

Toddy Palm (*Borassus Flabellifer*) Fruit Fibre Bundles as Reinforcement in Polylactide (PLA) Composites: An Overview About Fibre and Composite Characteristics

Nina Graupner^{1,*}, Koranat Narkpiban^{2,5}, Thiprada Poonsawat³, Porntip Tooptompong⁴ and Jörg Müssig¹

¹ The Biological Materials Group, Department of Biomimetics, HSB-City University of Applied Sciences, Bremen, Germany, Neustadtswall 30, D-28199 Bremen, Germany.

² Bioproduct Science, Department of Science, Faculty of Liberal Arts and Science, Kasetsart University, 1 Malaiman Rd, Kamphaeng Saen, Nakhon Pathom 73140, Thailand.

³ Department of Botany, Faculty of Liberal Arts and Science, Kasetsart University, 1 Malaiman Rd, Kamphaeng Saen, Nakhon Pathom 73140, Thailand.

⁴ Textile Technology and Innovation Research Unit, Department of Textile Science, Faculty of Agro-Industry, Kasetsart University, 50 Thanon Ngam Wong Wan, Khwaeng Lat Yao, Khet Chatuchuk, Bangkok 10900, Thailand.

⁵ Department of Science and Mathematics, Faculty of Science and Technology, Rajamangala University of Technology Tawan-Ok, Chonburi 20110, Thailand.

*Corresponding Author: Nina Graupner. Email: nina.graupner@hs-bremen.de.

Abstract: Toddy palm fruit have an apparent density below 0.8 g/cm³ and offer an interesting lightweight construction potential in polylactide (PLA) composites reinforced with 37 mass-% fibres. Single fibre bundles show similar mechanical properties compared with coir: tensile strength of 240 MPa, Young's modulus of 3.8 GPa and an elongation at break of 31%. However, density and diameter (~ 50 µm) of fruit fibre bundles are significantly lower. The compression moulded composites have a density of 0.9 g/cm³ and achieved an unnotched Charpy impact strength of 12 kJ/m², a tensile strength of 25 MPa, Young's modulus of 1.9 GPa and an elongation at break of 9%. Due to the high porosity of the composites and the different stress-strain behaviour of fibre and matrix the fibre-reinforcement potential could not be fully used. Maximum stress of the composite was reached at the elongation at break of the PLA-matrix (~2%) while the fibre achieved its maximum stress at an elongation of ~31%. After reaching the maximum stress of the composite, the fibres were pulled out from the matrix with low energy absorption, resulting in a decrease in stress and a limited reinforcement potential. Additionally, the study investigates whether an insect attack by the Asian fruit fly on the mesocarp has a significant influence on the mechanical fibre characteristics. The results have shown that only the rough surface of the fibre bundles is smoothed by insect infestation. The mechanical properties were not significantly affected. For this reason insect-infested fruits of the toddy palm, which are no longer suitable for food production, can be used for the production of sustainable composite materials.

Keywords: Toddy palm fibre; *Borassus flabellifer* fibre; polylactide (PLA) composite; impact strength; fibre/matrix adhesion

1 Introduction

The *Borassus flabellifer* palm belongs to the family *Arecaceae* (alt. *Palmae*), and is also known as toddy palm, sugar palm, doub palm, palmyra palm, tala palm or wine palm. *Borassus flabellifer* is found in coastal areas of India, northern Sri Lanka, and mainland southeastern Asia. The fruit and tuberous seedings are used for food, beverages and sugar. Fibres from the leaf and leaf sheath are used for brushes, septic tank based and clear water filters, doormats, carpets, ropes, chair and sofa cushion [1,2].

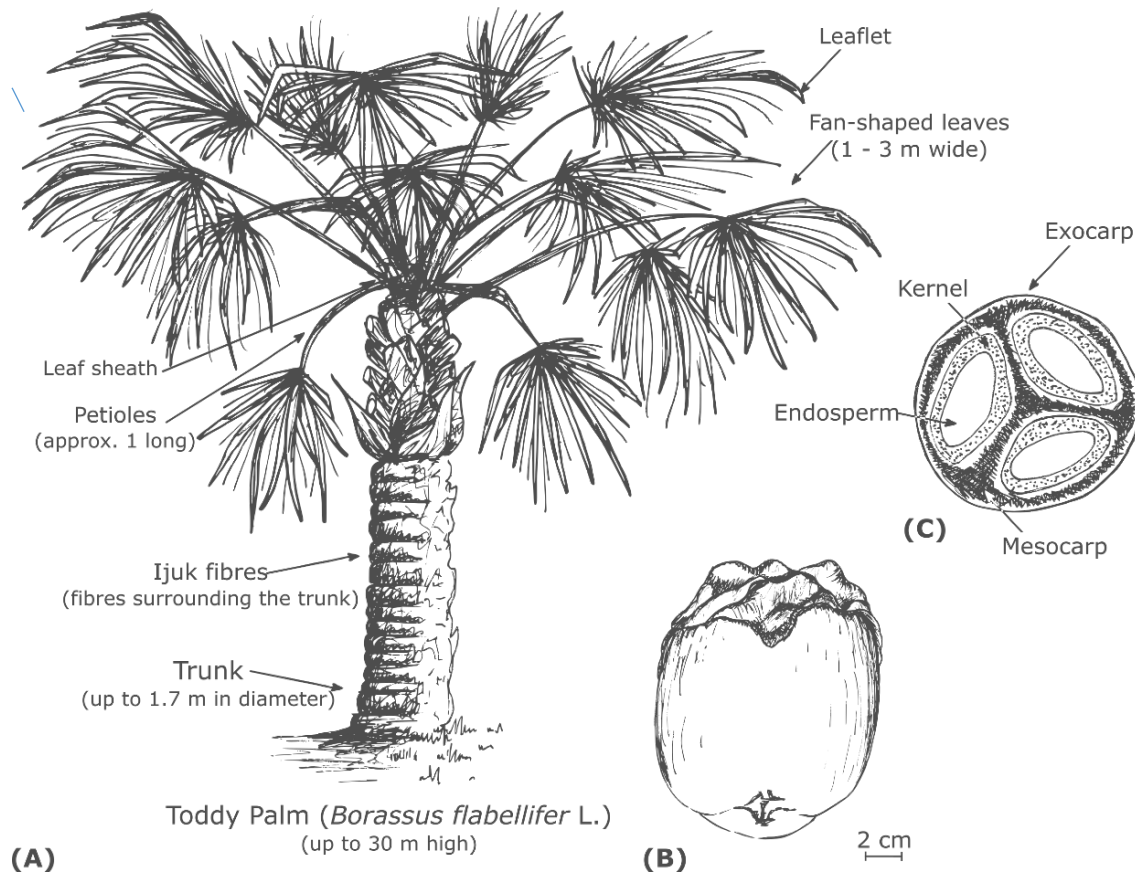


Figure 1: Origin of different toddy palm fibre bundles. A) palm tree, B) toddy palm fruit and C) cross-section of a toddy palm fruit

Fibres from different origin of the toddy palm such as fibres from trunk, leaflet, petiole, fruit, and black sugar palm fibres (ijuk-surrounding the trunk) (see Figs. 1(A) & 1(B)) were already investigated for their mechanical and chemical characteristics, e.g., in studies by Sahari et al. [3,4], Reddy et al. [5] or Ishak et al. [6]. Fibre bundles of different origin show apparent differences in their mechanical characteristics. While fibre bundles from the leaflet and the petiole show high strength and stiffness, ijuk and trunk fibre bundles have clearly lower strength and stiffness values but a higher elongation at break [3,4]. Moreover, Ishak et al. [6] have shown that tensile strength and Young's modulus of fibre bundles extracted from different heights of the trunk (between 1 and 15 m) increase significantly from bottom to top. Moreover, the elongation and toughness increase up to 9-11 m, in height. Above 11 m the values decrease.

