Study of Naturally Woven Coconut Leaf Sheath-Reinforced Polypropylene Matrix Based Composites

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ABSTRACT

Renewable materials have gained significant attraction as reinforcement in polymer matrix composites. Naturally woven coconut leaf sheath (CLS) fiber-reinforced polypropylene (PP) composites were prepared by compression molding. Different composites were fabricated varying the weight percentage of CLS fiber (5, 10, 15, 20 and 25 wt%). Hydrophilic nature of the CLS fiber was evaluated using water uptake test and revealed strong hydrophilicity by gradually absorbing water until it reaches to a constant water uptake value of 150%. Structural features of the composites were confirmed by infrared spectrophotometry. Mechanical performance was investigated using tensile and bending tests. Fiber reinforcement significantly increased the mechanical properties of the composites, and the optimum improvement of strength and stiffness was achieved for the 10% fiber content. The tensile strength and stiffness was improved by 60 and 40%, respectively, whereas, bending strength and stiffness was increased by 70 and 50% respectively. Moreover, thermal properties, and degradation nature of the composites were investigated. Addition of fiber decreased the thermal stability of the composites. Degradation test of the composites were performed up to six months in soil medium at ambient condition and showed slow degradation of mechanical properties over time. The optimized composite retained 70% to 75% of its initial tensile strength and modulus, after six months degradation time.

Key word: Coconut leaf sheath fiber, polypropylene, composite, mechanical properties, thermal properties, soil degradation.

INTRODUCTION

The world is now going towards natural things. Environmental polices of developed countries are increasing pressure on manufacturers to consider the environmental impact of their products. Therefore, the interest in using natural fibers as a reinforcement of polymer–based composites is growing mainly because of its renewable origin [1]. Polymer matrix composites are finding widespread applications in chemical industry and many other areas as structural materials [2]. In recent years, a great interest has been generated in the use of agro-fiber as substitute for synthetic fiber (e.g., carbon and glass) in the development of polymer matrix composites due to their more favorable properties such as low density, non-abrasive,

stiffness, high specific properties and non-brittleness. The environment as well as economic advantages such as biodegradability, abundance throughout the world, rural jobs generation, non-food/agricultural based economy, low cost, low energy consumption and utilization are also among its advantages [3,4]. Throughout the past two decades, natural fibers have been utilized in many applications including consumer goods, low-cost housing, automotive, aircraft, paper industries, textile, building and construction industries [5]. Extensive studies have been carried out on lignocellulosic fibers such as sisal [6-8], pineapple [9-11], banana [12-14], bamboo [15], vakka [16], date [17], and oil palm empty fruit bunch fibers [18, 19], and the results have shown that lignocellulosic fibers can be used as an effective reinforcing agents. This is because of its several advantages such as abundance, renewability, biodegradability, environmentally friendly, high strength-to-weight ratio, and low cost compared to their synthetic counterparts.

Coconut palm (*Cocos nucifera*, Linn.), a valuable tropical resource for food and energy, is plenteous in the coastal districts of Bangladesh. The coir fibers, obtained from coconut palm, are being used to make a variety of materials including carpets and mats. However, there are many other parts of the palm other than coir from which fibers can be extracted but are not currently used as resources for materials in an organized way. In fact, all of these fibers are currently used as fuel in the rural area, which does not take advantage of their high length to diameter ratio. Some of these fibers occur in nature already in woven form e.g. leaf sheath (Figure 1 (a) and b), which can be readily impregnated with plastics or cement. In order to find better utilization of coconut leaf sheath (CLS), particularly as fillers or reinforcement in relatively low cost applications (where high strength or high modulus is not required), it is necessary to evaluate the properties of fiber-reinforced composites. Hence, we processed the naturally woven leaf sheath fibers into short fibers for thermoplastic polymer reinforcement.

As such, sandwiching this fiber with thermoplastics could offer a strategy for producing advanced composites based on end-use requirements within a framework of low cost, recyclability, availability, bio-degradability and environmental consideration. Moreover, thermoplastic matrix materials are the most important part of a composite. Polypropylene (PP) is an amorphous thermoplastic polymer and are widely used as engineering thermoplastic because of several vital and useful properties such as transparency, dimensional stability, flame resistance, high heat distortion temperature, and high impact strength. PP is also very suitable for filling, reinforcing, and blending. PP with fibrous natural polymers of biomass origin is one of the most promising routes to create natural-synthetic polymer composites [20-26]. In this paper, we present the fabrication and assessment of new cellulosic short fiber-reinforced polypropylene composites. The structural, mechanical, degradation and thermal properties of the composites are studied to find out its potential towards diversified applications ranging from automotive to consumer products within the environmental legal framework. The aim is to find out the optimum fiber volume fraction in the composite to maximize the mechanical properties, reveal trends of mechanical properties variation over degradation time in soil and determine the high temperature degradation under air and nitrogen atmospheres.

EXPERIMENT
Chemicals and instrumentation

PP was purchased from Polyolefin Company Private Ltd., Singapore and was grinded to small particles (50-60 µm) for appropriate and homogeneous matrix-fiber adhesion. Coconut leaf sheath fibers (*Cocos nucifera*, Linn.) were collected from the rural area of

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Bangladesh and chopped into small pieces (2-3 mm) using hand scissors (**Figure 1(c)**). The chopped fibers were cleaned with distilled water and exposed thoroughly to sunlight for about 24 h. Finally, the cleaned fibers were vacuum dried under 100 ^{o}C for 5 h prior to the composites fabrication.

