Sains Malaysiana 48(1)(2019): 145–154
http://dx.doi.org/10.17576/jsm-2019-4801-17

Revisiting the Morphology, Microstructure, and Properties of Cellulose Fibre from Pineapple Leaf so as to Expand Its Utilization

(Mengkaji Semula Morfologi, Mikrostruktur dan Sifat Serat Selulosa daripada Daun Nanas untuk Memperluaskan Penggunaannya)

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ABSTRACT

Pineapple leaf waste is an agricultural product that is available in large quantities and is still under-utilized. Therefore, the aim of this work was to investigate the morphology, microstructure, and mechanical properties of pineapple leaf fibre (PALF) such that its full potential may be realized. Pineapple leaf, its fibre bundles and elementary fibres have been investigated. Morphology, size, and mechanical properties of fibre bundles extracted from different parts (i.e. bottom, middle and top) of a leaf were studied. It was found that the PALF obtained from vascular tissue and from the mesophyll have different macroscopic shapes. Both, however, contain micron-size elementary fibres of similar size and shape. Size and properties of fibre bundles change from the bottom end of a leaf toward the top end. Pineapple leaf microfibre (PALMF) was found to be smaller in diameter than other natural fibres. It is also very long and its structure changes according to its position along the leaf. At the bottom end a clear and large central hole or lumen can be observed. At the top the lumen becomes almost undetectable. The mechanical strength of PALMF appears to decrease, albeit very slightly, toward the tip of the leaf. The mechanical properties of the fibres are relatively high and comparable to that of flax and hemp fibres which are widely studied and used as reinforcing materials in composites. Very long microfibre can easily be obtained from fibre bundles by dissolving the binding matrix. Potential applications for this microfibre are suggested.

Keywords: Cellulose microfibre; elementary fibre; high aspect ratio fibre; natural fibre; pineapple leaf fibre

ABSTRAK

Sisa daun nanas adalah satu produk pertanian yang tersedia dalam kuantiti yang besar dan masih kurang dimanfaatkan. Oleh itu, tujuan kajian ini adalah untuk mengkaji morfologi, mikrostruktur dan sifat mekanik serabut daun nanas (PALF) supaya potensi penuhnya boleh dicapai. Daun nanas, berkas serabut dan unsur serabut telah dikaji. Morfologi, saiz dan sifat mekanik berkas serabut yang diekstrak daripada bahagian yang berbeza (bahagian bawah, tengah dan atas) daun telah dikaji. Didapati bahawa PALF yang diperoleh daripada tisu vaskular dan mesofil mempunyai bentuk makroskopik yang berbeza. Kedua-duanya, bagaimanapun mengandungi unsur serabut saiz mikron dengan saiz dan bentuk yang sama. Saiz dan sifat berkas serabut berubah dari hujung bawah daun hingga ke atas. Mikroserabut daun nanas (PALMF) didapati lebih kecil diameternya daripada serabut semula jadi lain. Ia juga sangat panjang dan strukturnya berubah mengikut kedudukannya di sepanjang daun. Di bahagian bawah, lubang pusat yang jelas dan besar atau lumen dapat diperhatikan. Di bahagian atas pula, lumen hampir tidak dapat dikesan. Kekuatan mekanik PALMF dilihat berkurangan, walaupun sedikit, ke arah pucuk daun. Sifat mekanik serabut ini agak tinggi dan setanding dengan serabut flaks dan hem yang dikaji secara meluas dan digunakan sebagai bahan pengukuh komposit. Mikroserabut yang panjang boleh diperoleh dengan mudah daripada unsur serabut dengan melarutkan matriks mengikat. Potensi aplikasi untuk mikroserabut ini adalah dicadangkan.

Kata kunci: Mikroserabut selulosa; nisbah serabut aspek tinggi; serabut daun nanas; serabut semula jadi; unsur serabut

INTRODUCTION

Natural fibres have been widely used in various applications, especially as reinforcement for polymer matrix composites (Bledzki & Gassan 1999; Faruk et al. 2012; Wirawan et al. 2009). This is because of their lower environmental impact, lower carbon footprint and embodied energy, renewability and carbon storage potential (Pervaiz & Sain 2003). Although all natural fibres are cellulosic materials, different natural fibres have different properties. This is due mainly to their cellulose content and micro-fibrillar angle (Akin et al. 2010). The size and shape of the elementary fibres are also different (Bos et al. 2002). Pineapple leaf fibre (PALF) is a potentially significant natural fibre as it can be obtained easily from pineapple leaf waste and it has excellent mechanical properties. PALF has been studied as a reinforcement in polymer composites (Asim et al. 2015; Mishra et al. 2004). In addition, our group has shown that PALF, with a novel extraction method, can be used effectively to reinforce both plastics (Kengkhetkit & Amornsakchai 2014, 2012) and rubbers (Prukkaewkanjana