Fruit fibre bundles from the mesocarp (see Fig. 1(C)) which are the focus of the present study shall protect the seeds against impact loads. The fruit needs to resist high impact forces when it falls to the ground from heights up to 30 m. Hence, fruit fibre bundles have higher elongation and toughness [5] compared to ijuk or trunk fibre bundles leading to higher energy absorption in the case of applied impact loads. Considering the fruit fibre bundles, Reddy et al. [5,7] found two different fibre types. Fine fibre bundles

which are attached at the fruit shell and coarse fibre bundles which are present edge to edge in the fruit nut. The authors found higher tensile strength and stiffness values with a slightly lower elongation for fine fibre bundles as compared to coarse fibre bundles [5]. This finding may be based on the function of the fibres in nature. A similar phenomenon was found in the coconut pericarp, where fibres of higher strength and stiffness were found in the outer part of the pericarp (exocarp) compared to the inner part (mesocarp). An opposite trend was detected for the elongation at break which increased from the exocarp to the mesocarp structure leading to mechanical gradients improving the energy absorption potential of the fruit when it falls to the ground [8].

Some authors are working on the investigation of the suitability of fibres from different origins of the toddy palm for composite applications. Examples are the works of Sahari et al. [3] who investigated the reinforcement potential of fibres from petiole, trunk and ijuk in unsaturated polyester (UP) resin or Balakrishna et al. [9] and Dhoria et al. [10] who worked on epoxy (EP) composites reinforced with fibres from the petiole. Other examples are the studies of Srinivasababu et al. [11] who investigated fibres from the leaflet in UP, Prabowo et al. [12] and Rashid et al. [13] who analysed ijuk fibre-reinforced polypropylene (PP) and phenolic composites, respectively. Reddy et al. [14], Maheswari et al. [15] and Sarasini et al. [16] examined the characteristics of fruit fibre bundles in combination with EP, high-density polyethylene (HDPE) or polycaprolactone (PCL) for potential composite applications.

The present study focuses on the fruit fibre bundles from the mesocarp, which were processed with a PLA matrix to compression moulded composites. Fruit of the toddy palm contains approximately 2.4% mesocarp fibres corresponding to 66 g when a nut with a total mass of 2,790 g is considered [1]. Due to the low density of toddy palm fibre bundles extracted from the mesocarp of the fruit (0.7 to 0.8 g/cm³; [17,18]) compared to other cellulose-based fibres like bast fibres (density 1.4 to 1.5 g/cm³; [19]) or coir fibres (density of 1.15 to 1.5 g/cm³; [19]) these fibres promise an interesting potential for lightweight composite applications. Additionally, fruit fibre bundles from the toddy palm show a lower diameter of around 120 µm [17] as compared to coir fibre bundles with a fibre diameter between 50 and 460 µm [19]. The resulting higher specific fibre surface of fibre bundles leads to a higher bonding surface between fibre and matrix. Both coir and toddy palm fibre bundles are separated from the thick mesocarp layer at the fruit (compare Fig. 1(C)). When the coconut is ripe, it falls off the palm. The mesocarp is dry, and the colour changes from green to brown. The fruits of the toddy palm fall down at ripening while the mesocarp remains fresh, yellow jelly liked. Both the coconut and the fruit of the toddy palm have a hard inner shell of endocarp, which protects the endosperm and the cotyledon.

This study aims to find out if fruit fibre bundles of the toddy palm are suitable as reinforcement to improve the toughness of a brittle PLA matrix. Since the pericarp of the toddy palm is often attacked by insects like the Asian fruit fly (*Bactrocera sp.*) it is additionally worth to investigate if the mechanical fibre and composite characteristics are affected by the infestation. The fruit flies penetrate the skin of the mature fruit and deposit eggs. The fruit is then damaged by larvae which feed in the fruit, and the fruit is no longer suitable for consumption [20,21]. It should be examined whether the fibres are still usable for composite materials.

2. Materials & Methods

2.1 Fibres and Matrix

Two different kinds of toddy palm fibre bundles were supplied from Kasetsart University (Kamphaeng Saen Campus, Kamphaeng Saen District, Nakon Pathom, Thailand). The mesocarp (fruit) fibre bundles were separated manually with a sharp knife from the endocarp. A water boiling process removed the yellow gum of the jelly-like parenchyma. It was not distinguished between, fine fibre bundles, which are attached at the fruit shell and, coarse fibre bundles, which are present edge to edge in the fruit-nut as described in [5,7]. Fibre bundles from, Toddy palm yellow, (specified in the text as ‘Toddy palm y’) are derived from the ripe mesocarp of the toddy palm fruit. Fibre bundles from, Toddy palm brown’ (specified in the text as ‘Toddy palm b’) come also from a ripe mesocarp but were infested with an insect leading to a darker colour as shown in Fig. 2. It is assumed that the Asian fruit fly (*Bactrocera tau* or *Bactrocera dorsalis*) infest the

fruit. Fibre bundles were carded to separate the agglutinated fibres and bundles from each other with a laboratory carding machine (Shirley Developments Limited SDL, Stockport, UK; serial number 02895) before testing.

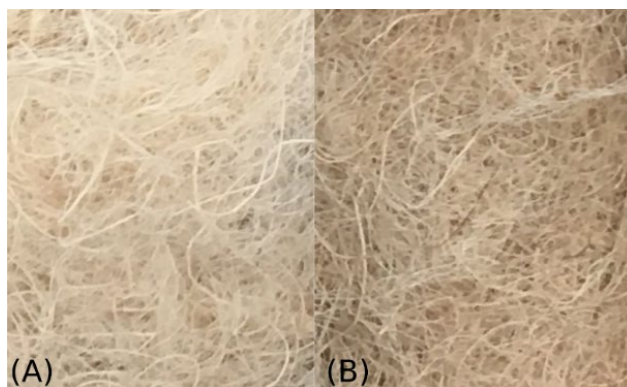


Figure 2: ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles from Thailand

As a matrix polylactide (PLA) staple fibres (type Ingeo SLN 2230E2; Eastern Textile Ltd., Taipei, TW) were supplied with a fineness of 3.0 dtex and a length of 60 mm. Fibres were produced from a NatureWorks™ 6202 D PLA with a density of 1.24 g/cm³ (melting temperature of 160-170°C; glass transition temperature of 60-65°C). The tensile strength of compression moulded PLA was determined to be 52 MPa in a previous study [22].

2.2 Fibre Investigations

Fibre bundles were conditioned before tensile testing and width measurement at least for 18 h in a standard climate according to DIN EN ISO 139 (20°C, 65% relative humidity) [25].

Tensile characteristics were investigated with a Fafegraph M tensile testing machine (Textechno, Mönchengladbach, DE) working with a pneumatic clamping system. A clamp combination of PVC/PMMA was used. 50 fibre bundles were investigated with a load cell of 1000 cN at a gauge length of 20 mm and a testing speed of 10 mm/min. The elongation was determined from the traverse path without compliance correction of the testing machine. The uncorrected Young’s modulus was calculated in the linear-elastic region of the stress-strain curves.