Fourier Transform Infrared Spectroscopy (FTIR)
    The structural analysis of the PP and composites was conducted using a Perkin Elmer Fourier Transform Infrared (FTIR) Spectrometer with an attachment of attenuated total reflectance accessory. The spectra were recorded over the range of 4000-650 cm^{-1} at a resolution of 4 cm^{-1}.

Mechanical properties of the composites
    The tensile properties of the composites (F1-F5) were measured using a universal testing machine (model H50 KS-0404, Hounsfield Series S, UK) with a load capacity of 5000 N at a crosshead speed of 10 mm/min, according to DIN 53455. The gauge length was 20 mm. For bending properties measurement, the crosshead speed was 10 mm/ min, and span distance was 40 mm. The three point bending tests were carried out according to DIN 53452 standard method. The average results of minimum five samples for each composite were reported.

Surface morphology
    The fracture surfaces of the tensile specimens of F2 and F5 composites were examined using a Hitachi S-4000 field emission scanning electron microscope, operated at 5 kV. Samples were mounted with carbon tape on aluminum stubs and then sputter coated with carbon tape on aluminum stubs and then sputter coated with platinum and palladium to make them conductive prior to scanning electron microscopy (SEM) observation.

Thermal analysis
    The thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) of the polypropylene and optimized composite were recorded on a NETZSCH instrument (model no. STA 449 F3, Jupiter). The temperature was increased from 0 to 900 ^{o}C at a rate of 20°C/min under inert nitrogen atmosphere.

Procedure
Composite fabrication
    The PP matrix based unidirectional composites were made by compression molding. At first, the mould (12 x 15 cm^{2}) was cleaned with wax as a cleaning agent. The mixture of dried CLS short fibers and PP powder were prepared according to the formulation of **Table 1** (5, 10, 15, 20 and 25 wt% fiber) and poured into the mould. The processing temperature was maintained at 190˚C for 5 min under 5 bar consolidation pressure in the heat press (Carver, INC, USA Model 3856). The molds were then cooled for 1 min in a separate press under 5 bar pressure at room temperature. **Figure 1 (d)** shows the final composite product.

Water uptake of the CLS fiber
    The water uptake of the CLS fibers was periodically monitored up to 10 days by soaking the pre-weighed dried samples in the distilled water at room temperature. The samples were removed from time to time, lightly wiped to remove surface water and weighed. Water uptake is determined using the following equation.

$$\text{Water uptake} = \frac{W_f - W_i}{W_i} \cdot 100 \qquad\qquad (1)$$

Where, W_i is the initial weight of the samples, and W_f is the final weight of the samples after fixed time.

Degradation test of the optimized composite

The degradation test of the optimized composite was carried out for six months burying the samples in humid soil whose moisture level was at least 25 %. After set time point, the samples were taken out, washed with distilled water and dried at 80 °C for 8 h. The variation of tensile strength and tensile modulus (stiffness) was periodically noted in order to determine the degradable character of the samples in this environment.

Table 1. The composition of the different composites.

Formulation no.	Weight percentage of PP and CLS in different composites
F1	95% PP + 5% CLS
F2	90% PP + 10% CLS
F3	85% PP + 15% CLS
F4	80% PP + 20% CLS
F5	75% PP + 25% CLS

Figure 1. (a) Coconut palm, arrows show woven leaf sheath, (b) naturally woven coconut leaf sheath, (c) chopped coconut leaf sheath; and (d) chopped coconut leaf sheath-reinforced PP composite.

RESULT AND DISCUSSION
Water uptake of the CLS fiber

The hydrophilic nature of the fiber influences its water retention capacity. **Figure 2** shows the water uptake profile of the CLS fiber immersed in distilled water at room temperature for different time periods. Initially, the fiber gradually absorbed water and reached a plateau value of 150% after 8 days. The degree of hydrophilicity gave a quick

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idea of the strength of fiber-matrix adhesion, structural stability, and potentiality of the composites to a wide range of outdoor exposures and final biodegradability after being discarded. It is well known that hydroxyl groups (-OH) present on the surface of the fibers is responsible for water absorption [27]. CLS fiber contains about 70% cellulose (α-cellulose and hemicelluloses) and 28% lignin [28]. The cellulose structure of CLS possesses a lot of – OH groups. These hydroxyl groups are responsible for the strong hydrophilic nature of CLS. The cellulosic –OH groups are very reactive, they and other oxygen-containing groups attract water through hydrogen bonding giving a strong hydrophilic nature to CLS fiber [29]. However, high water uptake of fiber is unwanted for a wide range of fiber reinforced composites in outdoor applications.
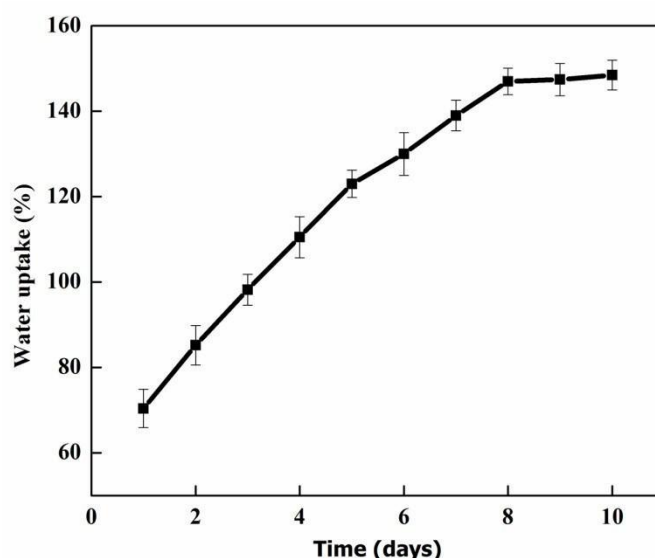

Figure 2. Water uptake profile of naturally woven coconut leaf sheath (CLS) soaked in distilled water for different time period.

