

**STUDY ON MECHANICAL PERFORMANCE OF  
HYBRID GREEN COMPOSITES OF  
POLYLACTIC ACID REINFORCED BY  
KENAF, BAMBOO AND COIR FIBERS**  
(ケナフ, 竹, コイアによって強化されたポリ乳酸系ハ  
イリッドグリーンコンポジットの機械的性能に関す  
る研究)

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## **Abstract**

This study investigated the mechanical properties of unidirectional kenaf, bamboo and coir fiber-reinforced polylactic acid (PLA) composites. A hand lay-up technique was used to prepare the composites with fiber weight content varying from 40 wt.% to 70 wt.%. The various composites were fabricated to investigate the effect of fibers' composition in monolithic green composites and hybrid green composites. These composites were named as green composites since they were made up from fully biodegradable materials. To investigate the performance of the composites, numerous tests were done. At first, fiber bundle tensile tests were conducted, followed by tensile, flexural and impact tests for monolithic and hybrid green composites to analyze their properties. In addition, two stacking sequences of low and high modulus fibers in the outer layers were compared to evaluate the composites' performances. Scanning electron microscopy was used to observe the fracture surface morphology.

The result of tensile strength on monolithic green composites was used to estimate the compatibility of fibers and PLA matrix. It was shown that at up to 70 wt.% fiber content, kenaf/PLA achieved approximately 290 MPa, and bamboo/PLA achieved approximately 210 MPa, producing higher strength than coir/PLA (55 MPa). Based on these results, with a higher content of kenaf and bamboo fibers, the strength would increase while a higher content of coir fibers does not contribute to the strength properties of the composites. On the other hand, coir fiber produced a higher elongation in the composites. Therefore, to optimize the strength properties in this hybrid, kenaf and bamboo fibers are the main factors that influence strength. In terms of adhesion between PLA matrix and fibers, bamboo was incompatible with PLA due to evident

voids and fibers pull out from composites upon fracture. A further chemical modification is recommended for bamboo fibers.

According to the results of tests on three types of high modulus fibers in the outer layer of hybrid composites: KBCCBK/PLA, KCCK/PLA and BCCB/PLA, the tensile strength of KBCCBK/PLA composite achieved 187 MPa, approximately 20 and 78% higher than that of BCCB/PLA and KCCK/PLA, respectively. The Young's moduli of the three composites ranged from 6 to 7.5 GPa. High flexural strength was obtained in both KBCCBK/PLA (199 MPa) and BCCB/PLA (206 MPa) composites, approximately 16 and 20% higher than that of KCCK/PLA, respectively. However, KCCK/PLA composites showed the highest flexural modulus, approximately 70% higher than those of other combinations. Higher strain energy per unit volume required to break (toughness) was the characteristic of KBCCBK/PLA composites. It was found that the combination of high strength and stiffness of bamboo and kenaf fibers (outer layer) and high ductility of coir fiber improved tensile and flexural strengths compared to monolithic fiber green composites, particularly the coir fiber-reinforced PLA.

As high mechanical properties were obtained from using high modulus fibers in the outer layers, the effect of two symmetrical stacking designs of hybrid green composites were compared with high modulus fibers in the outer layers (KBC/PLA) and low modulus and high strain fibers in the outer layer (CBK/PLA). Tensile, flexural and impact tests were conducted to investigate their mechanical properties with total fiber content varying from 50 to 70 wt.%. Field emission scanning electron microscopy was used to observe the microstructural failures. The tensile strength of both composites had a similar trend and increased linearly up to 158 MPa. The tensile modulus was

approximately 6 to 7 GPa. It showed that the stacking sequence with high modulus fibers in the outer layers (KBC/PLA) improved flexural strength, approximately 49% higher than that of CBK/PLA with low modulus fibers in the outer layers. In contrast, the impact strength of composites with low modulus fibers in the outer layers (CBK/PLA) was approximately 21% higher than that of its counterparts. It was also found that the stacking sequence had no significant effect on tensile strength, while affecting tensile strain, which was increased approximately 58% due to the low modulus and high strain in the outer layer. As a consequence of water absorption, both composites' stacking sequences had weight increase significantly with fiber content after 48 hours of submerging in water (50 °C) when the weight increased 26-34 % and 28-39% for KBC/PLA and CBK/PLA, respectively. It revealed that these composites are sensitive to water.

A further study was also done to investigate the damping loss factor of high modulus fiber in the outer layer (KBCCBK/PLA) composite to compare with those of selected synthetic composites. It was found that the loss factor of the hybrid green composites was considerably higher than those of synthetic composites due to the viscoelastic property of plant fibers. Based on hybridization results:

1. The combination of three kinds of fibers which are kenaf, bamboo, and coir produced composites with better strain, toughness and damping loss factor properties.
2. High modulus fibers in the outer layer produced composites with better flexural properties.
3. Low modulus fibers in the outer layer produced composites with better impact strength.

4. The compatibility of kenaf-coir fibers and PLA matrix produced composites with high flexural modulus.
5. Stacking sequences had a small effect on tensile strength but had a major impact on flexural strength.
6. Hybrid green composites had higher damping loss factor than those of GFRP and CFRP.

In conclusion, hybrid green composites containing kenaf, bamboo, and coir fibers with bio-based polymer (PLA) were fabricated and their mechanical properties were assessed. The hybrid green composites provide fully eco-designed material, tailoring the inherited low mechanical properties of certain monolithic green composites.

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## **Publications**

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### **Conferences:**

1. Rosni Binti Yusoff, Hitoshi Takagi, Antonio Norio Nakagaito. (2015). Mechanical performance of hybrid green composites. *Proceedings of the International Conference on Advanced Technology in Experimental Mechanics*, Toyohashi, Page 101.
2. Rosni Binti Yusoff, Hitoshi Takagi, Antonio Norio Nakagaito. (2015). Preparation and characterization of hybrid green composites. *Proceedings of the Advances in Mechanics of Composite Materials and Structures*, Seoul, Page 66.
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# CHAPTER ONE

## Introduction

### 1.1 Background

Composites have become indispensable materials in our daily lives for numerous applications. The “green” constituent in the composites has been discussed to treat the ever-existing environmental impact. Both fibers and polymer matrices used in composites should be eco-designed, in order to degrade after the end of service. Apart from that, the bio-resources should be abundantly available for continuous production to replace the usual scarce petrochemical matrices (polypropylene, polyethylene etc.). Therefore, plant-based composites are suitable for environmental concerns and widely available around the globe. Plants offer numerous kinds of fibers and process-ability with high specific strengths. Their strength properties depend on physical and chemical properties influenced by the growing regions. The mechanical performance of composites is dominated by fibers and matrices (polymers). In the case of plant-based polymer matrices, it can be renewable as well as providing a barrier to the environmental effect. This study begins with the evaluation of fiber bundles properties, then interfacial strengths of fiber-matrix through mechanical performances (such as tensile strength, flexural strength and toughness).