et al. 2015). The use of pineapple leaf as a source of fibre for composite application is still very limited. Although the structure and properties of PALF have already been reported (Alwani et al. 2014; Khalil et al. 2007; Mukherjee & Satyanarayana 1986; Neto et al. 2015), the information seems to be limited and not quite complete as that known for flax. In summary, the fibre is known to be multicellular (Mukherjee & Satyanarayana 1986) vascular bundles (Khalil et al. 2007). Its ultimate cell length and breadth are 3-9 mm and 4-8 mm, respectively (Mukherjee & Satyanarayana 1986). It is one of the fibre with the highest a-cellulose content and thus has high tensile strength (Khalil et al. 2007; Neto et al. 2015). Strength of the fibre could vary with diameter and length (Mukherjee & Satyanarayana 1986). This is probably the reason that mechanical properties of the fibre cover relatively wide range (Neto et al. 2015). However, it is still not clear to what degree that strength of fibre from different position along the leaf length varies. In order to fully utilize the fibre especially for composite reinforcement, its structure, properties and characteristics should be known better. Therefore, it was the objective of this study to further investigate the morphology, microstructure, and properties of PALF. In addition, the characteristics of PALF from different parts of the leaf are analyzed. This is simply by dividing the leaf into three parts with equal length and named bottom, middle and top according to its height.

Materials and Methods

Pineapple leaves were collected from a cultivation area in Kok Kwai sub district, Ban Rai district, Uthai Thani province, Thailand. Sodium hydroxide (NaOH) was commercial grade. Formalin-Acetic Acid-Alcohol (FAA) solution was commercial grade which contains formalin, glacial acetic acid, and 70% ethanol at the volume ratios of 0.5:0.5:9.

Sample Preparation for Morphological Studies

For light microscopy observation, a transverse section of pineapple leaf fibre was prepared using a wax-embedding method (Johansen 1940). The materials were cut to about 10-20 µm thickness using a Leica SM2000R sliding microtome. The sections were pre-stained with Safranin O, counterstained with Fast Green FCF and finally mounted in DePeX mounting media (Johansen 1940).

For scanning electron microscopy (SEM) observation, three types of sample were prepared, i.e. transverse-section of a leaf (Figure 1), transverse-section of a fibre bundle and longitudinal view of a fibre bundle. To obtain leaf transverse-sections, the leaf was immersed in an FAA solution for at least 24 h, and then stored in 70% ethanol. The samples were cut with an automatic microtome MT-3 to about 5-10 µm thickness and dried at room temperature. The sample was air-dried at ambient temperature so that shrinkage would occur slowly and not causing damage to the specimen. To prepare fibre bundle transverse-sections hand scraped fibre was sandwiched between two pieces of styrene-ethylene butylene-styrene block copolymer. The sample was cut across the fibre axis with a microtome (American optical 820) and then soaked in sodium hydroxide solution for about an hour. The sample was finally cleaned with distilled water and dried in a hot air oven at 60°C. To obtain the fibre longitudinal view hand scraped fibre was fixed at the both ends on a metal disc with epoxy glue. The sample was soaked in 10%w/v NaOH solution and the fibre was washed along its length with a jet of NaOH solution from a pipette fitted with a rubber bulb. The process was repeated several times with total washing time of about an hour. The fibre was finally washed with distilled water. All of the chemical treatments were carried out at ambient temperature.

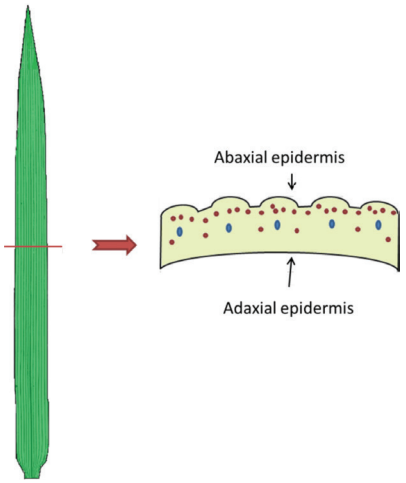

FIGURE 1. Schematic illustration for the preparation of transverse section of pineapple leaf

Fibre Extraction for Mechanical Testing

Each pineapple leaf was cut into 3 equal lengths being the bottom, middle, and top positions counted from the attachment to the plant. Fibres were separated from each part of the leaf by hand scraping. For mechanical property testing, the extracted fibre was mounted on a paper frame having a rectangular window of 1×2 cm^2 cut along the frame's long axis for ease of handling. After mounting the fibre with a frame on the testing machine, the paper frame was cut on both side and the test initiated similar to Yusoff et al. (2009).

Characterization

Morphology The structures of the leaf and the fibre were observed using both a light microscope (Olympus; model BXS1TRF) and a scanning electron microscope (SEM) (Hitachi Tabletop Microscope; model TM 1000, Japan) and the sample was coated with gold before being observed under the electron microscope.

Mechanical Properties Fibre strengths were determined with a universal testing machine (Instron, Model 5556, High Wycombe, UK) with a 100 N load cell and a gauge length of 20 mm. A crosshead speed of 10 mm/min was used. The tensile testing was adapted from ASTM D3379-75 (1998) and Charlet et al. (2010). At least 30 fibre specimens were measured for each sample. Fibre diameters were determined with an optical microscope using the ImageJ, a public domain image processing program (Rasband 1997-2015).

Fourier-Transform Infrared Spectroscopy (FTIR) Functional groups on the surface of PALF before and after treatment were studied using a FTIR spectrometer (Frontier, PerkinElmer) in Attenuated Total Reflection (ATR) mode. The scans were conducted in the range of 4000-500 cm^{-1} at resolution of 4 cm^{-1}. The number of scans was 32.

