118
PLA/TC 1004 (70/30)	8.6	8.5	7.0	168
PLA/TC 1004 (60/40)	9.0	8.9	7.9	181

Fay *et al.* [23] reported that the reduction in the tan delta peak is due to the immobilization of polymer matrix in the presence of the filler. The reduction in tan delta also denotes an improvement in the hysterisis of the system and a reduction in the internal friction [22, 23].

Figs 6A and B show respectively the temperature dependence of the storage modulus and tan delta for the PLA/TC 1004 composites filled with 20, 30 and 40% cellulose fibers. The storage modulus increased with increasing cellulose fiber content (Table VII). At low fiber loading, the matrix is not restrained by fibers and highly localized stresses occur [10]. The stress is more evenly distributed as the fiber concentration increases, resulting in a higher strength of the composite. The reduction of modulus with increase in the temperature is associated with softening of the matrix at higher temperature [24]. Further, the drop of matrix modulus is compensated by the fiber stiffness on increasing the temperature. The thermal expansion mismatch between the cellulose fiber and the PLA matrix might result in reduced properties [25]. It is also interesting to note that the amplitudes of tan delta peaks of the composites are lower than that of neat PLA as the amount of cellulose content increases in Fig. 6B.

3.4. Heat deflection temperature (HDT)

The HDT of the cellulose fiber-reinforced PLA composites increased when compared with the HDT of PLA resin (Table VIII). HDT represents the upper stability limit of the material in service without significant physical deformation under load and temperature. Saha *et al.* [26] found that the HDT of the composites increased with increasing glass fibers content on the study of unidirectional polyethylene glass fiber poly(methyl methacrylate) hybrid composite laminates. Similar results were also seen in the study of pultruded fiber-reinforced polyurethane composites [27].

TABLE VIII HDT of the PLA and PLA based composites

Polymer/cellulose (wt%)	HDT (°C)
PLA/cellulose (100/0)	64.5
PLA/TC 1004 (70/30)	73.1
PLA/R 0083 (70/30)	72.5
PLA/R 0084 (70/30)	73.0

TABLE IX TGA characterization of the composites

Polymer/cellulose (wt%)	T_5 (°C)	T_{25} (°C)	T_{50} (°C)	T_{75} (°C)
PLA/ cellulose (100/0)	356	385	401	414
PLA/TC 1004 (0/100)	177	305	330	348
PLA/TC 1004 (70/30)	308	336	349	356
PLA/R 0083 (0/100)	284	360	396	>600
PLA/R 0083 (70/30)	307	339	354	369
PLA/R 0084 (0/100)	282	355	391	>600
PLA/R 0084 (70/30)	325	360	376	394

3.5. Thermogravimetry

The thermal stability of pure PLA and PLA based composites was investigated with thermogravimetric analysis. In Table IX, the 5, 25, 50, and 75% weight-loss temperatures (T_5, T_{25}, T_{50}, and T_{75}, respectively) are listed for all specimens shown in Fig. 7. 88, 93 and 96% weight-loss were observed at 500°C for the composites of R 0084, R 0084 and TC 1004, respectively.

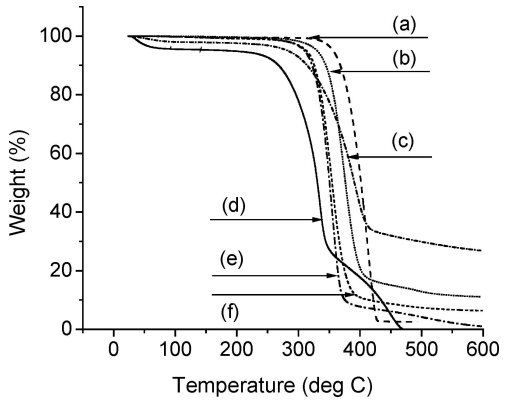

Figure 7 Thermogravimetric curves of the composites: (a) PLA/Cellulose (100/0) (□), (b) PLA/R 0084 (70/30) (□), (c) PLA/R 0084 (0/100) (□), (d) PLA/TC 1004 (0/100) (□), (e) PLA/TC 1004 (70/30) (□), and (f) PLA/R 0083 (70/30) (□).

The thermal stabilities of the composites with the same cellulose fiber content are in the order: PLA/R 0084 > PLA/R 0083 > PLA/TC 1004. Usually, most of the cellulose decomposes at a temperature of 350°C, and the thermal stability of the composite materials increases with fiber content [28]. It was also observed that the weight loss of neat TC 1004 fiber sample is relatively high in comparison to the neat R 0084 and R 0083 samples as seen in Table IX.