The past research on conventional composites was intended for long-term durability which are made from the non-degradable matrices and synthetic fibers (Netravali & Chaba, 2003). Carbon and aramid fibers are reinforcing fibers made from synthetic polymers, such as poly-acrylic acid and poly(p-phenylene terephthalamide) (Wallenbergger & Weston, 2004). The initial glass fiber contained 0.3-0.9% fluorine



and 6-8% boron oxide which emitted toxins during processing, but it has been replaced without these two ingredients. Common glass fibers (designated E-glass) that were developed in the late 1940s are currently being used in more than 65 % of composites produced, which are still in high demand (Raju, 2012), and used to combine reinforcing fiber with natural fibers such as kenaf, flax, and sisal for environmental friendly purposes. They are referred to as partial green composites, or plant-synthetic composites. Despite the high strength, waterproofness, and strain resistance of composites-synthetic-based fibers, the prolonged lifetime of the synthetic composites leads to over environmental impact through the accumulation of landfill sites. The environmental impact occurs when the high loading of these non-decomposing materials ends up in landfills. Presently, undeveloped land has been used as disposal areas (the cheapest way of waste disposal), and will soon be fully occupied due to the increasing non-biodegradable waste generation (Assamoi & Lawryshyn, 2012).

Compared to synthetic fiber-based composites, plant fiber-based composites have renewability, sustainability, and eco-efficiency. Their capability of absorbing carbon dioxide shows promising value in mitigating the environmental pollution (La Mantia & Morreale, 2011). Meanwhile, their natural properties such as low density, flexibility during the processing filler and lack of harm to the equipment, and acceptable strength properties at a low cost per volume have made them popular reinforcements in polymer-based composite materials. On the other hand, the disadvantages of plant fibers such as poor adhesion, variability, low thermal resistance and hydrophilic nature (Céline, Fréour, Jacquemin, & Casari, 2013), could be solved by fiber surface treatments in which the fibers are suitably modified by either physical or chemical treatments (Pandey, Ahn, Lee, Mohanty, & Misra, 2010), processing method (Sathishkumar, Naveen, & Satheeshkumar,

2014) (La Mantia & Morreale, 2011) and proper fiber alignment (McGregor, Duhovic, Lin, & Bhattacharyya, 2010; Pickering & Aruan Efendy, 2016).

Currently, an extensive use of plant fibers, based on environmental and low-cost factors rather than on their mechanical properties, are expected to replace synthetic fibers like carbon fibers, aramid fibers, and glass fiber. For example, many established plastics companies in developed countries have changed to using plant fibers to alleviate environmental impacts (Satyanarayana, Arizaga, & Wypych, 2009). The acceptable performance obtained in the structural and non-structural, and automotive industries (Begum & Islam, 2013; Mohammed, Ansari, Pua, Jawaid, & Islam, 2015) has pushed the way to fully use green composites. Therefore, as an alternative to the partial green composites, fully green composites are attempted in this study.

## **1.2 Objectives of the study**

This research is focused on biodegradable green composites and hybrid composites based on three kinds of natural fibers, which are kenaf, bamboo and coir fibers, to investigate the mechanical properties of hybrid green composites. Tensile tests for fiber bundles and single fiber green composites were carried out to estimate the properties of each fiber for theoretical and experimental comparison. Currently, research on hybrid green composites is limited, unlike that for “partial hybrid green composites” in which both synthetic fibers and plant fibers are used as reinforcement. For example flax/glass/phenolic matrix (Zhang, Li, Ma, & Yu, 2013), jute/sisal/epoxy/hardeners (Sudhir, Madhukiran, Rao, & Madhusudan, 2014), sisal/glass fiber/epoxy/hardeners and jute/glass fiber/epoxy/hardeners (Ramesh, Palanikumar, & Reddy, 2013).

Therefore, this research is intended to fabricate eco-design composites, the fully hybrid green composites to replace the synthetic materials that offer acceptable strength for current applications in order to alleviate the negative environmental effect. To achieve the above goals, the outlines of this research were as follows.

1. Determining each fiber bundle and matrix properties (tensile properties, densities).
2. Fabrication of monolithic green composites.
3. Fabrication of hybrid green composites.
4. Evaluation of the composites properties through mechanical testing:
  - Tensile test – tensile properties
  - Bending test – flexural properties
  - Izod impact test – impact strength
  - Centre impedance method – damping loss factor
5. Studies on the effect of stacking sequences and fiber content of the composites.
6. Studies on the effect of fiber-matrix interfacial properties by the optical and scanning electron microscopy

### **1.3 Structure of the thesis**

This thesis consists of seven chapters. Chapter One explains the general introduction of composites used in current applications and the emerging trend in composites materials due to environmental concerns. The objectives of this study, as well as the structure of this report, is also presented.

Chapter Two discusses the literature review on plant fibers (coir, bamboo and kenaf) and polylactic acid as the main constituent materials in this study. Chapter Three

presents the materials testing and fabrication methodology of monolithic green composites and hybrid composites.

The experimental results are discussed in three consecutive chapters (Chapters Four, Five and Six). Chapter Four explains the density and tensile properties of untreated fiber bundles, and tensile properties of single type of reinforcement green composites. Chapters Five and Six present the hybrid green composites of various stacking sequences. Finally, Chapter Seven gives the conclusions of this research.

# CHAPTER TWO

## Literature Review

### 2.1 Plant fiber

Plant fibers are promising materials to be used as a reinforcement or filler in a composite material. The fibers have been considered as lower cost reinforcements (Beckwith, 2003). However, the selection of plant fibers as reinforcement or filler is the biggest concern in order to make them suitable and compatible in the matrix used for composites' production, especially for advanced and hybrid composites. The choice of plant fibers often determines the final properties of a composite due to the variation of fibers' strength embedded in the matrix resin.