Structural characterization

Naturally woven CLS short fiber-reinforced PP composites were prepared varying the fiber content from 5 to 25 wt%, and the five formulations, namely, F1, F2, F3, F4 and F5 were presented in the **Table 1**. The FTIR spectra of PP and F2 composite were illustrated in **Figure (3)** and **(4)**, respectively. As shown in **Figure 3**, the fingerprint region of pure PP was observed from 1455-650 cm^{-1}, particularly 1455 and 997 cm^{-1} ascribed to C=C stretching and C-H bending vibrations, respectively. The peak observed at 2916 cm^{-1} assigned to C-H stretching vibration. The spectrum of composite showed a peak at about 1739 cm^{-1} associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose in the natural CLS fiber [30], along with characteristic peaks of PP (**Figure 4**).
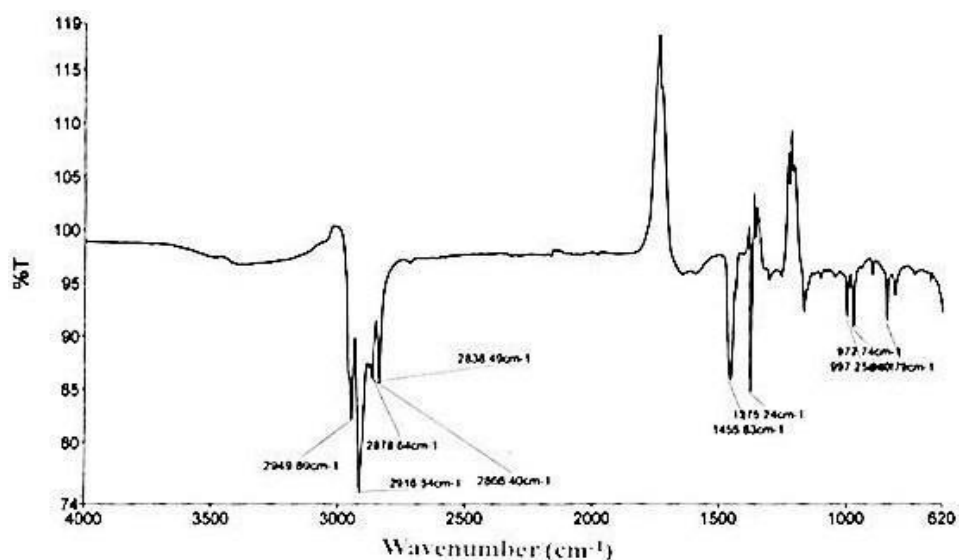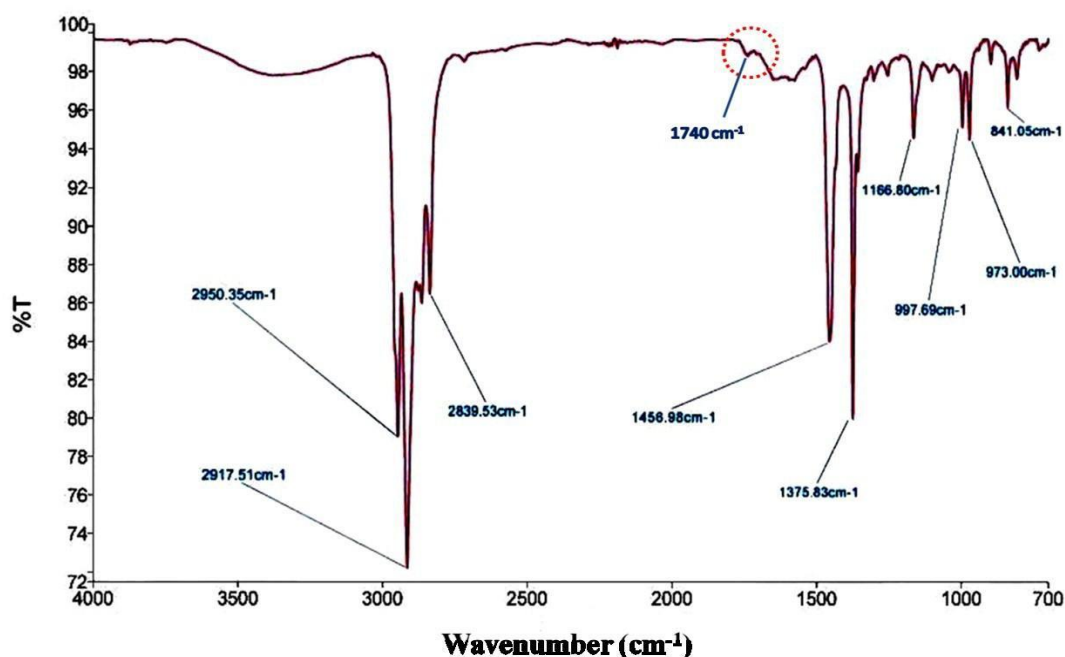
Figure 3. FTIR spectra of polypropylene.