Thermogravimetric analysis (TGA) In order to compare the thermal stability of PALF, tests were carried out with a Mettler Toledo TGA instrument (Model SDTA851, Switzerland). The samples were heated from 40 to 600°C at a heating rate of 20°C/min under oxygen atmosphere.

Results and Discussion

General Structure of the Leaf and Fibres

The cross-section of pineapple leaf is prepared as shown schematically in Figure 1. The light micrographs of the stained transverse-section of a pineapple leaf are shown in Figure 2. The leaf is rather spongy and can be roughly divided into two unsymmetrical parts, i.e. the upper less dense and the lower more dense parts. According to their positions and morphologies, the fibre bundles are found in the vascular tissue (A) and in the mesophyll (B). These are called fibrovascular bundles and mesophyll bundles, respectively.

Figure 3 displays high magnification SEM images of the two types of fibre. Fibrovascular bundles have a crescent-like shape and seem to pair up facing each other, so forming a structural support for vascular tissue. Mesophyll bundles have a somewhat rounded shape and are placed rather regularly in the mesophyll. It seems that both types of fibre bundle differ only in this macroscopic shape or morphology. Each bundle is made up of a number of discrete microfibres of similar size and shape. These microfibres have an average diameter of approximately 4-5 mm and a thick wall. Microfibres have hole or lumen in the middle. For some, however, this hole is not apparent. At this stage, it may be assumed that the overall microfibre wall consists of a primary wall and three sub-layers of secondary walls (Khali et al. 2007).

Rough calculation using multiple images of leaf cross-section suggests that approximately 56% of PALF are mesophyll bundles and 44% are fibrovascular bundles. It is, however, very difficult to differentiate the two types of microfibre after extraction from the bundle. Hence, in all that follows, PALF refers to fibre bundles in general.

Some Physical Properties of Fibre Bundles

Since pineapple leaf has a long narrow shape, it is natural to question if the shape, size and properties of the microfibres

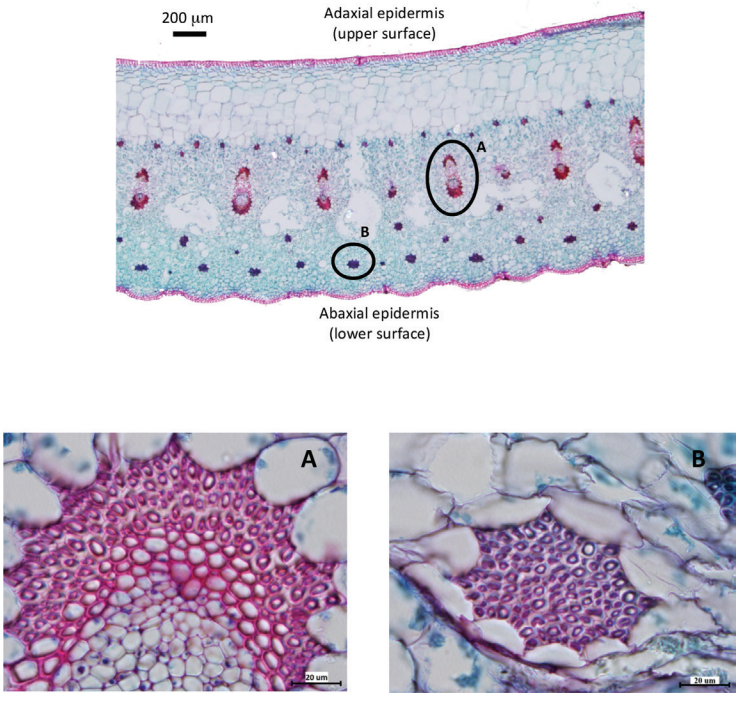

FIGURE 2. Light micrograph of stained transverse section of pineapple leaf displaying vascular fiber (fibrovascular) bundles (A) and mesophyll fiber bundles (B)