Plant fibers are greatly distinct from one another, typically based on the source of their climax region, the age of plant and extraction method (Sreekala, Kumaran, & Thomas, 1997). Kenaf fibers, for example, are grown in different conditions which influence length. Fiber taken from longer bast has the greater strength (Ochi, 2008). Moreover, according to a study, the chemical composition of kenaf (lignin, cellulose, pentosans, etc.) is different between the leaves and stem and increases with age (Rowell, Young, & Rowell, 1996). As for bamboo, it grows vertically and the growth rate is fast, two inches per hour in some moso bamboo species and can achieve a height of 60 feet in three months (Abdul Khalil et al., 2012). Fibers' length also affects the composites' mechanical properties (Shringi, Solanki, & Sharma, 2014) as well as the diameter and thickness. The dimensions of bamboo and coir fibers vary along the fibers' length (Sen

& Reddy, 2011). Therefore, the density of fibers differs depending on the kinds of fibers, which affects the volumetric fraction of the fiber content in the composites.

## **2.2 Properties of plant fibers**

Plant fibers are derived from several sources (such as bast, leaf, seed, and wood), and basically found in cellular form, which has a degree of inherent strength and stiffness due to the geometric and internal structure of plants (Beckwith, 2003). The fibers are taken from a plant cell, which is glued together by a pectin and or lignin-rich meddle lamella. Unlike natural fibers from animal cells, a plant cell has primary and secondary cell walls (Célino et al., 2013), basically to provide mechanical stability (Müssig, 2010). It has a chemical composition that determines the fibers' individual properties.

Due to the growing season, different parts of a plant fiber are diverse around the globe. Fibers taken from root, stem, trunk and leaf are different in chemical and physical properties (Rowell et al., 1996), and most properties are governed by the geometry of the secondary cell wall. This wall dominates properties in tension as it swells, breaking into fibril, winding to the helical path that inclines at the angle of the fiber's axis, which is called the cellulose microfibril angle (MFA) (Müssig, 2010; Mwaikambo, 2006). Generally, the bast fiber has an MFA below  $10^\circ$ , and its cell wall thickness often increases until the lumen almost disappears when it stays alive for a longer period (Müssig, 2010). Figure 2.1 illustrates the arrangement of the MFA and cellulose in cell walls (Gibson, 2012).

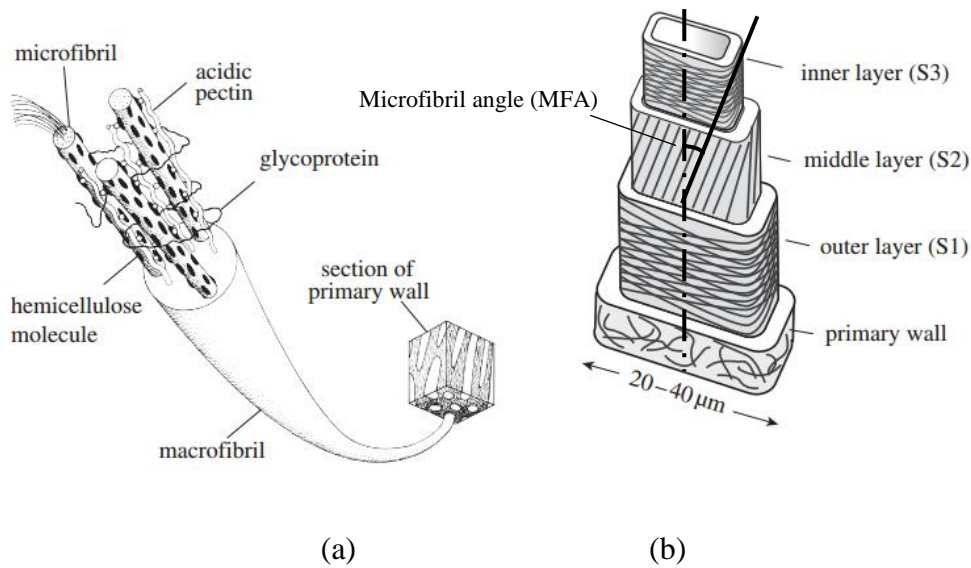


Figure 2.1 (a) Arrangement of microfibril formed from primary cell wall with the chemical composition (cellulose, hemicellulose, pectin), and (b) MFA and cell wall of wood made up of primary and three secondary layers.

## 2.2.1 Chemical composition of natural fibers

Vegetable or cellulosic or plant fibers are simple materials in term of chemical and physical properties. Cellulose, hemicellulose, and lignin are the main chemical composition of plant fiber. Approximately, half of chemical composition is cellulose, and approximately a quarter of that is hemicellulose and lignin, while pectin and waxes are extractive compositions in plant sources (Biagiotti, Puglia, & Kenny, 2004). Table 2.1 shows the compositions of selected plant fibers used intensely for composites production.

### 2.2.1.1 Cellulose

Cellulose is the main component of plant fibers. It is found that cellulose content was the highest in the early part of the growing season (Rowell et al., 1996). Generally, cellulosic fibers contain 60-70% cellulose (Thakur & Thakur, 2014). Meanwhile, lignin

and ash content vary and may be influenced by the amount of fertilizer. As in Fig. 2.2, the elementary unit of cellulose macromolecule is anhydro-d-glucose. It is almost pure in cotton fiber, whereas in wood, leaves, and stalks, it exists with other materials (lignin and hemicellulose) (Kalia et al., 2011). Cellulose molecules align to form microfibrils (Gibson, 2012). The three alcohol hydroxyls (-OH) give a hydrophilic property of cellulose via bonds between intramolecular and intermolecular as well as hydroxyl groups in the air which are an oxidizing agent. Exposure to chemical and solution treatments (Azwa, Yousif, Manalo, & Karunasena, 2013) will degrade the fiber since it swells with water a lot without dissolving in water.

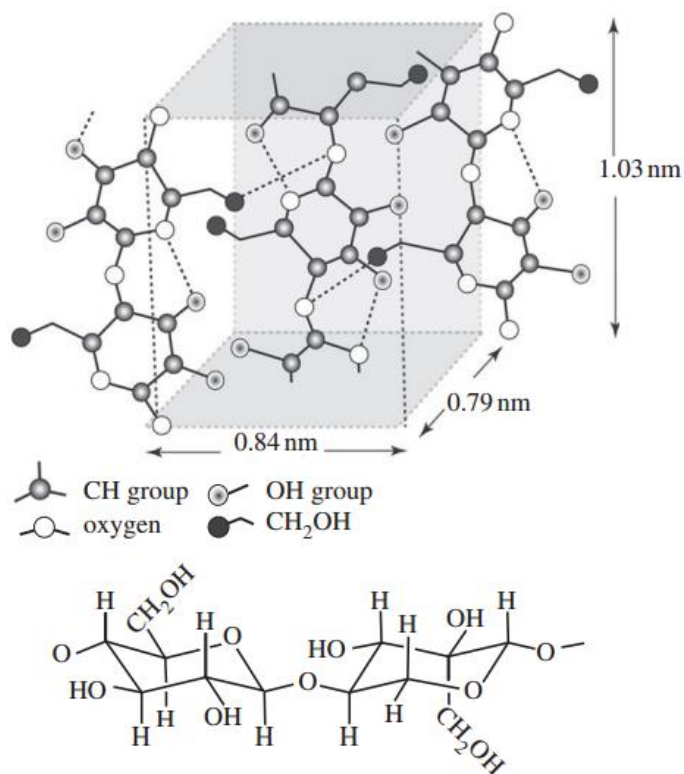


Figure 2.2 Molecular structure of cellulose, alternately rotated 180° (solid line, covalent bonds; dashed line, hydrogen bonds) and macromolecule of anhydro-d-glucose unit.