The measurement of the fibre bundle width distribution was performed for 3921 ‘Toddy palm y’ elements and 2551 ‘Toddy palm b’ elements with the image analysis software Fibreshape 5.1.1 (IST AG, Vilters, CH). Fibre bundles were prepared on a flatbed scanner (type Epson Perfection Photo V700, Epson, Meerbusch, DE) and scanned at 2000 dpi resolution in transmitted light mode. The evaluation was done with a measuring mask prepared for long fibres in the thickness range between 10 and 10,000 µm with a zoom factor of 0.71.

The density of Toddy palm fibre bundles was determined using a displacement method [23] with a micro-balance KERN ABT-120-5DM (Kern & Sohn GmbH, Balingen, DE). For the investigations commercially available canola oil was used. The density of the oil was specified by the manufacturer with 0.92 g/cm³ and was checked with a pycnometer (25.2089 ml; Paul Marienfeld GmbH & Co. KG, Lauda Königshofen, DE) at 20°C. The density of the fibres was determined after drying at 105°C for 3h. 3 samples (0.5 g) were analysed for each fibre type.

The chemical composition of both todody palm fibre bundles was exemplary analysed according to Van Soest [24] with an Ankom A200 fibre analyser (ANKOM Technology, Macedon, NY, USA) using a gravimetric analysis technique. The percentual amount of neutral detergent fibre (NDF), acid detergent

fibre (ADF) and acid detergent lignin (ADL) were determined for the calculation of the cellulose (ADF-ADL), hemicellulose (NDF-ADF) and lignin (ADL) content.

2.3 Composite Production

Multilayer webs were manufactured with a laboratory carding machine with a working width of 200 mm (Shirley Developments Limited SDL, Stockport, UK, serial number 02895). Toddy palm fibre bundles and PLA fibres were mixed during the carding process. The initial mass of toddy palm and PLA fibres was calculated based on the dry mass for an aimed fibre mass fraction of 30%, which corresponds to 40% in volume. Multilayer webs were cut to the dimensions of $80 \times 150 \text{ mm}^2$ and were then dried for 2 h at 105°C in a forced air oven (Vötsch VCL 4003, Vötsch Industrietechnik GmbH, Balingen, DE). Composite boards were produced with a compression moulding technique in a self-manufactured small press (press rheometer, $150 \times 80 \text{ mm}^2$) with shearing edges usable for a Zwick/Roell Z020 universal testing machine equipped with a 20 kN load cell (Zwick/Roell GmbH & Co. KG, Ulm, DE). The press plates were heated with heating cartridges to 180°C . The multilayer-webs which had been wrapped in Teflon foil (foil type 0903 AS; Böhme Kunststofftechnik GmbH + Co KG, Schwarzenbek, DE) were heated for a residence time of 10 s with a tool spacing of 10 mm and a temperature of 180°C . Subsequently, a pressure of 1 MPa was applied for 3 min, and finally, the pressure was boosted to 1.1 MPa for 5 seconds. The composites were then removed from the press and cooled between two aluminium plates with a mass of 965 g ($30 \times 12 \times 1 \text{ cm}^3$) at room temperature. The expected plate thickness was 1.6 mm. Due to the carding process, the fibre bundles have a predominant fibre orientation in the machine direction of the carding process. Composites were analysed parallel to the main fibre orientation.

2.4 Composite Testing

Composites were conditioned at least for 18 h in a standard climate according to DIN EN ISO 291 (23°C , 50% relative humidity) [26].

Tensile tests were carried out for 6 test specimens (type CP2 according to DIN EN ISO 20753:2014 [27]; thickness around 1.3 mm) with a universal testing machine type Zwick Z 020 (Zwick/Roell, Ulm, DE) working with a load cell of 500 N and a Zwick/Roell manual clamping specimen holder type 8135/20 N with vulcolan coating (Zwick/Roell GmbH & Co. KG, Ulm, DE). The gauge length was set to 35 mm. After reaching a preload of 0.5 N the test was performed with a speed of 1 mm/min. The elongation was measured with a video extensometer between two measuring marks spaced 10 mm apart (VideoXtens, Zwick/Roell, Ulm, DE). The Young's modulus was calculated between 0.05 and 0.25% elongation via linear regression.

Flexural characteristics for 6 specimens per sample were measured with the Zwick Z020 universal testing machine, using a load cell of 20 kN. The bending device was equipped with a load applicator having a 5 mm radius and bearings of 2 mm radius according to DIN EN ISO 14125 [28]. The test specimen geometry was calculated according to annexe A.1 and A.2 of DIN EN ISO 14125 [28] resulting in dimensions of $30 \times 25 \times 1.3 \text{ mm}^3$. The preload was set to 10 N, the bearing distance to 21 mm, and the testing speed to 1 mm/min. The elongation was calculated from the traverse path of the machine. The flexural modulus was calculated between 0.05 and 0.25% elongation via linear regression.

The unnotched Charpy impact strength was determined with a pendulum impact testing machine (type 5102, Zwick, Ulm, DE) operating with a pendulum hammer of 0.5 J according to DIN EN ISO 179 [29]. In deviation from the standard, 6 test specimens per composite with the dimensions of $80 \times 10 \times 1.4 \text{ mm}^3$ were investigated at a bearing distance of 40 mm. The sample was hit on the flatwise impact direction.

The density of the composites was determined as described for the fibres in a conditioned state (23°C , 50% relative humidity). 3 bending test specimens were analysed for each composite type.

The porosity of the composites was calculated from the difference of the theoretical calculated density based on the fibre and matrix volume fraction and the measured density from the bending test specimens after storage in a standard climate according to DIN EN ISO 291 [26].

2.5 Fibre/Matrix Adhesion

The fibre/matrix adhesion was investigated by microbond testing (see [30,31]). For the specimen preparation single toddy palm fibre bundles were fixed in a metal frame at both ends with adhesive tape (Tesa SE, Hamburg, Germany). Three to five PLA fibres (Ingeo fibres type SLN 2660 D; Eastern Textile Ltd., Taipei, TW) having a fineness of 6.0 dtex were twisted and knotted around the toddy palm fibre bundle. Afterwards, the ends of the PLA-fibres were removed with a razor blade close to the fibre bundle. The samples in the metal frame were wrapped in aluminium foil and heated for 5 min at 185°C in a forced-air oven type BW91270 (Zwick/Roell GmbH, Ulm, DE), and cooled down at room temperature. The result is a tiny PLA-droplet around the fibre. The embedding length was in the range between 170 and 320 µm. After conditioning according to DIN EN ISO 291 [26], the specimens were fixed on a self-manufactured microbond device. The microbond-test was carried out with a testing speed of 1 mm/min at a gauge length of 5 mm with a 5 N load cell at the Zwick/Roell Z020 testing machine. 26 valid readings were obtained for both types of toddy palm fibre bundles in a PLA matrix. The interfacial shear strength (IFSS) was determined from the maximum force divided by the embedded area of the fibre bundle. The interfacial friction (IFF) was calculated from the mean force value after fibre debonding within a traverse path distance of 1 mm divided by the embedded area of the fibre bundle.

2.6 Statistics

The statistical evaluation of the results was done with the open-source R software (<http://www.rproject.org/>). The results were examined by means of a Shapiro-Wilk test for normal distribution. To find out if there are significant differences between ‘Toddy palm y’ and ‘Toddy palm b’ data, for normally distributed data with homogenous variances the Tukey-test was used and for data which were not distributed normally the Wilcoxon test was chosen. All tests were performed with a level of significance $\alpha = 0.05$. Results are presented as Box-Whisker plots showing the median, the 2nd (Quartile 25) and 3rd quartile (Quartile 75) and 1.5 times the interquartile length (Whiskers). Significant differences within one series of experiments are marked with different letters above the plots, and an asterisk indicates results which are not distributed normally. Since not all results are normally distributed, the text refers to the median values, for all series of measurements with a higher number of samples (n) than 3, unless otherwise described.