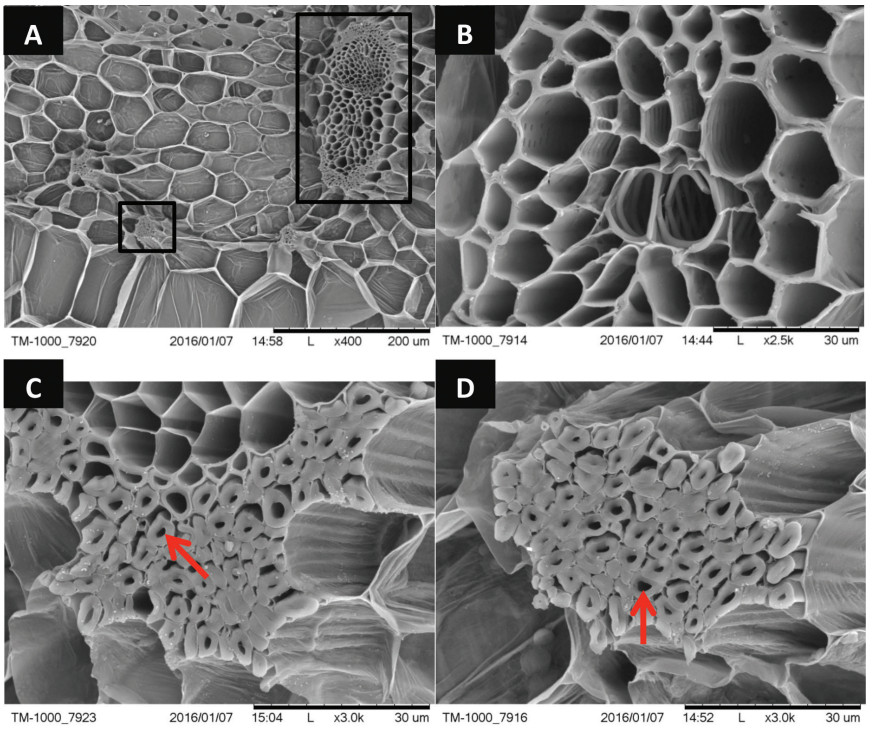

FIGURE 3. SEM micrographs of transverse section of pineapple leaf at low magnification (A). Fibrovascular bundles in the large rectangular in (A) are shown at higher magnification in (B) and (C) and mesophyll bundles in (D). The arrows show the lumen of each cell

vary along its length. To elucidate this, transverse sections of a leaf were prepared from the extreme ends of a leaf and representative samples are displayed in Figure 4. Generally, the size and shape of the fibre bundles are rather similar. However microfibres extracted from the bottom part of the leaf display clear lumens while those from the top rarely show the lumens. Also, microfibres from the bottom part are rounder than the rather elongated ones from the top.

Tensile strengths and diameters of fibre bundles extracted from three different positions in the leaf, i.e. top, middle and bottom, were determined and the results are shown in Figure 5. The tensile strengths of the fibres from different parts are not too different. Fibres from the middle part have the highest tensile strength, those from the bottom are slightly weaker, while those from the top are the weakest. However, the differences between middle and bottom parts were not much different. The values observed fall in the mid-range of the reported values of 170 and 1,627 MPa (Kalia et al. 2011). Presumably this variation is due to a difference in the pineapple varieties studied (Neto et al. 2015). Generally the strength of PALF is considered relatively high when compared to other plant fibres. It compares very well with that of flax and hemp fibres which are already used in the industry (Kalia et al. 2009). The average fibre cross-section areas gradually decrease from the bottom to the middle and then to the top parts of the leaf. The representative stress-strain curve is shown in Figure 6 and average mechanical properties of fibre bundles extracted from each part of the leaf are shown in Table 1.

In order to confirm such a change in the tensile strength of fibre bundles from different positions on the leaves, a larger number of specimens were tested and the results are shown in Figure 7 in the form of a Weibull plot. This is a continuous probability distribution widely applied to describe mechanical properties of fibres (Rinne 2008). The plot can be constructed from the probability of failure (Pf) of fibre specimen at different stress (s). The fibre bundles from the top position of the leaves are weakest while those from the middle and the bottom parts have a similar distribution. These more numerous results agree well with the earlier ones discussed above.

NATURE OF THE FIBRE BUNDLES AND OF THE MICROFIBRE

PALF consists of bundles of microfibres (Kengkhetkit & Amornsakchai 2012; Neto 2015), which may break up into smaller bundles during extraction process. Although plant cell walls are bonded together with a very thin pectin layer, which is called the middle lamella (Evert 2006a; Khalil et al. 2007), it is difficult to visualize how these microfibres bond together to form the larger bundle structure. Investigation of a number of broken bundle ends from the tensile test (discussed earlier) suggests that the microfibres are glued together with some matrix material. Broken bundle ends, displaying microfibres separated from the matrix material, are shown in Figure 8. To confirm this point, a fibre bundle end was washed with NaOH solution and this is also shown in Figure 8. Since the bonding material is soluble in NaOH solution, it is likely to be