However, cellulose is largely crystalline in structure, organized into microfibrils that are stable in the normal environment. The hydrogen bonds existing in each cellulose micromolecule form microfibrils and interact to form fibers. Therefore, hydrogen bonding provides tensile strength of cellulose by the cumulative of the covalent bond of hydrogen in a fiber, based on the molecule structure of macrocellulose (Biagiotti et al., 2004). The fiber of cellulose macromolecules forms a spiral along the fiber axis which determines the strength and stiffness of the fiber. The smaller MFA means greater mechanical properties (Bledzki, Reihmane, & Gassan, 1996).

Apart from that, the degree of polymerization (DP) affects cellulose molecules that are determined by the number of glucose units. The average of DP for plant cellulose ranges from about 50 to approximately 600 (Rowell et al., 1996). The complex polymer homolog ( $C_6H_{10}O_5$ ), basically, bast fibers possess high DP values among all the natural fibers (approximately 10,000) (Lewin & Pearce, 1998) that will produce higher mechanical strength and melting temperature.

### **2.2.1.2 Hemicellulose**

Along with cellulose in the cell wall, there is hemicellulose, which builds up with a combination of 5- and 6-ring carbon ring sugar polymers. Hemicellulose branched structure consists of a group of polysaccharides which is lower in molecular weight than cellulose (Thakur & Thakur, 2014). Some monomers of hemicellulose are present in Fig. 2.3. Compared with cellulose that is crystalline in structure, hemicellulose has a random and amorphous structure with little strength (Biagiotti et al., 2004). Hemicellulose cements cellulose microfibril to form the main structure of the fiber cell. However, it is easily hydrolysed by acid (Azwa et al., 2013). The reactive surface covered with hydroxyl

groups provides extensive chemical modification (Kalia et al., 2011). Hemicellulose degrades mostly into sugar molecules under wet conditions by acid hydrolysis.

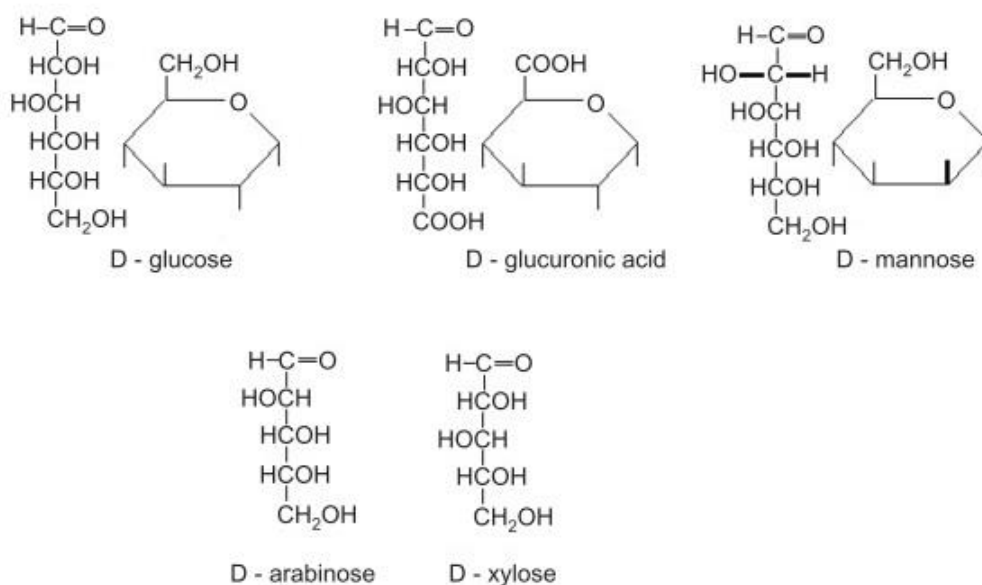


Figure 2.3 Some monomers of hemicellulose.

### 2.2.1.3 Lignin

Lignin was obtained by removing water from sugar to create an aromatic structure by irreversible reaction (Biagiotti et al., 2004). The cell walls of plant fibers are firmly cemented by lignin or hemicellulose. Lignin is a hydrocarbon polymer, giving rigidity to the plant as well as transferring water to the plant (Azwa et al., 2013). Polymers of lignin, types, and proportions depend on the natural source. The typical monomers of lignin are shown in Fig. 2.4 (a), while 2.4 (b) shows the first stage of the condensation process of these monomers to form lignin. The lignin content in the bast fiber influences its plant structure properties and morphology (Bledzki et al., 1996).

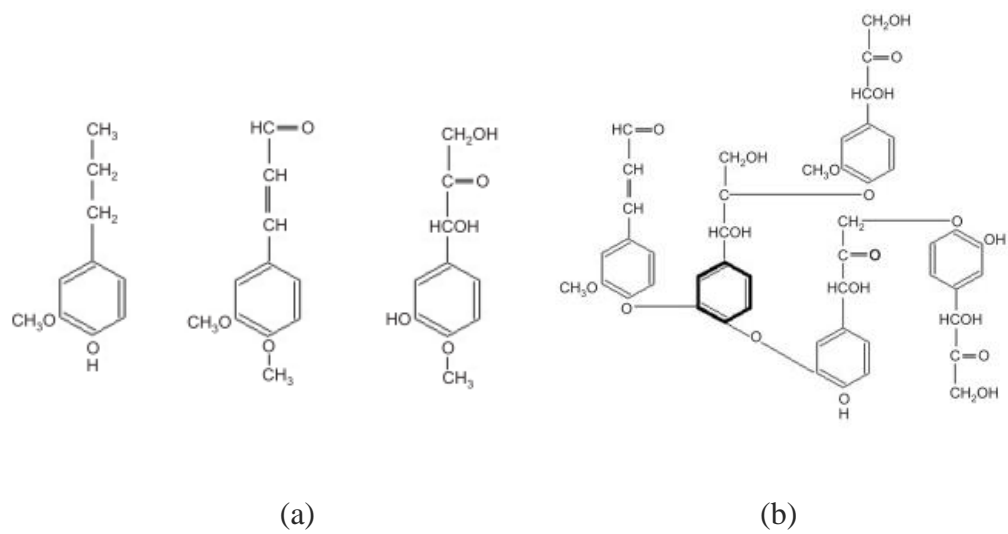


Figure 2.4 (a) The typical monomers of the lignin, and (b) early stage in their condensation.