3 Results and Discussion

3.1 Fibre Characteristics

As shown in Tab. 1 the measured fibre characteristics such as width, tensile properties and density do not differ significantly between ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles. Concerning the reinforcement potential for the use in composite materials, both types of fibres have a comparable potential from a mechanical and morphological point of view, and the infestation of the fruit by the Asian fruit fly has no significant influence on the investigated fibre properties itself.

When considering the fibre bundle width in Tab. 1, it is noticeable that the deviation between the mean and median value is huge. This difference indicates that very coarse fibre bundles were included in the measurement, which led to a significantly higher mean value. These results are in agreement with optical investigations using SEM micrographs taken from some cross-sections of toddy palm fruit fibre bundles (see Fig. 3(A)).

The low density values of the fibre bundles (0.772-0.791 g/cm³) promise an excellent lightweight construction potential for the use in composite materials. These low density values of 0.778-0.800 g/cm³ were also reported by Saravanan et al. [17] and Kini et al. [18]. Compared to ijuk fibre bundles and fibre bundles from the trunk, the density of fruit (mesocarp) fibre bundles is significantly lower (compare Tab. 2).

Comparing the measured mechanical characteristics of untreated fruit fibre bundles (Tab. 1) with data from the literature in Tab. 2 strength values of ‘Toddy palm y’ and ‘Toddy palm b’ are clearly higher. The measured Young’s moduli are in the range of values reported by Sarasini et al. [16]. The elongation at break is similar to the data described in studies by Reddy et al. [5,7] or Saravanan et al. [17]. But the width of fruit fibre bundles investigated in the present study is clearly lower than reported in other studies. The smaller width value could be based on the carding process which leads to a refinement of coarse fibre bundles.

Table 1: Characteristics of ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles (width, tensile strength, uncorrected Young’s modulus, elongation at break and density as mean values with standard deviation (std), median value and the number of tested elements (n))

	Mean	Std.	Median	n
<i>Width in μm</i>				
‘Toddy palm y’	53.9	36.8	31.9	3921
‘Toddy palm b’	49.8	32.6	29.4	2551
<i>Tensile strength in MPa</i>				
‘Toddy palm y’	261	124	277	50
‘Toddy palm b’	224	69	221	50
<i>Uncorrected Young’s modulus in MPa</i>				
‘Toddy palm y’	3781	1834	3445	50
‘Toddy palm b’	3955	1312	3690	50
<i>Elongation at break in %</i>				
‘Toddy palm y’	33	13	37	50
‘Toddy palm b’	30	10	32	50
<i>Density in g/cm^3</i>				
‘Toddy palm y’	0.791	0.072	0.773	3
‘Toddy palm b’	0.772	0.012	0.769	3

Toddy palm fibre bundles show similar stress-strain behaviour as described for coir or oil palm mesocarp fibre bundles, and are in the range of the ascribed strength and modulus values (Tab. 2; [32,33]). Compared with coir fibre bundles (Fig. 3(C)) fruit fibre bundles from toddy palm show a similar structural arrangement (compare Figs. 3(A) & 3(B)): the bundle is composed of individual cells and the cross-section shows a relatively round shape. Figs. 3(D) & 3(E) show the typical longitudinal view of the surface of a toddy palm fruit fibre bundle. The unit cells run longitudinally with more or less parallel orientations in the form of shallow longitudinal cavities, similar as described for coir fibre bundles [34]. But the chemical composition is different. As shown in Tab. 3 fruit fibre bundles from toddy palm contain a clearly higher amount of cellulose and hemicellulose while coir fibre bundles have a higher lignin content which may have an influence on the fibre/matrix adhesion [38,39]. The mechanical characteristics of fruit fibre bundles from toddy palm are in a similar range as compared to coir fibre bundles with a higher fineness and a obviously lower apparent density.

Table 2: Characteristics of fibre bundles from different parts of the Toddy palm cited in literature

Origin	Tensile strength in MPa	Young's modulus in MPa	Elongation at break in %	Diameter (width) in μm	Density in g/cm^3	Annotation	References
Trunk	16-292	490-3370	6-28			Investigation between 1 and 15 m height of the trunk	[6]
Trunk	198	3100	30	596	1.12		[3,4]
ijuk	277	5860	22	221	1.20		[3,4]
Leaflet	421	10400	10	115	0.49		[3,4]
Leaflet	98	1170			0.73		[11]
Petiole	365	8600	13	255	0.51		[3,4]
Fruit	82		43			Fine bundle	[5]
Fruit	48	4912	46			Coarse bundle	[5]
Fruit	71	10800	35	120		Fine bundle	[7]
Fruit	65	4918	47	120			[14]
Fruit	24.3		26	120	0.78-0.80		[17]
Fruit	65	4918	47	130		Fine bundle	[15]
Fruit	51	1221	41	280		Coarse bundle	[15]
Fruit	150			101	0.80		[18]
Fruit	101-120	2720-3840		119-130		Influence of gauge length	[16]
Coir	95-270	2800-6000	15-51	50-460	1.15-1.50	Different references	[19]
Oil palm mesocarp	259-373	3867-4844	10-21			Influence of different testing speed	[33]

Table 3: Cellulose, hemicellulose and lignin content of fruit fibre bundles from toddy palm compared to coir fibre bundles

Component	Toddy palm fruit fibre bundle	References	Coir fibre bundle	References
α -Cellulose in %	45.7-53.4	[7,15]	32.7-39.3	[35,36,37]
Hemicellulose in %	29.6-32.8	[7,15]	2.0-22.6	[35,36,37]
Lignin in %	17.0-21.5	[7,15]	42.1-59.4	[35,36,37]

Comparing the chemical composition of 'Toddy palm y' and 'Toddy palm b' fibre bundles we see no clear differences regarding cellulose, hemicellulose, lignin and an ash content of the fibres indicating a non-significant influence of the insect attack on the chemical fibre composition (see Tab. 4).

Table 4: Measured chemical composition of ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles

Sample	Cellulose	Hemicellulose	Lignin	Ash
‘Toddy palm y’	50.15	21.80	8.33	0.90
‘Toddy palm b’	51.42	22.40	10.04	1.10