Figure 4. FTIR spectra of optimized composite F2 (90% PP +10% CLS).

Mechanical properties of the composites

Mechanical property of the composites is an important factor for the application of interests. As such, tensile and bending tests of the composites were done to measure the strength and stiffness of the composites. **Figure 5** showed typical stress-strain curves for the composites. All composites exhibited non-linear stress-strain curves demonstrating ductile behavior. These curves showed different ductile failure behaviours as a function of fiber content. The F1 formulation (90% PP + 10% CLS) exhibited more ductile nature and correspondingly decreased as the fiber content increased from 5 to 25 % in the composites,. These ductility variations might be dominantly originated from the PP matrix to fiber ratio and fiber-matrix interaction. Moreover, the stress to failure increased with increasing fiber content from 5 to 15 % but beyond 15% stress decreased. The maximum stress to failure was

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obtained for 10% fiber content and this might be due to more evenly distributed fibers as well as better uniform stress transfer. The tensile strength (TS), bending strength (BS), elongation at break (Eb%), tensile modulus (TM) and bending modulus (BM) of the composites are presented in **Figure 6 (a)** and **(b)**, respectively. It was found that fiber reinforcement occurred improving the strength and stiffness of the composites. The increase in fiber content from 5 to 10 wt% resulted in both increased tensile and bending strength. The highest TS and BS values were found for F2 (90% PP + 10% CLS) and was considered as an optimized composite. Interestingly, higher amount of fiber content (>10 wt%) reduced TS and BS of the F3, F4 and F5 composites. From F1 to F2 formulation, the TS and BS values increased, significantly, increase TS was also observed as they gradually declined from F3 to F5 formulations. As observed from **Figure 6 (a)**, the highest TS and BS values of 27 MPa and 46 MPa, respectively, was found for F2 formulation. Elongation at break (Eb%) is one of the significant mechanical properties of any engineering material. As a whole, the composite showed lower elongation at break for all formulations. However, F1 showed slightly higher Eb% (24%) compared to those of other formulations (17 to 18%). This phenomenon might be due to the low elongation at break (%) of the CLS fibers compared to the matrix PP. It was also noted that melted PP matrix filled the spaces between the fibers during the compression molding process. A higher volume fraction of PP matrix in F1 formulation and relatively lower fiber-matrix interaction may contribute to an increased Eb% of F1. However, there is no significant differences were observed with the variation of fiber content.

The significant enhancement in the tensile and bending modulus of the composites was obtained with increasing fiber content from 5 to 10 wt%, as shown in **Figure 6 (b)**. Beyond 10 wt% fiber content, the composites exhibited a decreased modulus with loading up to 25 wt% fiber content. Likewise strength, F2 formulation showed the maximum TM and BM values of 278 MPa and 525 MPa, respectively. A 10 wt% fiber loading increased correspondingly 170 and 140% of TM and BM of the composite.
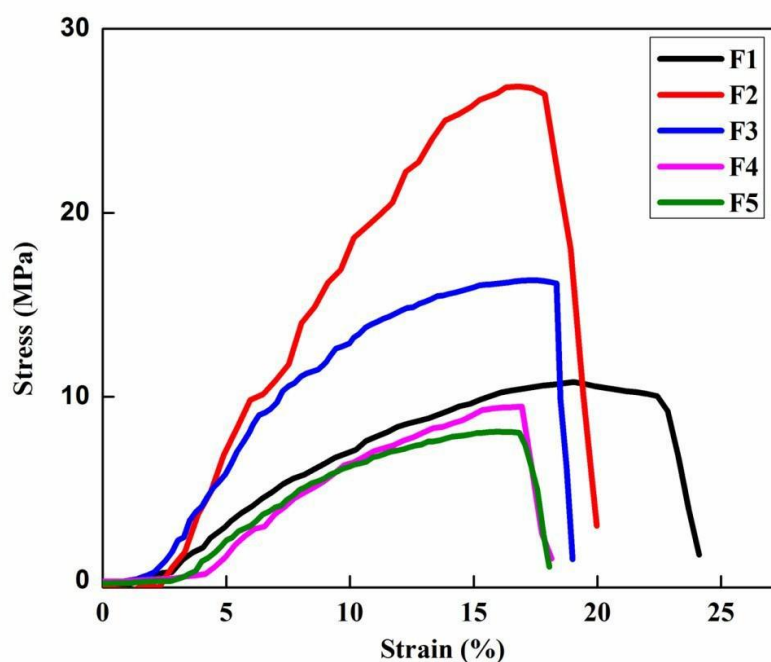

Figure 5. Representative stress-strain curves of the composites.