Table 2.1 Chemical composition and physical properties of selected fibers.

Fiber	Cellulose (wt.%)	Hemicellulose (wt.%)	Lignin (wt.%)	Microfibrillar angle (°)	Density $\rho$ (g/cm <sup>3</sup> )	Tensile strength $\sigma$ (MPa)	Tensile modulus E (GPa)	Specific modulus (E/ $\rho$ )	Ref.
Softwood	30-60	20-30	21-37	-	-	-	-	-	(M. Jawaid & Abdul Khalil, 2011)
Flax	62-72	18.6-20.6	2-5	5-10	1.4-1.5	343-2000	27.6-103	19-68	(Dittenber & Gangarao, 2012)
Bamboo	60.8	30	32	2-10	0.6-1.1	140-800	23-49	38-45	(Dittenber & Gangarao, 2012; Goda, Takagi, & Netravali, 2008)
Kenaf	72	20.3	9	9-15	1.4	223-930	14.5-53	10-38	(André, 2006; Dittenber & Gangarao, 2012; Goda et al., 2008)
Coir	36—43	0.03	41-45	41-45	1.15-1.46	95-230	4-6	3-4	(Dittenber & Gangarao, 2012; Goda et al., 2008)
Sisal	67-78	10-14	8-11	20	1.33-1.5	363-700	9-38	7-25	(Dittenber & Gangarao, 2012; Goda et al., 2008)
Cotton	83-90	5.7	<2	-	1.5-1.6	287-800	5.5-13	4-8	(Dittenber & Gangarao, 2012)
Curaua	71-73.6	9.9	7.5-11.1	-	1.4	87-1150	11.8-96	8-68	(Dittenber & Gangarao, 2012)
E-glass	-	-	-	-	2.5	2000-3000	70	29	(Pickering & Aruan Efendy, 2016)

### 2.2.2 Coir fibers

Coir is a cellulosic plant fiber that is extracted from coconut fruit, and *Cocos nucifera* is its scientific name. Coir fiber presents as fiber bundles since numerous cell walls are in the fiber. As shown in Fig. 2.5, each fiber cell has a narrow and thick wall which is made of cellulose (Harish, Michael, Bensely, Lal, & Rajadurai, 2009). It has a multicellular fiber, consisting of fiber, phloem, and parenchyma cells, the central portion, called “lacuna” is useful for better mechanical interlocking with the matrix during fabrication (Khalil, Alwani, & Omar, 2006; Sreekala et al., 1997). The fiber is chemically amenable, non-toxic, low in density, and not brittle, as well as less abrasive to plastic processing equipment. Likewise, as with most plant fibers, it is kind to the environment. However, it is highly hydrophilic because of the predominance of –OH groups on its surface.

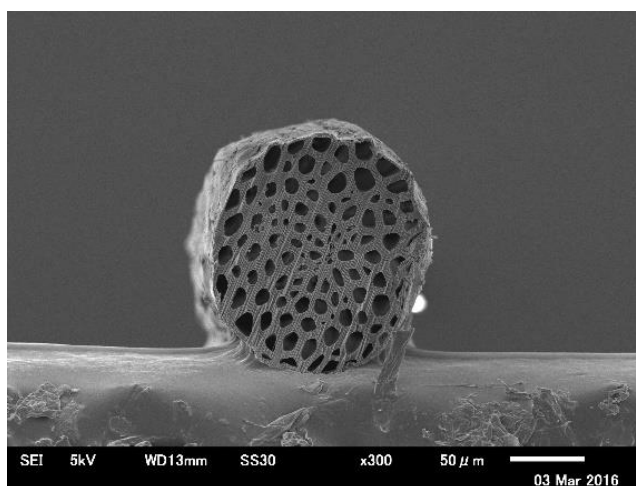


Figure 2.5 Circular shape of coir fiber with hollow, narrow and thick fiber cell walls.

There are many general advantages of coir fibers as follows; they are moth-proof, resistant to fungi and rot, provide excellent insulation against temperature and sound, are not easily combustible, flame retardant, extremely resistant to moisture effects and

dampness, tough and durable, resilient, spring back to shape even after constant use, totally static free and easy to clean (Ali, 2011).