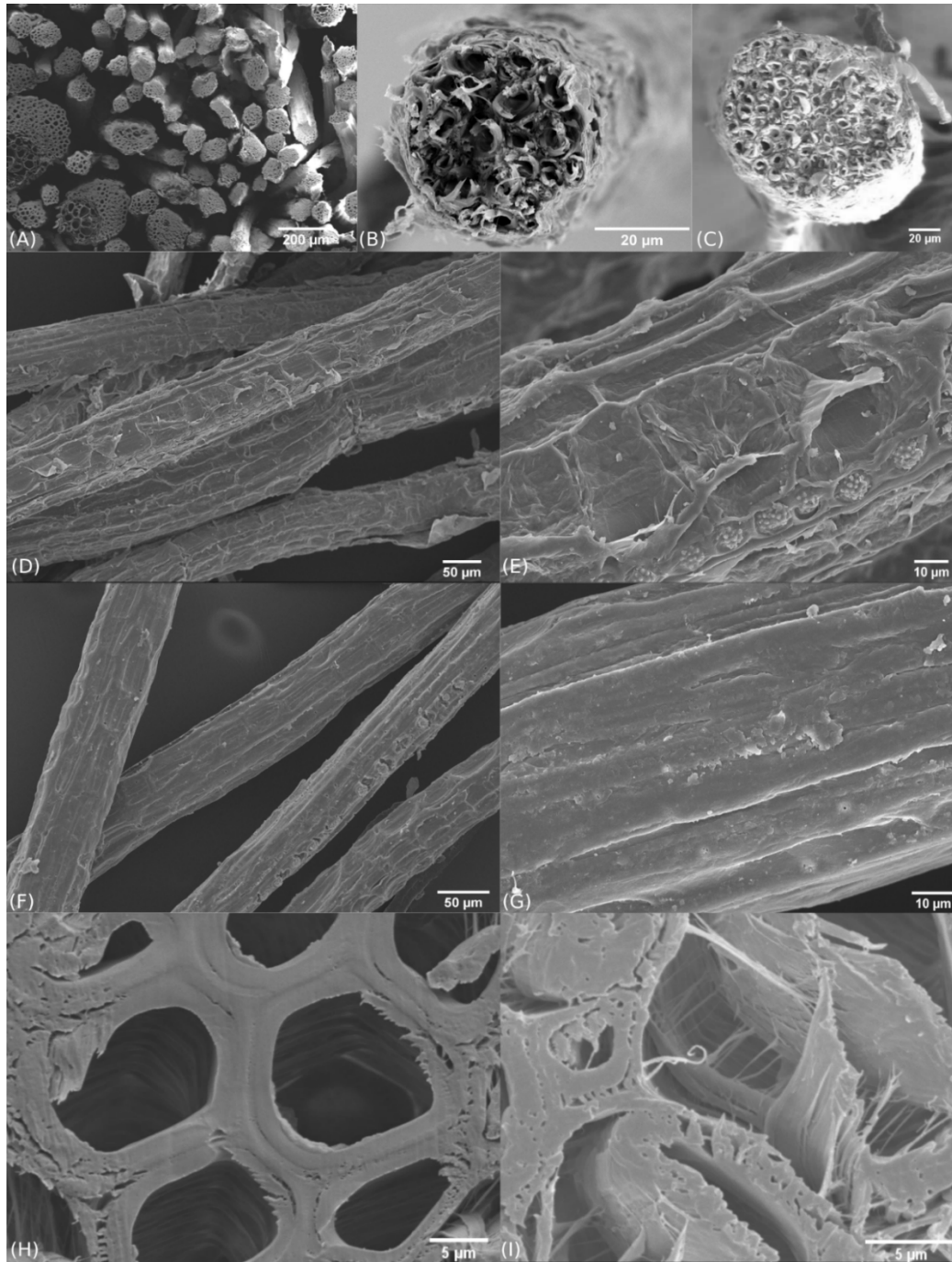


Figure 3: SEM micrographs of the cross-section of fruit fibre bundles from a toddy palm (A & B) and a coir fibre bundle (C) as well as the longitudinal view of ‘Toddy palm y’ fruit fibre bundles in overview (D) and detail (E) and ‘Toddy palm b’ fibre bundles in overview (F) and detail (G). (H) and (I) show the detailed cell wall structure (cross-section) of ‘Toddy palm y’ and ‘Toddy palm b’

3.2 Composite Characteristics

The compression moulded PLA composites were analysed by tensile, flexural and impact tests, and microbond tests were used to investigate the interfacial shear strength (IFSS). Noteworthy is the low density of the composites which was determined to be 0.90-0.92 g/cm³, which confirms the excellent potential of the fibres for lightweight construction.

The PLA composites produced from ‘Toddy palm y’ and ‘Toddy palm b’ fibres show no significant differences regarding their tensile and flexural properties (see Fig. 4). On the trend, the strength and elongation at break values from tensile and flexural tests show somewhat lower values for the samples reinforced with ‘Toddy palm b’ fibre bundles. The same (non-significant) trend has been already demonstrated for the tensile properties of the fibre bundles (Tab. 1). The tendency of lower elongation/ductility of ‘Toddy palm b’ fibres is also visible for the unnotched Charpy impact strength values of the composites (Fig. 5). The values are slightly lower as compared to ‘Toddy palm y’/PLA; however, from a statistical point of view, these differences are not significant.

In addition to the slightly lower strength of ‘Toddy palm b’ compared to the ‘Toddy palm y’ fibres, a significantly lower IFSS was determined in a PLA matrix (Fig. 6). The lower apparent IFSS may be based on the surface roughness of the toddy palm fibre bundles. As shown in Figs. 3(D) & 3(E) ‘Toddy palm y’ fibre bundles have a rougher surface compared to ‘Toddy palm b’ fibre bundles (Figs. 3(F) & 3(G)). The rougher surface influences the interfacial friction (IFF) which was determined after fibre debonding. ‘Toddy palm y’ resulted in a value of 3.2 ± 1.8 MPa while ‘Toddy palm b’ has shown a value of 2.7 ± 1.4 MPa. That means that the insect attack may lead to a smoother fibre surface without changing the overall chemical composition (Tab. 4).

Moreover, the adhesion of the individual cells within the fibre bundle could have an impact on the mechanical fibre and composite characteristics. As shown in Fig. 3(I), the middle lamellae in the ‘Toddy palm b’ fibre bundles is partially dissolved and porous. As a result, under tensile and bending load, individual cells may slide easily. However, in combination with the lower IFSS and the slightly lower strength of the fibre bundles a lower strength of the composites may be the result. The IFSS was measured with 8.4 MPa for ‘Toddy palm y’/PLA and 6.5 MPa for ‘Toddy palm b’/PLA (Fig. 6). These values are clearly higher compared to toddy palm fruit fibre bundles reinforcing a cashew nut shell liquid epoxy matrix which was determined in the range between 0.12 and 0.15 MPa [18], coir fibre bundles in a PP and a MAPP matrix with values of 2.4 and 5.6 MPa measured with microbond-tests [40], and coir in polyester (1.48 MPa) or in polystyrene (1.35 MPa) determined with a pull-out test [41]. But it should be noted that, e.g., bast fibre bundles usually exhibit a higher IFSS in a PLA matrix determined with a microbond or pull-out-test. Examples are values of 11.3 MPa for hemp/PLA [42], 15.3 MPa for flax/PLA [43] or 10.7 MPa for kenaf/PLA [44].

As visible from Fig. 4(D) the flexural strength is more than double as high (by factor 2.1) as compared to the tensile strength (Fig. 4(A)). A similar effect was observed by Duan et al. [45] for a coir fibre-reinforced PLA composite with 30 mass-% fibres (factor 2.3). A possible explanation for the different behaviour under tensile and flexural load is given by Wisnom [46] for carbon fibre-reinforced composites. Wisnom [46] pointed out that strength is assumed to be controlled by statistically distributed critical defects. A smaller volume of material is subjected to the maximum stress in a bending test than in a tensile test leading to a lower probability of critical defects which may lead to failure. Higher strength in bending can, therefore, be explained without the need to assume catastrophic failure initiating from a critical defect. However, composites are usually able to withstand quite large amounts of damage before they break entirely. The flexural failure of unidirectional carbon fibre-reinforced epoxy is usually a gradual process, with collectives of fibres breaking first at the surface and then progressively through the sample. Very often, the specimen does not actually break at all but just becomes very flexible due to the reduced effective thickness [46]. It is assumed that this effect is also present in toddy palm fibre-reinforced PLA composites. Due to the low thickness of the composites (1.3 to 1.4 mm), no complete breakage of the bending samples was found as compared to the tensile test specimens. Another aspect which may lead to higher flexural

strength is the non-linear stress transfer in the sample leading to an overestimation of the flexural strength in plastic based samples [47].