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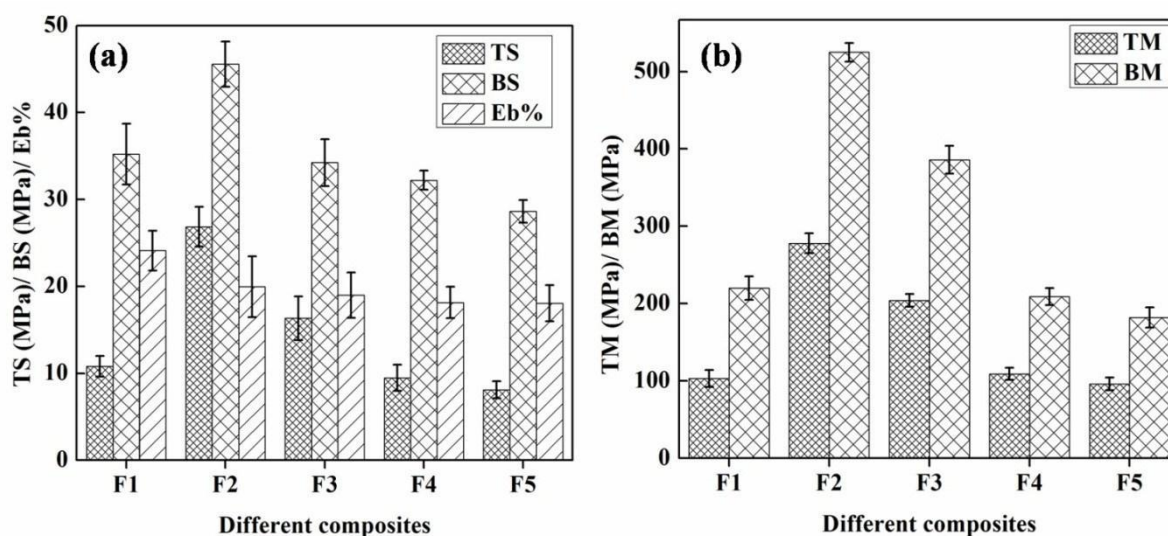**p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733**
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Figure 6. Mechanical properties (a) tensile strength, bending strength and elongation at break (Eb%) and (b) tensile and bending modulus of the composites.
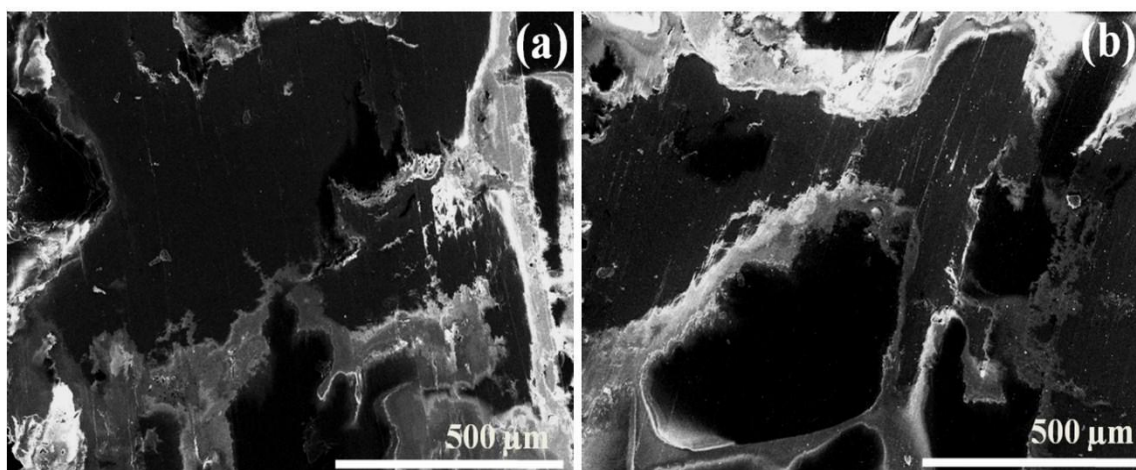

Figure 7. SEM images of (a) optimized composite F2 (90% PP +10% CLS) and (b) F5 composite (75% PP + 25% CLS).

It is expected that with the 10 wt% of fiber reinforcement, the fibers are able to contribute to the effective stress transfer between fibers and the PP matrix and concomitantly increases the tensile strength.

We performed the scanning electron microscopy (SEM) analysis on the fracture surfaces of the F2 (optimized composite) and F5 (overloaded fiber composite) to examine the effect of fiber content on the fracture mechanism of the composites. The improvement in the strength is attributable to the strong interaction between fibers and the PP matrix, as evidenced by a scanning electron micrograph being taken on fracture surface of optimized composite containing 10 wt % fibers (**Figure 7 (a)**). It is worthwhile to mention that the properties of hybrid composites depends not only on the properties of individual reinforcing fibers but also on the orientation and arrangements pattern of the fibers, fiber-matrix adhesion etc. The decreased strength and stiffness at higher CLS fiber content (15 to 25 wt%) could be attributed to poor adhesion between the fiber and the matrix, which promotes more micro

crack formations at the interface. A higher fiber content results in more (and probably larger) void formation during processing, which leads to micro crack formation under loading and therefore reduces the strength [31]. Moreover, a higher content of short CLS fiber might exhibit a large extent of pull-outs in the composites compared to the lower fiber content. In addition, the possibility of fiber entanglements, particularly in the composite with random fibers, increases as the fiber content increases. These entanglements act as defect for mechanical strength. SEM images of fracture composite with 25 wt% fiber content showed larger fiber breakages and gaps as well as delamination suggesting the reduced matrix to fiber interactions and dispersion problems (**Figure 7 (b)**). On the other hand, at lower fiber content (5 wt %), the matrix is not restrained by enough fibers and highly localized strains occur in the matrix at low stresses, hence reduces the strength and stiffness but improves extensibility by bit [32].