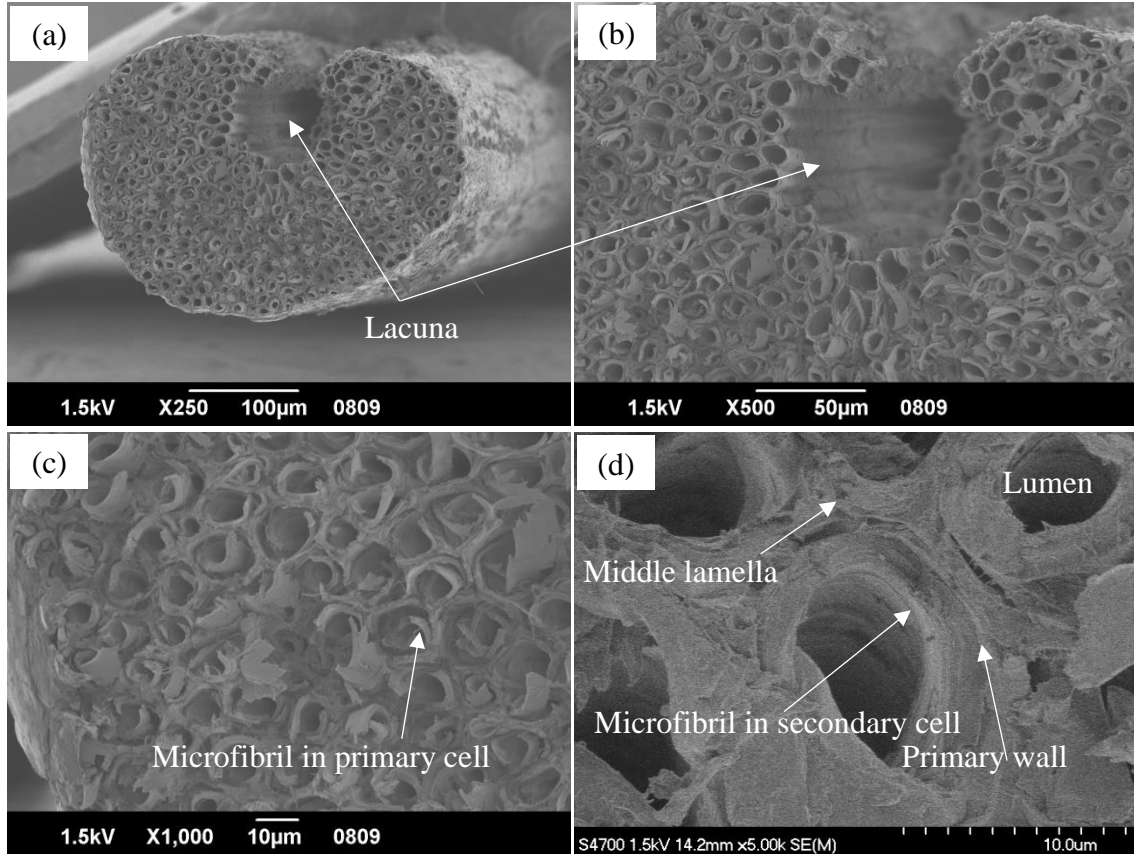


Figure 2.6: (a)-(b) Coir fiber central portion and (c)-(d) several cell walls in coir fiber bundles.

The hollow portion along the fiber axis (lacuna) is an advantage in acoustic and thermal insulating applications, also it reduces bulk density, providing lightweight fiber reinforcement properties. Figure 2.6 details the shape of coir fiber bundles with hollow fiber cells and its internal structure. The elementary fiber consists of two main cells, primary and secondary cell walls, which aligned at a high microfibril angle and vary among the species of the fibers (Table 2.1). Coir fibers have a considerable amount of porosity, ranging between 22-30% (Tran et al., 2014). It was also found that the coir fiber was not stiff, achieved Young's modulus of 4.6 GPa, and may achieve strength of 234

MPa. However, its high strain at break (20-40%) is an advantage in the toughness ability of composites as reinforcements.

### **2.2.3 Bamboo fibers**

Bamboos are grass species and extracted from the hollow and cylindrical stem (tube-like shape). Bamboo is regarded as a potential filler or reinforcement for polymer green composites to replace wood and glass fiber. Its high specific strength is equivalent to conventional glass fibers (Okubo, Fujii, & Yamamoto, 2004) based on low-density fiber. The inherent properties of bamboo fiber such as high tensile strength and modulus, and low elongation at break are derived from the effect of its unidirectional oriented fibers (Jain, Kumar, & Jindal, 1992). The bamboo stem is naturally reinforced by fiber bundles, comprising 50% of the stem cross-section area (Ashby, 2008). The percentage of fiber bundles of the bamboo plant influences hardness, which depends on the number of fiber bundles and the manner of their scattering. It increases from the bottom to the top of the culm (Fei, Fei, Yu, Xiong, & Tan, 2014; Jain et al., 1992). Moreover, the unidirectional configuration in bamboo fiber composites contributes superior mechanical properties, 3-5 times better than short-fiber randomly-oriented composites. The reinforcement efficiency of composites was reported to be achieved by the uni-directionality of bamboo fibers (Shah, 2013).

It was found that steam explosion is an effective method to extract lignin from the stem. However, the surfaces of bamboo fiber bundles have many organic particles (lignin/xylem/ cuticle) as shown in Figs. 2.7 and 2.8. A further mechanical cleaning or alkali treatment would clean the surfaces.

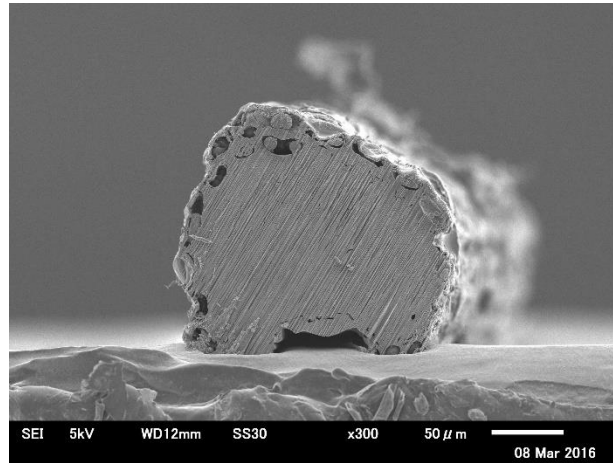


Figure 2.7 Circular shape of bamboo fiber bundles with particles on fiber cell walls.