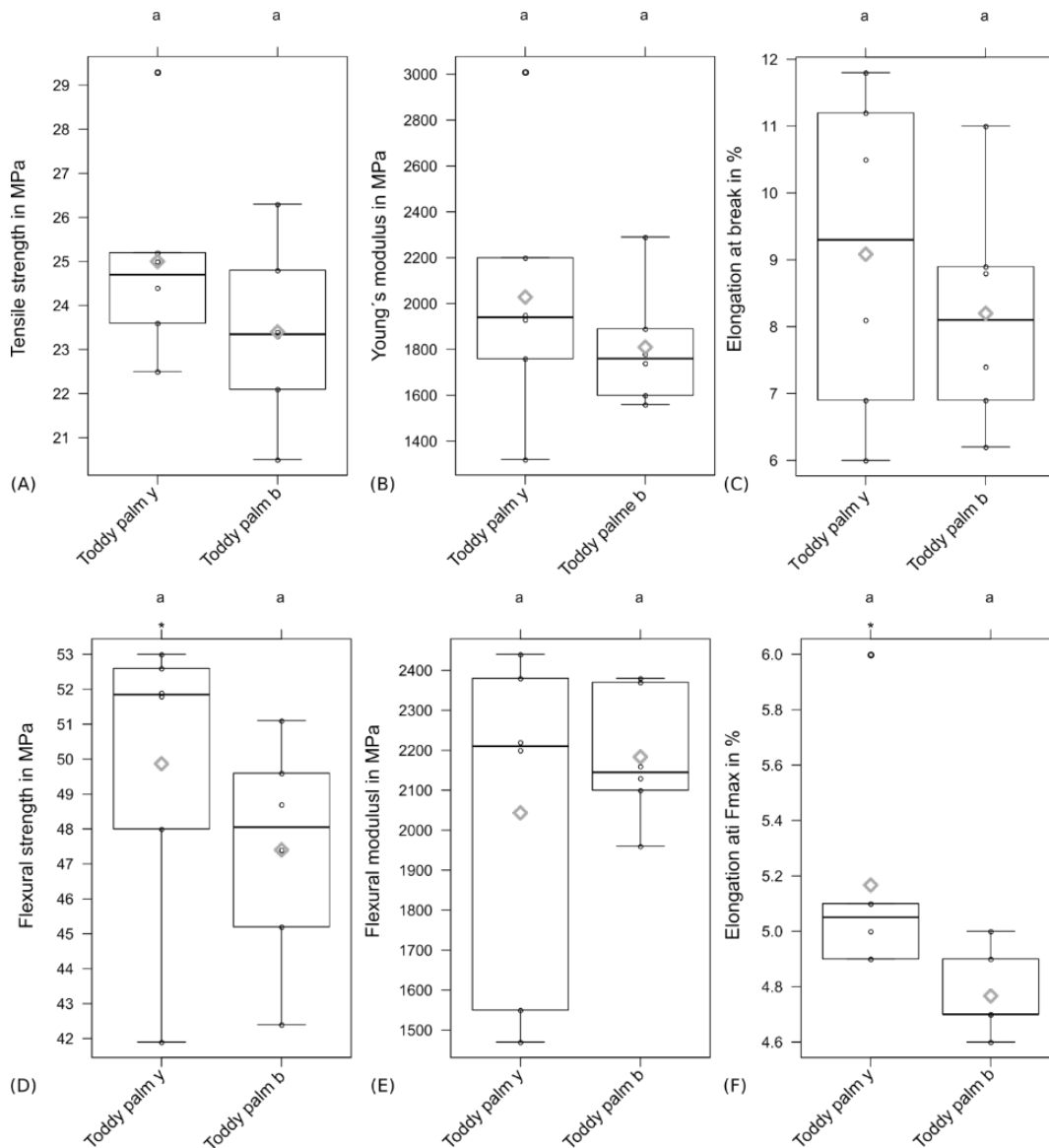


Figure 4: Tensile characteristics (tensile strength (A), Young's modulus (B) and elongation at break (C)) and flexural characteristics (flexural strength (D), flexural modulus (E) and elongation at maximum load (F)) of 'Toddy palm y' and 'Toddy palm b' fibre bundle/PLA composites. Results are shown as Box-Whisker-plots with mean values as rhombuses; an asterisk shows results which do not follow a normal distribution; different letters indicate significant differences

Table 5: Characteristics of composites reinforced with fibre bundles from different parts of the toddy palm cited in the literature (CM means compression moulded composites, IM means injection moulded composites)

Composite	Fibre fraction in volume (vol) or mass %	Manufacturing process	Fibre orientation	Tensile strength in MPa	Young's modulus in MPa	Elongation at break in %	Impact strength in kJ/m ²	Reference
Petiole/UP	30 vol	CM	random	13	430	5	7 kJ/m ²	[3]
Petiole/EP	20 vol	Hand lay up	random	14-23		2-17		[9]
Petiole/EP	30 vol	Hand lay up	random	16-23		3-19		[9]
Petiole/EP	40 vol	Hand lay up	random	17-27		2-16		[9]
Leaflet/UP	30 vol	CM	random	15	390	8	8 kJ/m ²	[3]
Leaflet/UP	12 vol	Hand lay up	UD	50	850		17 kJ/m ²	[11]
Leaflet/UP	20 vol	Hand lay up	UD	38	700		21 kJ/m ²	[11]
Leaflet/UP	27 vol	Hand lay up	UD	42	1000		40 kJ/m ²	[11]
Leaflet/UP	33 vol	Hand lay up	UD	45	850		57 kJ/m ²	[11]
Leaflet/UP	5 vol	Hand lay up	UD	37	550			[10]
Leaflet/UP	10 vol	Hand lay up	UD	40-42	680-700			[10]
Leaflet/UP	20 vol	Hand lay up	UD	42-48	700-730			[10]
Leaflet/UP	22 vol	Hand lay up	UD	62-71	710-790			[10]
Leaflet/UP	35 vol	Hand lay up	UD	73-85	1000-1130			[10]
Trunk/UP	30 vol	CM	random	10	560	3	4 kJ/m ²	[3]
Ijuk/UP	30 vol	CM	random	11	470	4	5 kJ/m ²	[3]
Ijuk/EP	10 vol	Hand lay up	n.s.	43	3330	1	47 J/m	[2]
Fruit/EP	4 mass	Hand lay up	random	42	2374			[14]
Fruit/EP	8 mass	Hand lay up	random	44	2560			[14]
Fruit/EP	12 mass	Hand lay up	random	46	2768			[14]
Fruit/EP	16 mass	Hand lay up	random	48	2926			[14]
Fruit/EP	20 mass	Hand lay up	random	46	2754			[14]
Fruit/EP	24 mass	Hand lay up	random	44	2517			[14]
Fruit/HDPE	5 mass	IM	random	20-22	537-604	156-166	80-86 J/m	[15]
Fruit/HDPE	10 mass	IM	random	21-23	548-650	165-183	86-97 J/m	[15]
Fruit/HDPE	15 mass	IM	random	21-25	541-663	167-197	83-105 J/m	[15]
Fruit/HDPE	20 mass	IM	random	20-23	535-638	172-214	80-95 J/m	[15]
Fruit/PCL	10 mass	IM	random	17	395	450		[16]
Fruit/PCL	20 mass	IM	random	17	580	100		[16]
Fruit/PCL	30 mass	IM	random	18	700	80		[16]
Coir/PLA	30 mass	CM	random	7.5	350	3.2		[50]
Coir/PLA	30 mass	CM	random	38	1520			[45]