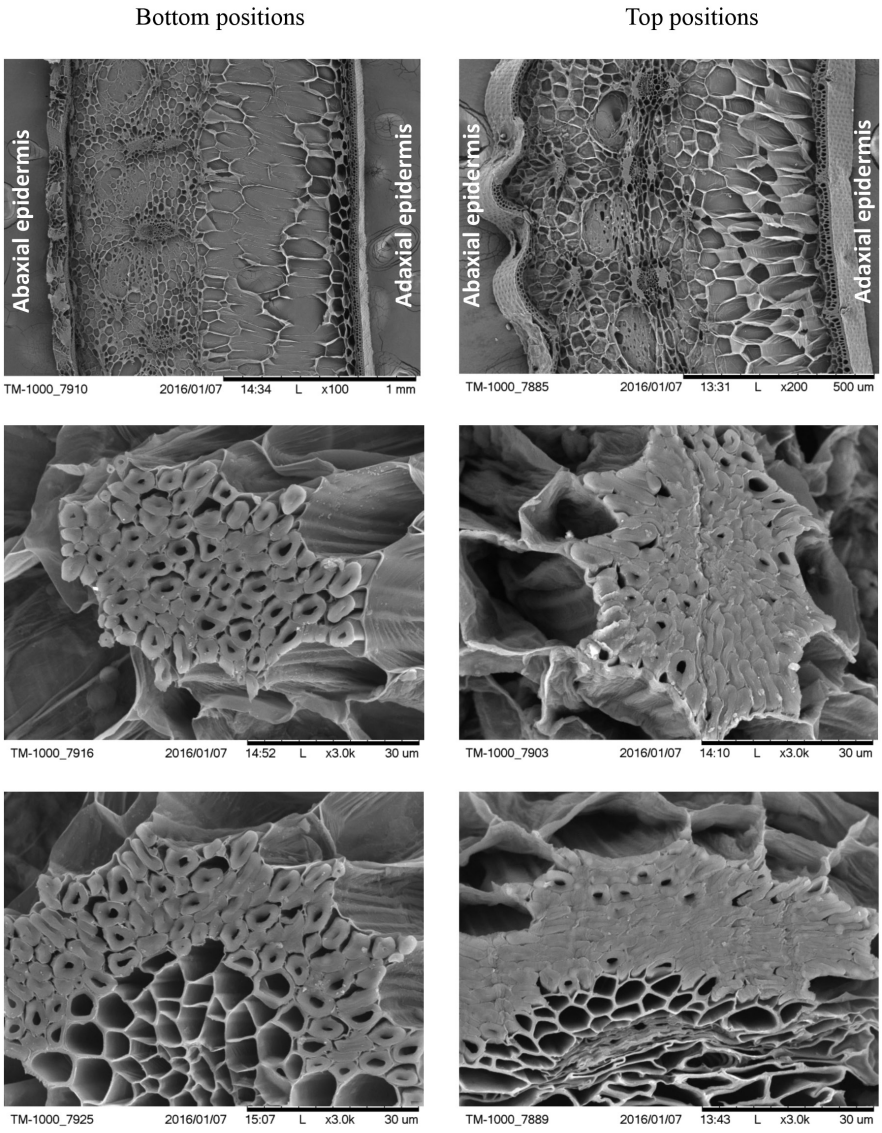

FIGURE 4. SEM micrographs of transverse-sections of a pineapple leaf at bottom (left column) and top (right column) positions. The top row displays the overall view, the middle row shows mesophyll fiber bundle and bottom row is a fibrovascular bundle

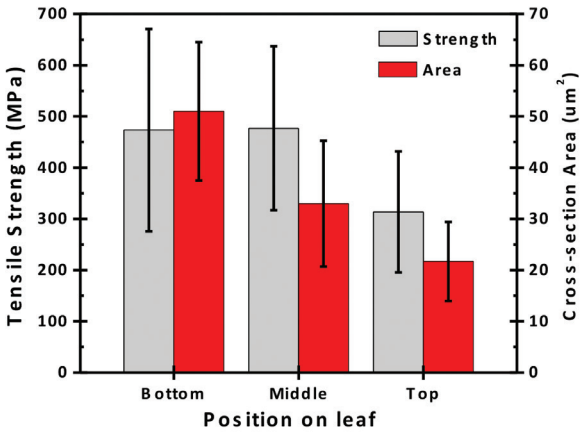

FIGURE 5. Tensile strengths and cross-section areas of fibers extracted from different parts of pineapple leaf

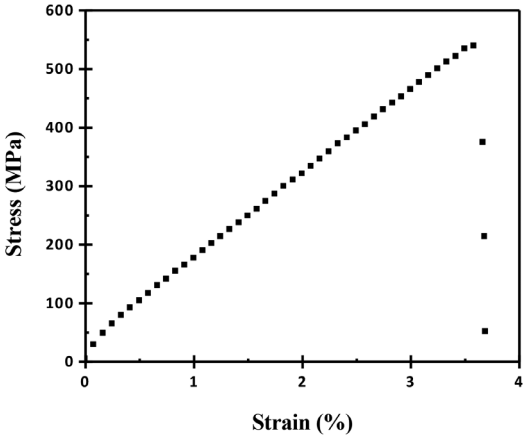

FIGURE 6. Stress-strain curve of pineapple leaf fiber (PALF) bundle

TABLE 1. Mechanical properties PALF bundles from different part of the leaf

Position of leaf	Strength (MPa)	Modulus (GPa)	Elongation at break (%)
Top	313.6 ± 118.1	16.2 ± 4.8	3.5 ± 0.8
Middle	476.9 ± 160.3	19.9 ± 8.2	3.7 ± 0.8
Bottom	473.3 ± 197.4	18.5 ± 5.7	4.1 ± 1.0

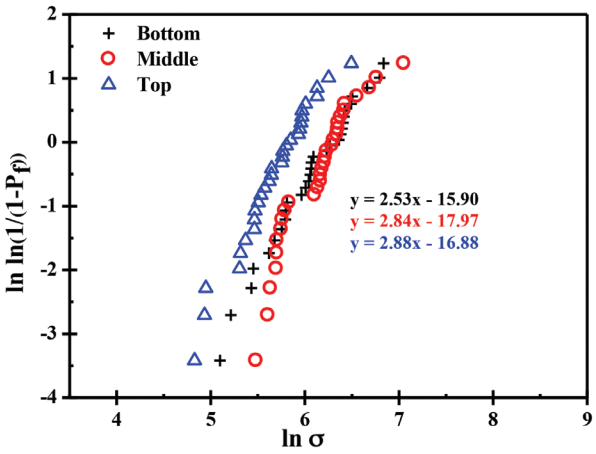

FIGURE 7. Weibull plot of strength distribution of PALF extracted from different positions of pineapple leaves. See text for explanations