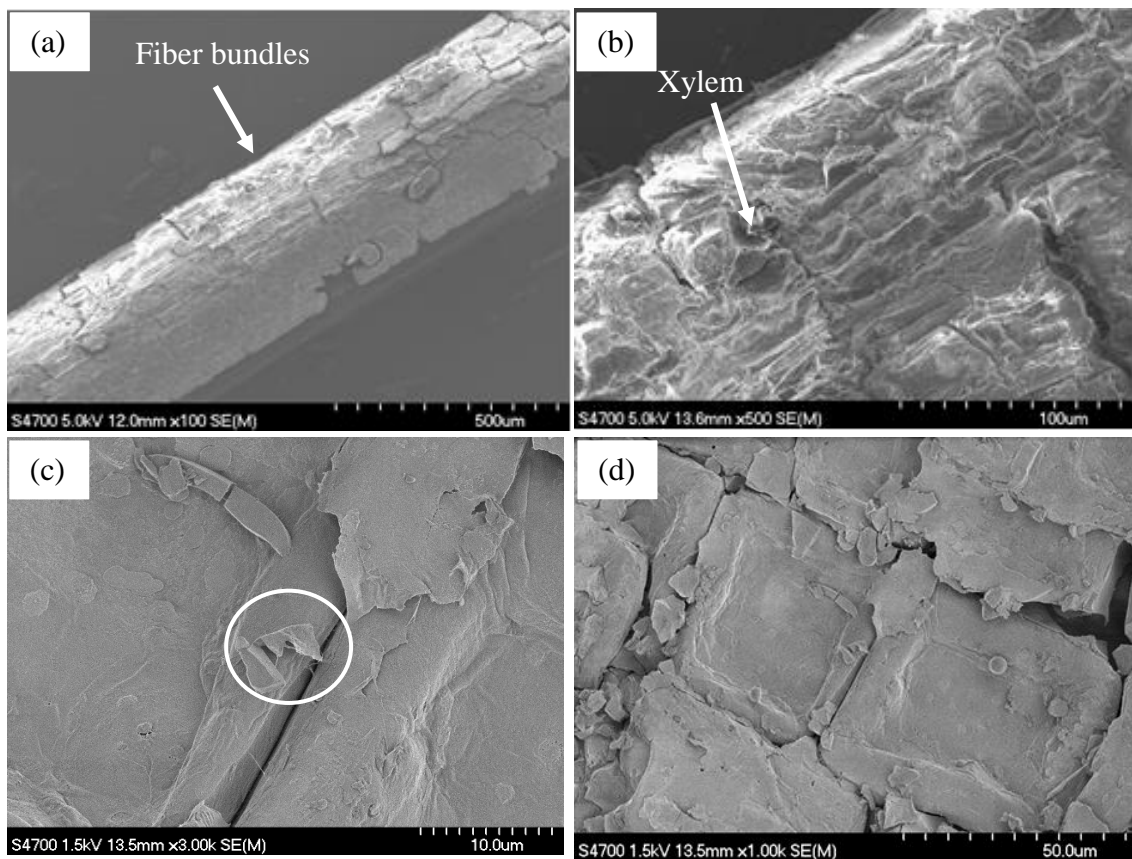


Figure 2.8 Surface condition of bamboo fibers bundle: (a) fiber bundles, (b) raw surface of fiber bundles, (c)-(d) high cuticles existing on bamboo surface.



Also, the organic particles will almost be entirely removed if the fibers are processed in the form of cotton-like fibers, using a mixer multipurpose blender, and improve their properties (Okubo et al., 2004). Due to the high strength and modulus of bamboo fiber, bamboo composites have been used in both structural and non-structural applications to produce building materials, such as bamboo plywood, laminated bamboo lumber, bamboo particle board, furniture and flooring (Yu, Huang, & Yu, 2014).

#### **2.2.4 Kenaf fibers**

Kenaf plant (*Hibiscus cannabinus L.*) is one of 50 species of *Hibiscus (genus)* section of *Furcaria* and was used to produce paper and pulp along with roselle (*Hibiscus sabdaffia L.var altissima*) (Rowell et al., 1996). It consists of two separate parts, core and bast (Jonoobi, Harun, Shakeri, Misra, & Oksmand, 2009). Kenaf bast fiber has better tensile strength than core fiber (H'ng, Khor, Tadashi, Aini, & Paridah, 2009). Moreover, kenaf fiber revealed the high value of both tensile strength and flexural properties compared to other natural fiber-reinforced PLA composites (Akil et al., 2011). However, the extraction of kenaf fiber is time-consuming since the fibers have a small cross sectional area ( $0.01 \text{ mm}^2$ ) with a density of  $0.62 \text{ g/cm}^3$  (Yousif, Shalwan, Chin, & Ming, 2012).

Kenaf is always available in long fiber form and found to be used in many insulator seals (Bernard et al., 2011). Kenaf has the capability to accumulate carbon dioxide at a high rate (Akil et al., 2011). Furthermore, kenaf fiber composites can replace synthetic fiber composites for flexural application (Edeerozey, Akil, Azhar, & Ariffin, 2007; Yousif et al., 2012). Kenaf fiber has to be combed carefully to clean the fibers by

removing the dissimilarity of fiber length and untangle the strong bonding between individual fibers and bast fibers.

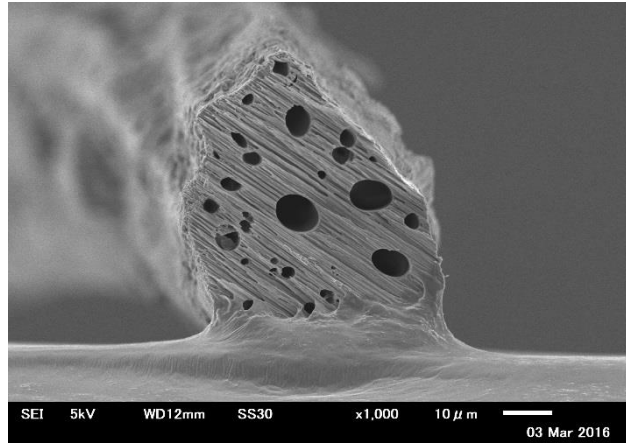


Figure 2.9 Circular shape of kenaf fiber bundles with lumens at fiber cell walls.