Tab. 5 gives an overview of composite tensile properties based on different matrices reinforced with fibre bundles of different parts of the toddy palm reported in the literature. Compared to compression moulded composites made from petiole, leaflet and ijuk fibre-reinforced unsaturated polyester (UP) composites [3], 'Toddy palm y' and 'Toddy palm b' fibre-reinforced PLA composites show considerably higher tensile strength and modulus as well as a higher unnotched Charpy impact strength. The measured data are in the range of petiole fibre-reinforced epoxy (EP) [9]. Unidirectional arrangement of leaflet fibre bundles in a UP matrix resulted in significantly higher strength values with a lower tensile modulus compared to toddy palm/PLA composites [10,11]. Compared to studies dealing with fruit fibre-reinforced

thermoplastics, the strength values of ‘Toddy palm y’/PLA and ‘Toddy palm b’/PLA are on the level of toddy palm fruit fibre-reinforced high density polyethylene (HDPE) [15] and slightly above the values of fruit fibre-reinforced polycaprolactone (PCL) [16] while Young’s moduli of toddy palm/PLA composites are significantly higher. Reddy et al. [14] achieved higher values for fruit fibre-reinforced EP compared to our toddy palm/PLA composites. Since the ‘Toddy palm y’ and ‘Toddy palm b’ have significantly higher strength values than the fruit fibres investigated by Reddy et al. [14] (compare Tab. 1 and Tab. 2), the lower strength of the composites is assumed to be based on the one hand on the manufacturing process and on the other hand on the combination of the ductile toddy palm fruit fibre bundles and the brittle PLA matrix. Due to the high elongation of the mesocarp fibres (~31%) and the low elongation of the PLA matrix (~2%), the potential reinforcement effect of the fibre cannot be used to the full extent (compare [22] for ductile regenerated cellulose fibres in a PLA matrix). ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles reach their ultimate strength at an elongation above 30% (compare Fig. 8). However, the composites fail already at an elongation of 8-9%, and the maximum stress is already achieved at ~2% elongation. Fig. 8 illustrates that the elongation at break of the fibre exceeds the elongation at break of the PLA matrix; the elongation at break of the composite increases significantly by the use of ductile fibres compared to the neat PLA matrix but it can also be seen that the strength of the composite is reached at an elongation only slightly higher than the elongation at break of the PLA matrix. Afterwards, the stress drops considerably down. At this point, the matrix is already broken and the fibre bundles are elongated and pulled out from the matrix (compare Fig. 7). Due to the insufficient fibre/matrix adhesion, a stress transfer from the matrix to the fibre is no longer possible. As a result, the full reinforcement potential of the fibre bundles cannot be used. Future work should focus on the improvement of the fibre/matrix adhesion and/or the use of a matrix having a higher elongation for better use of the reinforcement potential of the fibre.

To evaluate the reinforcing potential of the toddy palm fruit fibre bundles, a modelling approach developed for ductile regenerated cellulose fibres in a PLA matrix according to Graupner et al. [48] was applied to the toddy palm/PLA composites. Eq. (1) depicts this model with the composite strength σ_C in MPa, the fibre length efficiency factor η_L , the fibre orientation factor η_O , the agglomeration factor η_a , the stress value of the fibre at the elongation where the tensile strength of the composite is achieved σ_{Fcom} in MPa, the fibre Volume fraction V_F , the tensile strength of the matrix σ_M and the volume fraction of the matrix V_M . The mass-based calculated fibre volume fraction of ‘Toddy palm y’/PLA and ‘Toddy palm b’/PLA was determined to be 40.2 and 40.8%, respectively, for a composite with perfect compaction and the intended dimensions of $150 \times 80 \times 1.6 \text{ mm}^3$. However, the measurement of the dimensions of the specimens has shown that a mean plate thickness of 1.3 mm was achieved due to the manufacturing process in a compression moulding tool with shearing edges. Thus, the fibre volume fraction is with 46.7% for ‘Toddy palm y’ and 47.8% for ‘Toddy palm b’ (fibre mass fraction of around 37%) considerably higher than expected. Due to the lack of spacers, the matrix has leaked out, and the fibre volume fraction has increased due to the smaller plate volume.

σ_{Fcom} was determined to be 72.1 MPa for ‘Toddy palm y’ and 70.1 MPa for ‘Toddy palm b’ fibre bundles, respectively. Due to the high fibre length $> 50 \text{ mm}$ η_L was set to 1, η_O was taken from a previous study [22,48] for 30 mass-% lyocell 15.0 dtex fibre-reinforced PLA (0.7618). The agglomeration factor is calculated according to El-Sabbagh et al. [49] with $\eta_a = 1 - ((1/0.78) * V_F)^2$ and results in 0.642 for ‘Toddy palm y’/PLA and 0.624 for ‘Toddy palm b’/PLA. The agglomeration factor is dependent on the processing method and the fibre volume fraction. The probability of agglomerates and pores in the composite generally increases with an increase in fibre volume fraction.

Using the calculated fibre volume fraction of 46.7% for ‘Toddy palm y’ and 47.8% for ‘Toddy palm b’ it turns out that the calculated strength (44.1 and 43.0 MPa for ‘Toddy palm y’/PLA and ‘Toddy palm b’/PLA) is by factor 1.8 higher than the measured strength. Thus, the potential of the fibres was not utilised, as illustrated in Fig. 8. The considerable deviations can be attributed to the suboptimal fibre/matrix adhesion and to the high porosity of the samples. The porosity is considered in the agglomeration factor in Eq. (1). However, the porosity was determined with 10.7% for ‘Toddy palm y’/PLA and 8.5% for ‘Toddy palm b’/PLA and is significantly higher than that of the investigated lyocell/PLA samples ($< 5\%$) from the

previous study [22,48]. It is expected that the strength can be significantly increased by the reduction of voids and the improvement of fibre/matrix adhesion by optimising the processing procedure. A better fibre/matrix adhesion could cause a better stress transfer from the matrix to the fibre. Another possibility is for the improvement of the stress transfer from the matrix to the fibre the use of a tougher matrix. The use of a tougher matrix may enhance the elongation at maximum stress of the composite and should cause the maximum stress of the fibre to be reached at higher elongation, resulting in higher composite strength.

$$\sigma_C = \eta_L \cdot \eta_O \cdot \eta_\alpha \cdot \sigma_{FCom} \cdot V_F + \sigma_M \cdot V_M \quad (1)$$

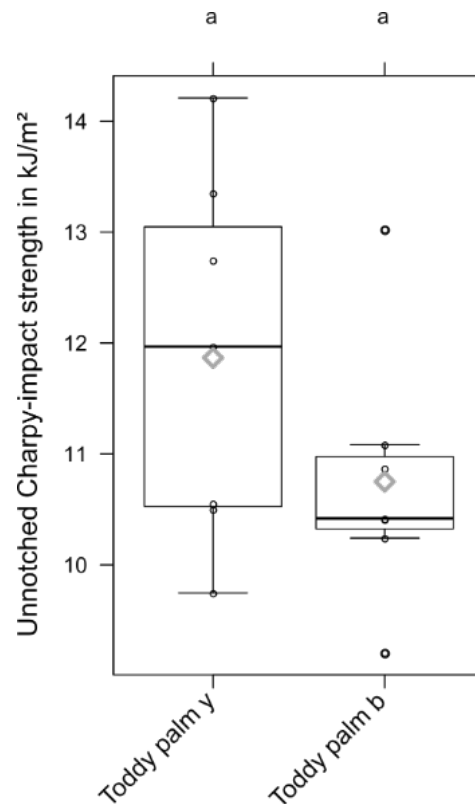


Figure 5: Unnotched Charpy impact strength of ‘Toddy palm y’ and ‘Toddy palm b’ fibre / PLA composites. Results are shown as Box-Whisker-plots with mean values as rhombuses; all results are distributed normally and show no significant differences