3.6. Morphology of the composites

The morphology of the cellulose fibers was investigated by SEM as shown in Figs 8–10. Figs 8–10 show evidence of fiber breakage for the R 0083, R 0084 and TC 1004 fibers. The SEM was used to sort out the fibers of different diameters ranging from 5 to 35 μm. The fiber-matrix interface in the composites was also investigated by SEM. SEM micrographs of the impact fracture surfaces of the composites with the same cellulose content (30 wt%) are displayed in Figs 11–13. These micrographs illustrate the individual separation and dispersion of the cellulose fibers in the form of single fibers, which indicates that the cellulose fibers have been separated during the extrusion process and well dispersed in the PLA matrix [29]. Some fibers are tightly connected with the matrix, and some cellulose fibers are broken and/or torn up. The micrograph of PLA/TC 1004 composite is shown in Fig. 13, which indicates the presence of voids in the matrix resulting in lowering of tensile strength and only a marginal

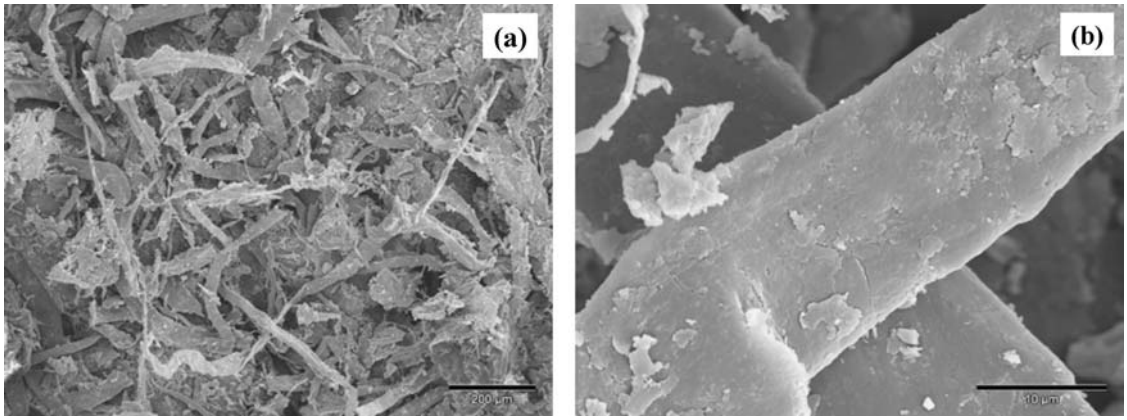

Figure 8 SEM micrographs of the R 0083 fibers: (a) 200 μm and (b) 10 μm.

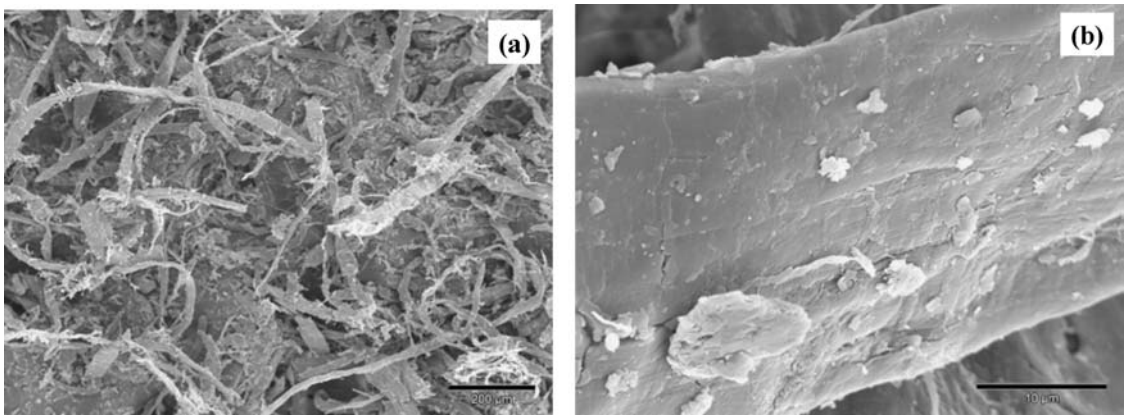

Figure 9 SEM micrographs of the R 0084 fibers: (a) 200 μm and (b) 10 μm.