hemicellulose (Saha et al. 1993; Xiao et al. 2001). By measurements carried out on SEM images from multiple areas, the microfibres diameter is approximately 3-4 mm which is consistent with that reported earlier (Kengkhetkit & Amornsakchai 2012; Tanpichai & Witayakran 2015). This is smaller than that of flax elementary fibre, which is about 12-17 mm (Charlet et al. 2010). The general appearances of the fibre bundles, before and after washing, are shown in Figure 8. The starting fibre is seen to be covered with some material of which the cellular-looking structure in the longitudinal direction of the fibres can be seen (cf Figure 3). After washing with NaOH solution, the covering material was removed leaving a very clean surface with a number of parallel straight lines. These lines are edges of microfibres. A small bundle was followed along its length to determine the length and to observe the shape of microfibre ends. No interruption of microfibre was observed for the entire length of approximately 10 mm. A numbers of fibres from many specimens were observed and it is confident that no interruption was observed. This is shown in Figure 9 as a stitched image (an image formed from several separate pictures of a long object).

FTIR AND THERMOGRAVIMETRIC ANALYSES OF FIBRE BUNDLES AND MICROFIBRES

Fibre bundles and microfibres were subjected to FTIR analysis to ascertain any changes in functional groups and chemical compositions. The FTIR spectra of fibre bundles (PALF) and NaOH washed fibre (PALMF) are displayed in Figure 10. The main differences between the PALF and PALMF spectra are that the peaks at 1732 and 1246 cm^{-1} appear in the former but not the latter. These peaks are due to C=O and C-O stretching in the acetyl groups of hemicelluloses (xylan) (Labbé et al. 2005; Marchessault 2009). This xylan can readily be removed by treatment with alkali (Marchessault 2009) thus confirming that the bonding material is primarily hemicellulose.

To further confirm this, the extracted material was precipitated from the alkali solution by neutralizing with glacial acetic acid. The precipitated material was separated by centrifugation. Since it is water soluble, it has to be washed several times with ethanol and then acetone. After drying under vacuum at ambient temperature, IR spectra

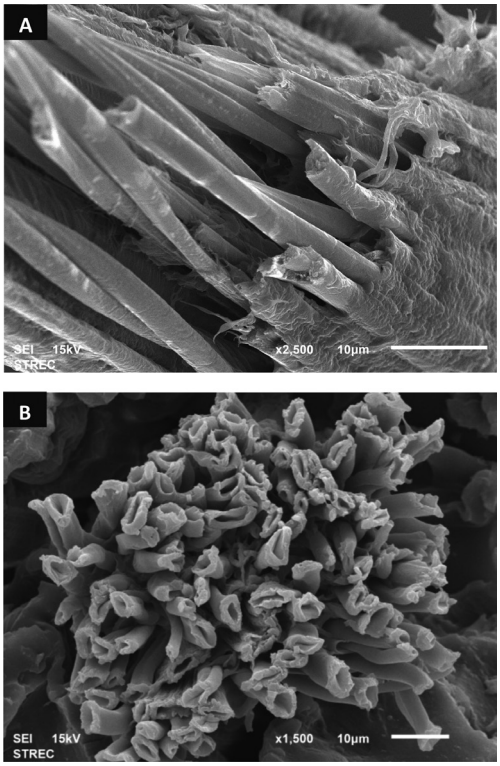

FIGURE 8. SEM images of a broken end of a fiber bundle (A) and of the cut end after washing with 10% w/v NaOH solution (B). The fiber bundle was from the middle part of the leaf

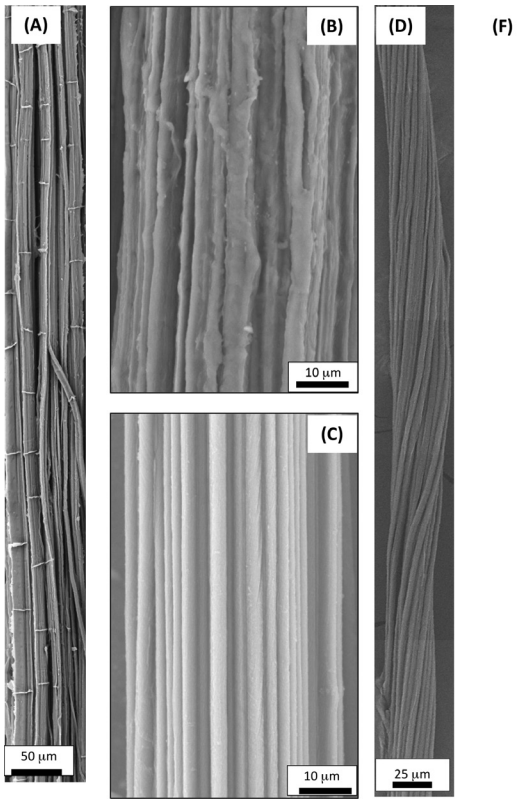

FIGURE 9. SEM images of hand scraped PALF (A, B) and that after washing with 10% w/v NaOH solution (C, D and F) with different magnifications