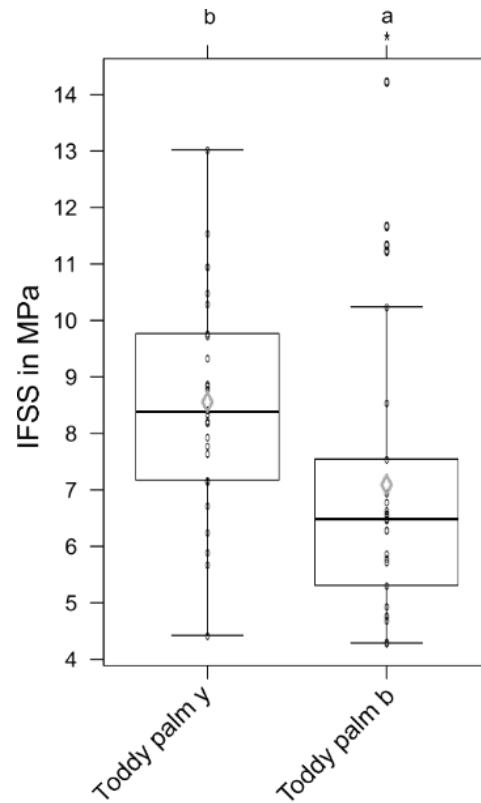


Figure 6: Interfacial shear strength of ‘Toddy palm y’ and ‘Toddy palm b’ fibre bundles embedded in a PLA matrix. The IFSS was measured with a microbond-test. Results are shown as Box-Whisker-plots with mean values as rhombuses; an asterisk shows results which do not follow a normal distribution; different letters indicate significant differences

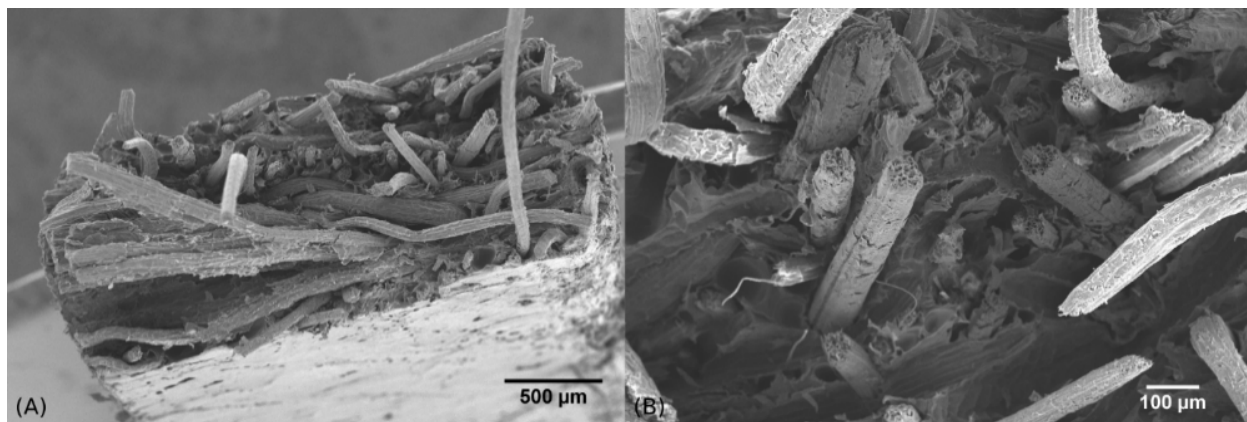


Figure 7: Tensile fracture surface of a ‘Toddy palm y’ fibre-reinforced PLA composite

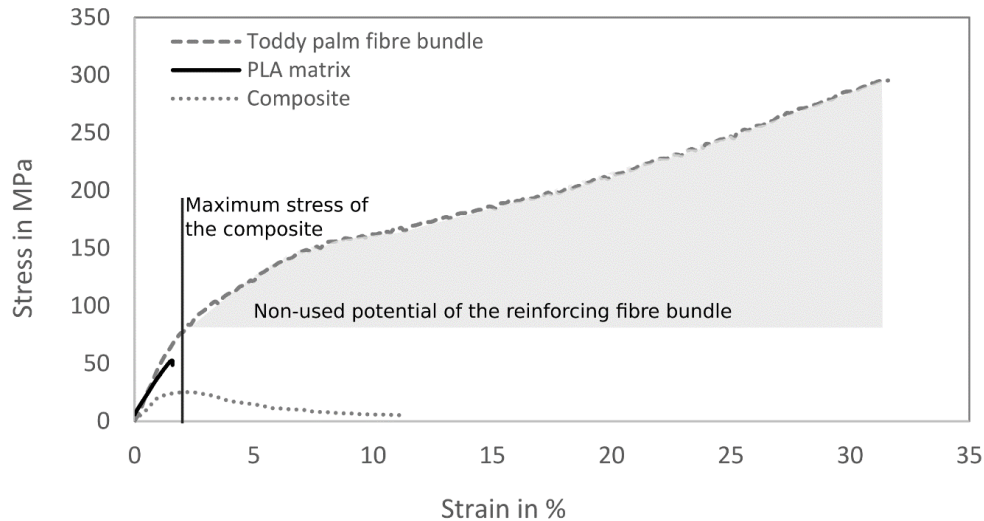


Figure 8: Stress-strain curves of neat PLA, a toddy palm fibre bundle and the composite reinforced with 47 vol.-% fibres

4 Summary and Conclusions

It has been demonstrated that fruit fibre bundles from the mesocarp of the toddy palm fruit show comparable characteristics to fibre bundles from the mesocarp of the oil palm fruit and the coconut. An infestation of the toddy palm fruit by fruit flies does not seem to have significant negative influences on the fibre properties. This means that infested fruits that are no longer usable for food products can be used for composite materials. With very high toughness, the fibre bundles from the mesocarp of the toddy palm have a very low density, resulting in a good potential for lightweight construction. The high toughness of the fibre bundles could not be transferred to the PLA composite, and the reinforcement potential of the fibre could not be achieved. The high porosity of the composites has a negative effect on the fibre/matrix adhesion, and thus on the load transfer from the matrix to the fibre as well as on the energy absorption under impact load. Besides, the low elongation of the brittle PLA matrix (~2%) prevents the maximum stress of the fibre could be reached. The fibre bundles were already debonded from the matrix at low stress. During the tensile tests, it was found that fibre pull-outs considerably delayed the complete failure of the samples while the brittle PLA matrix was already broken. The maximum load of the composite was reached too early so that the reinforcement potential of the fibre bundle could not be achieved. In future research, the fibre/matrix adhesion, as well as the manufacturing process, should be improved to reduce the void content. In addition, the use of a more ductile matrix should be tested in order to exploit the maximum stress of the fibres in the composite. After optimising the composite structure and the fibre/matrix adhesion, it is assumed that a lightweight material with a very interesting impact performance can be produced.

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