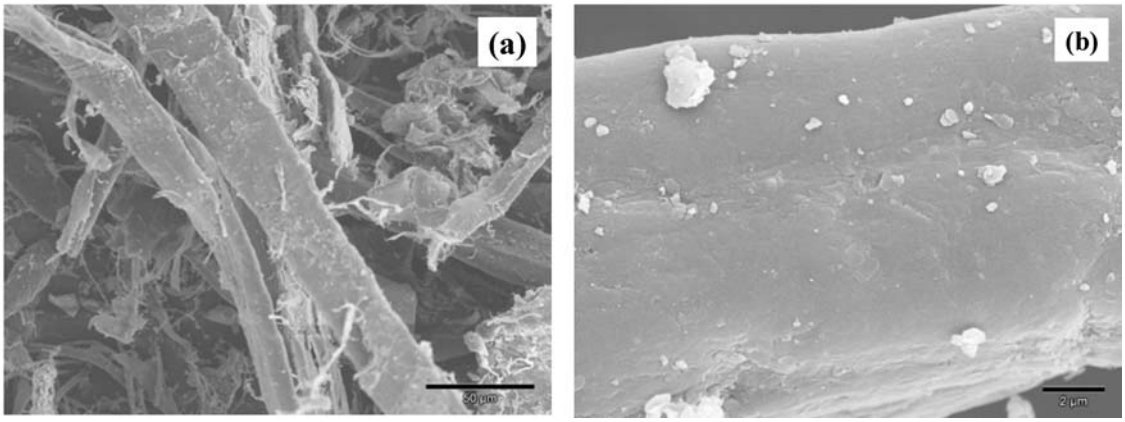

Figure 10 SEM micrographs of the TC 1004 fibers: (a) 50 μm and (b) 2 μm.

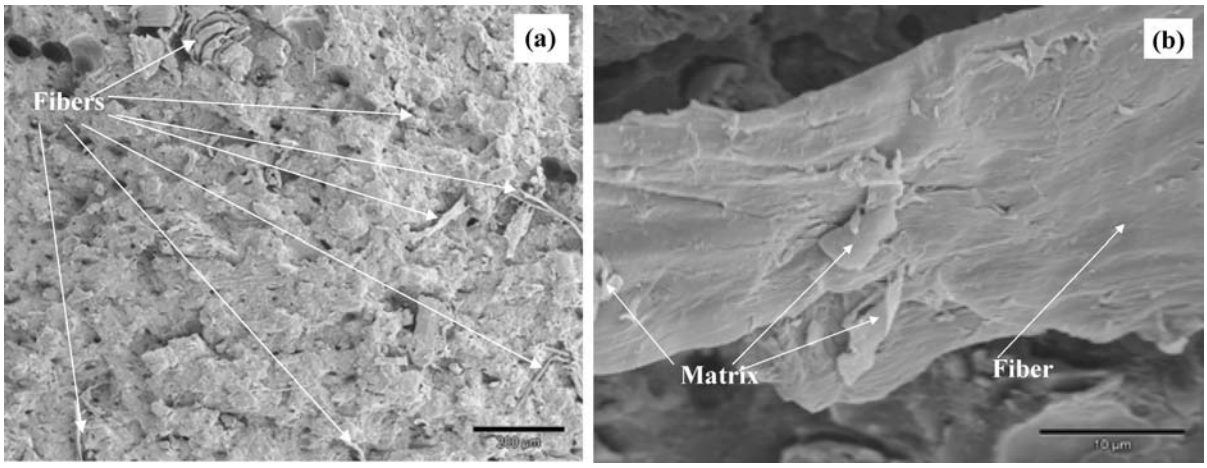

Figure 11 SEM micrographs of PLA/R 0083 composites: (a) 200 μm and (b) 10 μm.