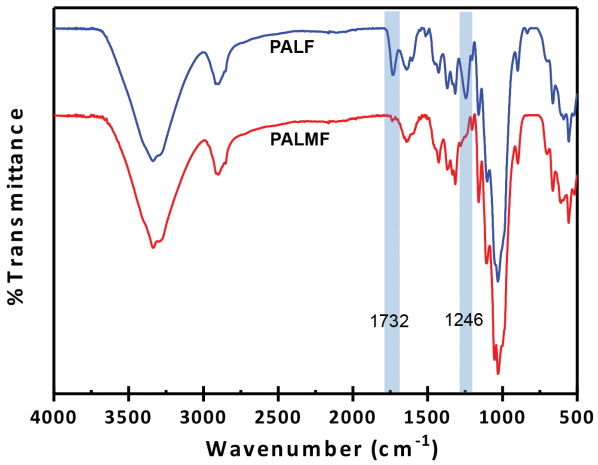

FIGURE 10. IR spectrum of hand scraped (PALF) and that after washing with 10% w/v NaOH solution (PALMF)

was obtained and is shown in Figure 11. The spectrum is similar to that of xylan. The absorption at 1648 cm^{-1} is associated with absorbed water while that at 1163 cm^{-1} with the C-O-C vibrations in hemicellulose (Xiao et al. 2001). The sharp band at 1042 cm^{-1} corresponds with either C-O, C-C stretching or C-OH bending in hemicellulose. The β-glycosidic linkages between sugar units in two hemicelluloses are seen as a small band at 903 cm^{-1} (Xiao et al. 2001). Therefore, it can be concluded that hemicellulose could be washed out from the pineapple leaf fibre with alkali solution. In addition, previous chemical analysis also indicates that the amount of hemicellulose decreases after washing the fibre with 10% w/v NaOH solution (Fu et al. 2013; Kengkhetkit & Amornsakchai 2014; Saha et al. 1993).

Hemicellulose is known to be thermally less stable than cellulose (Brígida et al. 2010; Yang et al. 2007). Thus removal of hemicellulose from the fibre bundles should be beneficial in terms of the thermal properties of the resulting PALMF. To confirm this point, PALF and

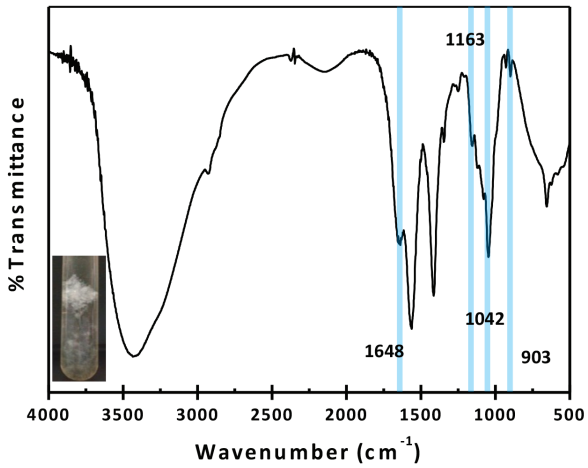

FIGURE 11. IR spectrum of materials extracted from pineapple leaf fiber (PALF) with 10% w/v NaOH solution

PALMF were subjected to thermo gravimetric analysis and the results are shown in Figure 12. There are two steps of weight loss at about 70°C and just after 250°C. The first is due to the loss of moisture in the fibres and the second to the thermal decomposition of the material. The onset of the thermal decomposition of PALMF is about 20°C higher than that of PALF, indicating better thermal properties of the treated fibre. The increase in decomposition temperature may seem to be quite small. This is because the starting PALF already contains relatively high cellulose content and thus removal of the small remaining hemicellulose causes a limited increase in the thermal decomposition temperature. The results are consistent with that reported by Fu et al. (2013). The TGA curves are similar to that of flax fibre reported by Kannan et al. (2013).

WAXS ANALYSIS OF FIBRE BUNDLES AND MICROFIBRES

Fibre bundles and microfibres were subjected to WAXS analysis for their crystal structures. Figure 13 displays their WAXS patterns. The patterns indicate very high orientation of cellulose crystals in the fibres. They are similar to that of jute fibres (Lin et al. 2014), ramie fibre (Woodcock & Sarko 1980) and also electron diffraction patterns of ramie (French 1978) and cotton microfibrils (Herbert & Muller 1974; Paralikar & Betrabet 1977) suggesting that the cellulose in PALF is of type 1 (Cellulose I). This is characterized by the three diffractions assigned (110), (200), and (004). The (110) reflection gradually split into two reflections as (10-1) and (101) after the PALF bundles were washed with NaOH solution to yield the microfibres. This change is similar to that observed in jute fibres (Lin et al. 2014).