From the overall results of **Figure 6 (a)** and **(b)**, it is found that 10 wt % is the optimum amount of fibers where TS, TM, BS and BM are recorded highest amongst the respective fiber composites due to optimum fiber-matrix interaction and more evenly distribution of stresses.

Thermal properties of the composites

Thermal characteristics of the PP matrix (granules) and the optimized composite (10 wt % fiber) were evaluated by TGA and DSC. The thermograms of both PP granules and optimized composite were presented in **Figure 8** and **9**, respectively. As shown in **Figure 8**, the degradation process of PP started at about 380 °C and 98.73% mass change was completed at 500 °C. However, the decomposition of the composite started earlier compared to the PP matrix which might be attributed to the fact that fiber-matrix adhesion in the composite (**Figure 9**). The addition of fibers decreased the thermal stability of the composites. As seen from the thermogravimetric curve of composite, the mass change started at 320 °C. The overall lower thermal stability of the composite might be due to the entrapped air during composite fabrication as well as the presence of thermally unstable fiber constituents such as hemicelluloses and pectins.
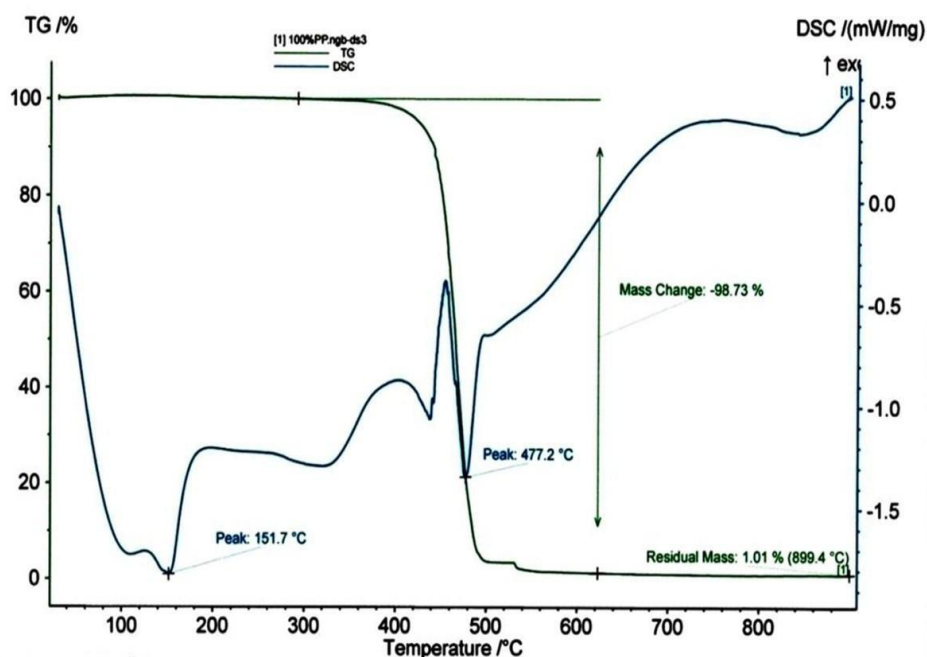

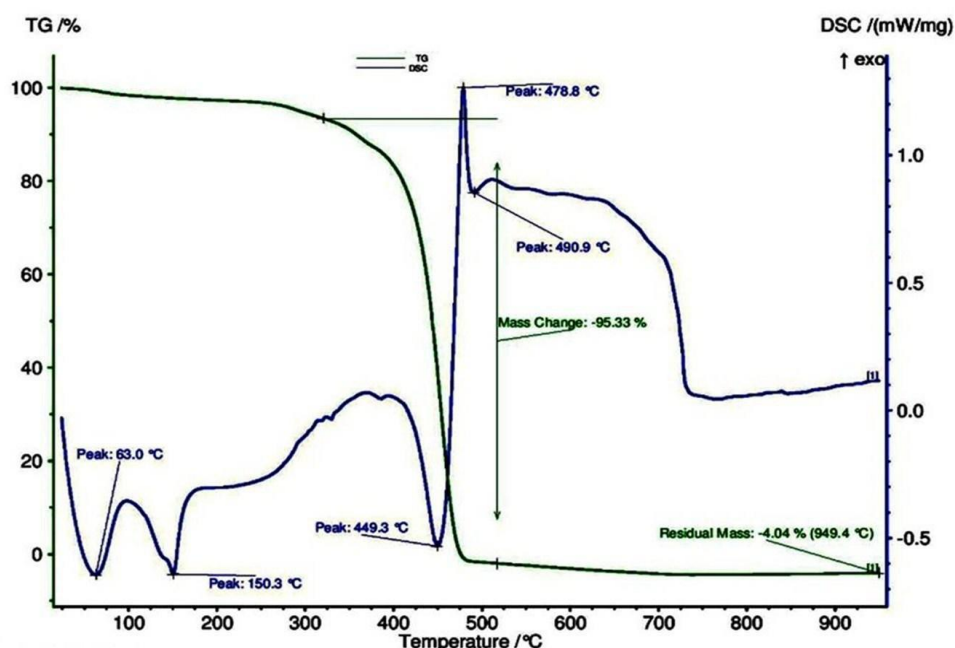**Figure 8.** TGA and DSC of polypropylene granules.