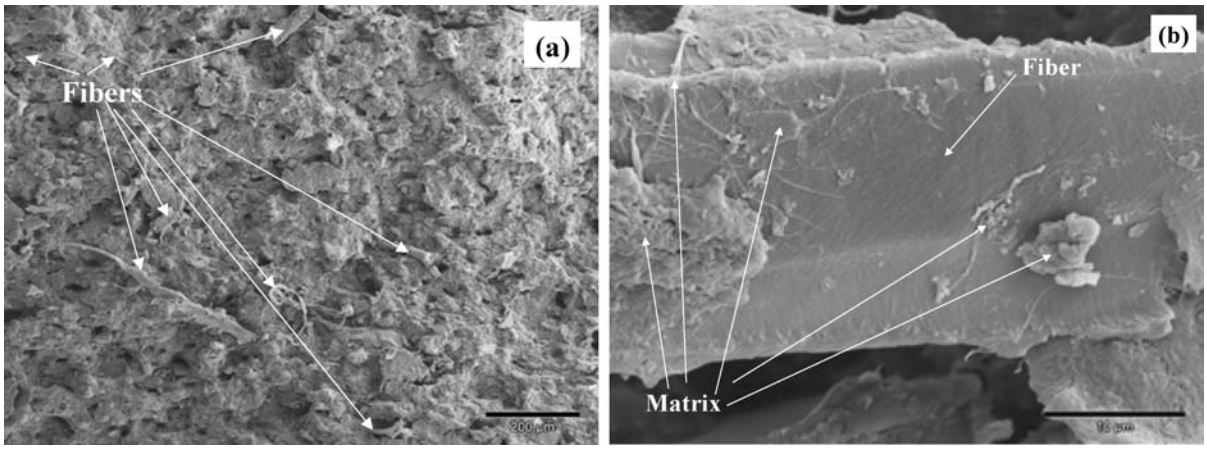

Figure 12 SEM micrographs of PLA/R 0084 composites: (a) 200 μm and (b) 10 μm.

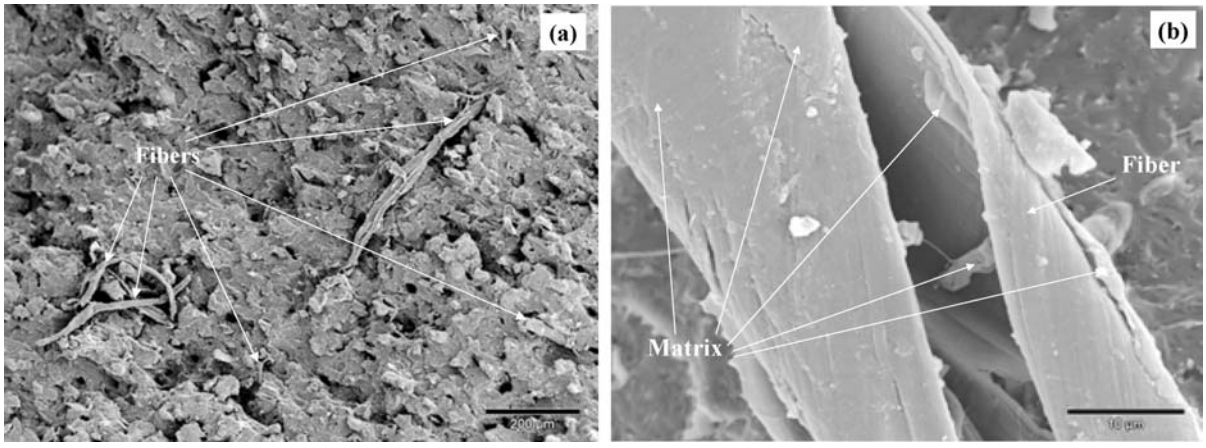

Figure 13 SEM micrographs of PLA/TC 1004 composites: (a) 200 μm and (b) 10 μm.

increase in flexural strength of the composite. In Figs 11b–13b, closer observation of the fiber pull-out in micrographs show that the fiber has been covered with a thin layer of the matrix, which lead to better stress transfer between the matrix and the reinforcing fibers [17]. Comparing Fig. 13a with Figs 11a and 12a, it is clear that there are less voids between fiber and matrix in PLA/R 0083 and PLA/R 0084 than PLA/TC 1004 composites. The fiber pull-outs are larger in Fig. 13a than those in Figs 11a and 12a.

4. Conclusions

Biobased composites were obtained from PLA and cellulose fibers that possess superior mechanical and thermal properties as a result of reinforcement. Generally these results together with other studies [1, 30, 31] indicate that PLA/natural fiber composites have mechanical properties of sufficient magnitude to compete with conventional thermoplastic composites. Hence the use of natural fibers, as reinforcements in PLA, gives interesting alternatives for production

of low cost and ecologically friendly composites. In the future, efforts will be made to evaluate the biodegradability of these cellulose fiber reinforced PLA composites.

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