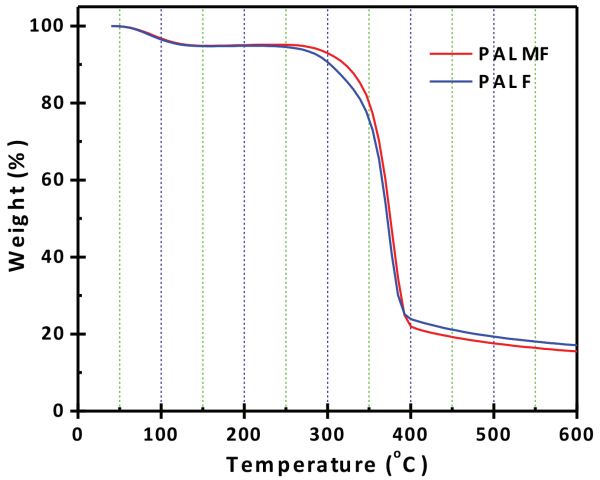

FIGURE 12. Thermograms of hand scraped PALF and that after washing with 10% w/v NaOH solution (PALMF)

The results described above lead to a general picture of the PALF. It is, in fact, groups of microfibres bonded with hemicellulose so forming structural support for vascular tissue and mesophyll (Evert 2006b). These observations suggest that the membered microfibres are continuous with no interruption along their length. They may be regarded as the elementary fibres since they cannot be separated into smaller building blocks without chemically degrading the structure. The dimensions measured in this work should not be confused with the fact that the fibres can be broken down to yield nanocrystalline cellulose (Cherian et al. 2010; Santos et al. 2013) or microfibrillated cellulose (Lavoine et al. 2012). This is because this degradation involves digesting away amorphous regions in the elementary fibre to show nanocrystalline fibrils, whiskers or needles.

As shown in Figure 4, microfibres do not all have clear lumen through their entire length. This lumen forms as a result of degeneration of intracellular organelles (Madsen & Gamstedt 2013). It is likely that as the plant grows, the leaf elongates and thus so too do the microfibres. Microfibres at the top part of the leaf are always younger in age than those at the bottom part. Therefore, degeneration, resulting in larger lumen size of intracellular organelles occurs more as we go down the microfibres from leaf tip to the leaf bottom. Greater degeneration leads to a slight drop in the apparent tensile strength of the fibres in the bottom part of the leaf. Thus, when this different in apparent density of the fibre (due to different lumen size) is taking into account, the strength of fibre from different positions may be regarded as very similar.

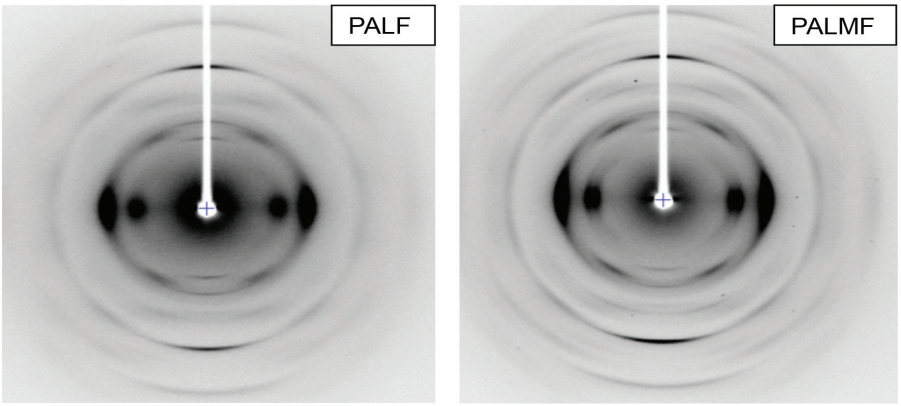

FIGURE 13. WAXS patterns of hand scraped PALF (left) and that after washing with 10% w/v NaOH solution (PALMF) (right). Fiber axis is vertical

The observed continuous nature of microfibres would suggest that ones with an extremely high aspect ratio can be prepared from pineapple leaf without changing crystal structure. The ratio is, in fact, determined solely by the cut length of the leaf. Such unique microfibres can be very useful in various applications, e.g. counterfeiting prevention markers in banknote, green support for immobilization of catalyst, enzyme and ion chelating groups in chemical reaction systems, and green reinforcement for composite materials. These microfibres can be prepared in large quantities more easily than that from steam explosion (Cherian et al. 2010; Tanpichai & Witayakran 2015).

Conclusion

It is shown that pineapple leaf fibre bundles are found in both vascular tissue and in mesophyll. They differ only in cross-section morphology. The pineapple leaf fibre bundle is a composite made of a number of small microfibres held together with hemicellulose. These microfibres are fine and very long, presumably as long as the full length of the leaf. At the base of the leaf microfibres are round and have large lumen. The lumen shape becomes more elongated, with smaller lumen, farther away from the leaf base. The tensile strength of the fibre bundles varies slightly along the length of the leaf, with the top part having the lowest strength and the middle part the highest. The change in strength may occur from the degeneration of the mature fibre cell at the middle of the cell (lumen). The strength of PALF is similar to that of flex and hemp fibres and thus can be used in similar applications. However, for practical use, the difference is probably too small to both cause any real problem and be worth the cost of separation.

Acknowledgements

The authors gratefully acknowledge the Center of Excellence for Innovation in Chemistry (PERCH-CIC), Office of the Higher Education Commission, Ministry of Education for partial financial supports of the project. WAXS patterns were kindly provided by Professor Kohji Tashiro from Toyota Technological Institute.

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Received: 13 July 2018
Accepted: 28 August 2018