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Figure 9. TGA and DSC of optimized composite F2 (90% PP +10% CLS).

Figure 8 and **9** revealed that the melting profile of composite did not change significantly. Only a little displacement was observed in respect to neat PP. The DSC scans of the PP matrix and composite showed peaks of melting at 151.7 and 150.3^{o}C, respectively. The temperature peak of melting slightly decreased because of the influence of added fibers, showing an interaction between fiber and matrix.

Soil degradation of the optimized composite

To understand the detrimental effects of soil environment on the mechanical property of the composite, it is necessary to carry out the soil degradation test. The degradation test of the optimized composite was performed in the soil medium (high moisture content) for up to six months. Tensile strength (TS) and tensile modulus (TM) values were plotted against degradation time and was shown in **Figure 10**. It was found that gradually TS and TM values were decreased over time, as expected. A slight decrease of TM was observed compared to TS of the composite.
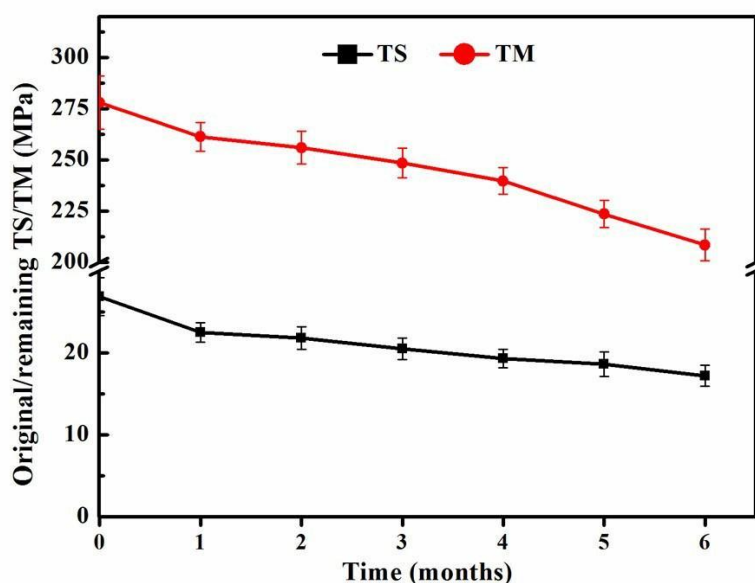
Figure 10. Degradation of tensile strength (TS) and tensile modulus (TM) of the optimized composite F2 (90% PP +10% CLS) in the soil medium.

After six months of degradation in the soil, composite retained 70 and 75% of its initial tensile strength and modulus. During immersion of the composite in the humid soil medium, water penetrated from the fiber edges of the composites and thus degraded the fibers slowly inside the composite as well as weakening of the bonding strengths between the fibers and matrix. Strength is lost as the cellulose polymers undergo degradation into oligomers of glucose (cellobiose, cellotriose, cellotetraose, cellopentaose, cellohexaose) through hydrolysis, as shown in the **Scheme 1**.
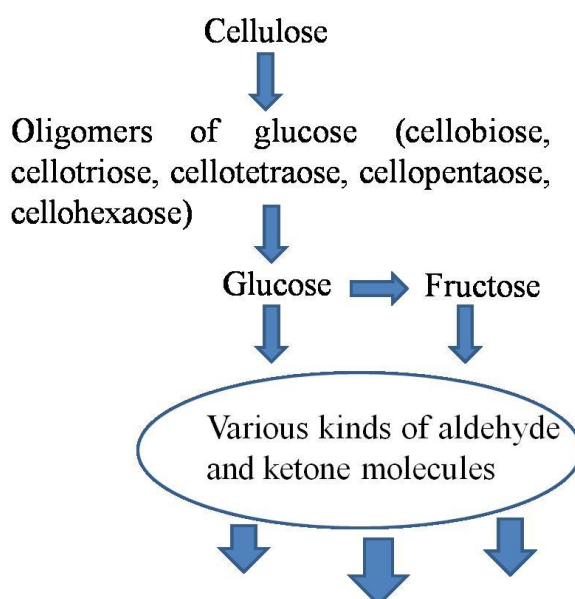
Scheme 1. The probable reaction pathways of cellulose hydrolysis.

Moreover, bacterial attack could also responsible for the loss of the mechanical properties because degradable fibers are generally attacked by microbial activity. Bacteria are ubiquitous in the soil and water. Lignocellulosic-degrading bacteria alone or work synergistically with other bacteria and soft-rot fungi to degrade lignocellulosic materials (lignin, cellulose, and hemicelluloses) as a part of the carbon cycle in nature, and the rate of biodegradation depends on various environmental conditions. Although bacteria can degrade lignified cell walls by extracellular enzymes, the degradation patterns differ distinctly from those caused by most fungi. The true identity of erosion bacteria is very difficult. However, in a recent work of European Commission funded research project, BACPOLES, using DNA technique, they concluded that erosion bacteria probably are a wide group different bacteria species, where some isolated species belongs to the group of Flavobacteria and Cytophaga [1, 33].

CONCLUSION

The objective of this study was to evaluate the benefits of reinforcing polypropylene with naturally woven coconut leaf sheath short fibers. CLS short fiber-reinforced polypropylene composites were prepared by compression molding varying the fiber loading. The incorporation and increase of the fiber content gave rise to considerable increase of tensile, bending strength and stiffness. Both strength and stiffness increased with increased fiber content up to 10 wt% and this was considered an optimum fiber loading. The highest TS, BS, TM and BM values of the optimized composite were found to be 27, 46, 278 and 525 MPa, respectively. However, reinforcement slightly decreased the thermal stability of the composites. The optimized composite retained about 70% of its initial strength and stiffness after six months of soil degradation. Hence, the composites might have potential utilization where lower strength is relatively required.

REFERENCES

[1]   Mohanty A. K, Misra M. and Hinrichsen G., *Macromol Mater Eng,* **2000**, 276/ 277, 1–24.
[2]   Mittal V., Saini R., and Sinha S., *Composites Part B,* **2016**, 99, 425-435.
[3]   Riccieri J. E., De Carvalho L. H. and Vazquez A., *Polym Composite,* **1999**, 20(1), 29–37.
[4]   Woodhams R., Thomas T. G., and Rodgers D. K., *Polym Eng Sci*, **1984**, 24(15), 1166-1171.
[5]   Klason C. and Kubat J., Cellulose in polymer composites, composite system from natural and synthetic polymers, Salmen L. et al. edition, **1986**, Elsevier Science, Amsterdam.
[6]   Kuruvilla J., Siby V., Kalaprasad G., Thomas S, Prasannakumari L and Pavithran C, *Euro Poly J*, **1996**, 32, 1243-1250.
[7]   Oksman K., Wallström L., Berglund L. A. and Filho R. D. T., *J App Polym Sci,* **2001**, 84, 2358-2365.
[8]   De Albuquerque A. C., Kuruvilla J., De Carvalho L. H. and d'Almeida J. R. M., *Comp Sci Tech*, **2000**, 60, 833-844.
[9]   Abdullah-Al-Kafi and Abedin M. Z., *J Reinf Plast Comp,* **2006**, 25, 575-588.
[10] Wisittanawata U., Thanawanb S., and Amornsakchai T., *Polym Test*, **2014**, 35, 20–27.
[11] George J. Sreekala M. S. and Sabu T., *J Reinf Plast Comp*, **1998**, 17, 651-672.
[12] Srinivasana V.S., Boopathya S.R., Sangeethaa D., and Ramnath B.V., *Mater Design*, **2014**, 60, 620–627.

[13] Benítez A. N., Monzón M. D., Angulo I., Ortega Z., Hernández P. M. and Marrero M. D., *Measurement*, **2013**, 46, 1065–1073.

[14] Pothan L. A. Sabu T. and Neelakantan N. R., *J Reinf Plast Comp*, **1997**, 16, 744-765.

[15] Kabir H. Gafur M. A. Ahmed F. Begum F. and Qadir M. R., *Universal J Mater Sci*, **2014**, 2(6), 119-124.

[16] Murali Mohan R. K. and Mohana R. K., *Compos Struct*, **2007**, 77, 288-295.

[17] Mohanty J. R., Das S. N., Das H. C., and Swain S. K., *Polym Composite*, **2013**, 34(6), 959-966.

[18] Rozman H. D., Tay G. S., Kumar R. N., Abubakar A., Ismail H., and Ishak Z. A. M, *Polym Plast Tech Eng,* **1999**, 38, 997-1011.

[19] Abdul K., Hanida S., Kang C. W., and Fuad N. A., *Reinf Plast Comp*, **2009**, 26, 203-218.

[20] Dey K., Sharmin N., Khan R. A., Nahar S., Parsons A. J., and Rudd C. D., *J Thermoplast Compos,* **2011**, 24, 695-711.

[21] Garcia M. Vliet G. V., Jain S., Zyl W. E..V., and Boukamp B., *Rev Adv Mater Sci,* **2004**, 6, 169-175.

[22] Karmaker A. C. and Hinrichsen G., *Polym Plast Technol Eng*, **1999**, 30, 609-621.

[23] Khan M. A., Hinrichsen G., and Drzal L. T., *J Mat Sci Lett*, **2001**, 20, 1211-1713.

[24] Bledzki A. K. and Gassan J., *J Prog Polym Sci*, **1999**, 24, 221-274.

[25] Wambua P., Ivan J., Verport I., *J Comp Sci Tech*, **2003**, 63, 1259–1264.

[26] Czvikovszky T., *Nuclear Instr Methods Phys Res B*, **1995**, 105, 233–237.

[27] Rahman M. M. and Khan M. A., *Compos Sci Tech*, **2007**, 67, 2369-2376.

[28] Poddar P., Asad M., Islam M. A., Sultana S., Nur S. P., and Chowdhury A. M. S., *Adv Mater Sci*, **2016**, 1(2), 1-4.

[29] Mishra S., Mohanty A. K., Drzal L. T., Misra M., Parija S., Nayak S. K., and Tripathy S. S., *Compos Sci Technol*, **2003**, 63(10), 1377.

[30] Cantero G., Arbelaiz A., Llano-Ponte R., and Mondragon I., *Compos Sci Technol*, **2003**, 3, 1247-1254.

[31] Thwe. M. M. and Liao K., *Compos Part A- Applied Sci Manuf,* **2002**, 33, 43–52.

[32] Soykeabkaew N., Supaphol P., and Rujiravanit R., *Carbohy Polym*, **2004**, 58, 53-63.

[33] Björdal C. G., *J Cult Herit.* **2012**, 13(3), 118–S122.