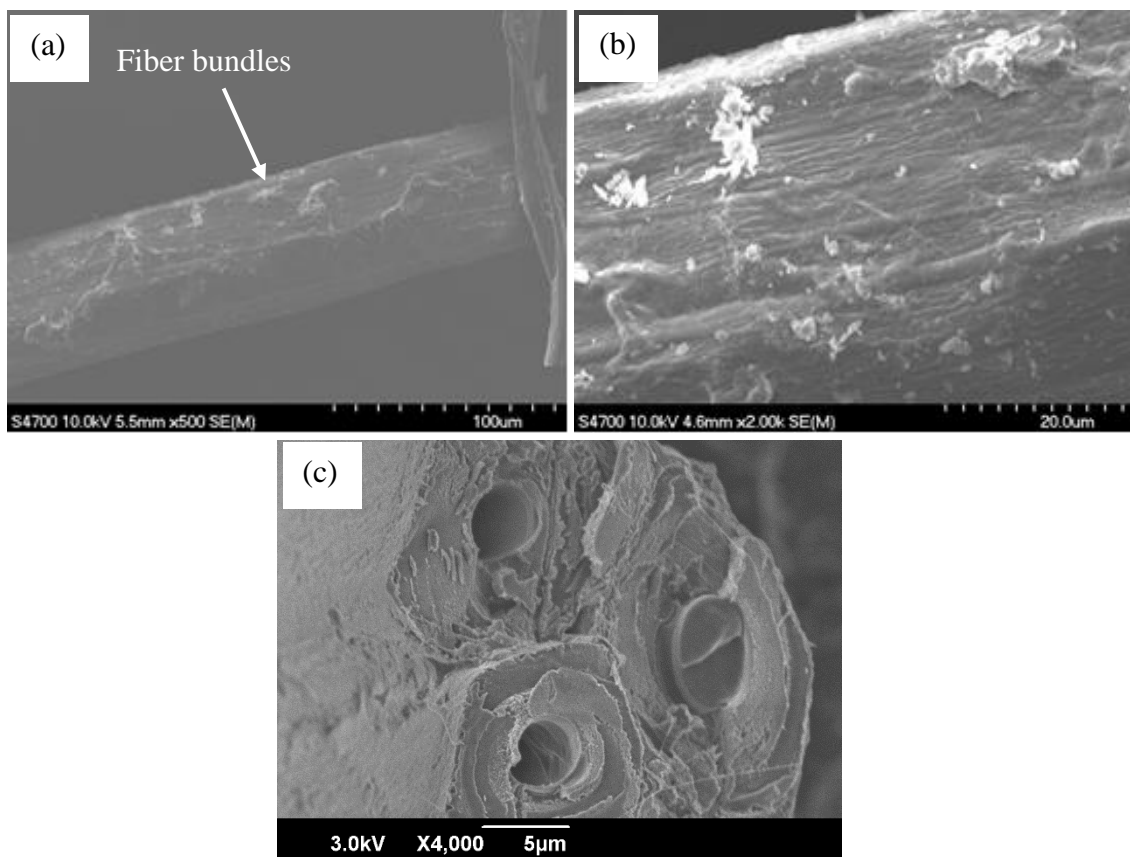


Figure 2.10 Surface condition of kenaf fibers bundle: (a) fiber bundles, (b) raw surface of fiber bundles with organic particles, (c) lumen on fiber bundles.

## **2.3 Polymer matrix**

### **2.3.1 Polylactic acid as a biodegradable matrix**

The biodegradable polymer matrix of polylactic acid (PLA) is frequently used to reinforce plant fibers. Produced via fermentation of corn starch to lactic acid, PLA can be reconverted and naturally decomposed to reduce the environmental impact. Compared to polypropylene (PP), PLA not only exhibits higher modulus but also higher storage modulus and flexural properties (Han, Karevan, Sim, et al., 2012) as well as high mechanical and thermal properties (Masud S. Huda, Drzal, Mohanty, & Misra, 2008; Nishino, Hirao, Kotera, Nakamae, & Inagaki, 2003), which is comparable to polystyrene (Suryanegara, Nakagaito, & Yano, 2009).

The polymer matrix provides a barrier against adverse environments and mechanical abrasion. It influences the mechanical properties by transferring load to fibers, particularly at initial stress condition. Polylactic acid (PLA) has high mechanical and thermal properties and forms a strong bond with kenaf fibers (Masud S. Huda et al., 2008; Nishino et al., 2003).

## **2.4 Composites**

Composites are made from fibers and matrix, characterized by the reinforcements used, which are particulate, fiber, laminate and hybrid composites. Filler or particulate composites are used for high temperature, friction resistance and to reduce shrinkage. Fibers such as carbon, aramid and glass fibers are the synthetic fibers used to produce synthetic composites or conventional composites. However, fibers from plant-based fibers, such as jute, kenaf, flax, bamboo, etc. are considered green composites, and are environmentally friendly. To produce the laminated composites, stacking of fiber ply in

certain directions alternatively is applied to improve better adhesion both of fiber and matrix, aiming for high strength of composites. The laminated layers are applied mostly in monolithic and hybrid composite fabrication which are suitable for structural applications that evolved from a monolithic structure to a multicomponent composite to be fabricated as the top layer, core, and base layer (Andreas, 2007). The stress distribution across the top and core layers is dominated by individual material properties, such as yield strength and Young's modulus. The higher modulus layers will carry a greater portion of total stress.

The performance of composites is basically based on some factors, for example the fiber's individual strength, alignment and laminating configuration, fiber length and fabrication method used. As for individual fibers, high strength and stiffness, a small diameter (Monteiro, S. et al., 2011), and a volume fraction between 60-70% (Campbell, 2010) were recommended. Apart from that, a continuous unidirectional configuration (Fei et al., 2014; Okubo et al., 2004; Salleh, Berhan, Hyei, & Isaac, 2012; Shah, 2013) yields a higher strength of composites than short fiber reinforced composites.

The matrix itself can be classified into three materials, which are metal, ceramic and polymer matrix composites (Ichhaporia, 2008). However, polymer matrix is the research interest in this study due to its low cost and low environmental impact.

#### **2.4.1 Monolithic green composites made from polylactic acid**

This monolithic concept was used based on the previous study (Buchmeiser, 2007; Gnanapragasam, Chitra, & Ravi, 2016; Nema, Chan, & Ho, 2014; Paci, Flores, & Ayson, 2014). Monolith was derived from the Greek word "monos," which means "single" and

lithos, which means “stone”, and thus referred as the column consisting of a single large block of stone (Nema et al., 2014). In general, the basic nature of materials used for preparation was classified as monolithic. In polymer synthesis, monolithic materials were used to refer to the alternative packed column based on beaded polymers or inorganic oxides as polymeric devices in separation science as well as in another area of chemistry (Buchmeiser, 2007). Therefore, in these green composites terms, it represents a composite made from continuous plant fiber reinforced with thermoplastic.

PLA is a thermoplastic polymer that has been used to be reinforce continuous plant fibers. These composites have been discussed widely and provide high composite strengths as shown in Table 2.2. Although they have a high variability in properties, PLA polymers ease modification with plant fibers and enhance brittleness. The size of testing samples, fiber orientation, fiber content, fiber treatment, fiber length and fabrication methods are a major influence on composite strengths. Polylactic acid or polylactide is polyester of lactic acid, fermented from renewable agricultural materials. Then, via a cyclic dilactone, lactide, ring opens polymerized to the polylactic acid polymer (Oksman, Skrifvars, & Selin, 2003). This polymer can be improved for temperature stability by reinforcement with fibers.

Commonly, the selection of individual plant fibers determines the end properties of green composites, along with other qualities such as strength of interfacial bonding, fiber’s alignment and fiber’s orientation (Pickering & Aruan Efendy, 2016). For example, a unidirectional PLA matrix reinforced with yarn flax fibers was found to have superior mechanical properties as a consequence of highly ordered treated fibers and PLA matrix modification in the composites (Duhovic, Horbach, & Bhattacharyya, 2009; McGregor et al., 2010). Fibers such as kenaf, coir, bamboo, sisal and jute have been used in consumer

goods, low cost housing and structural parts due to their high specific properties, comparable to glass fiber (Ramesh et al., 2013; Sudhir et al., 2014; Zhang et al., 2013) and conventional materials (Gosline, Guerette, Ortlepp, & Savage, 1999). The comparison with other synthetic materials is also presented in Table 2.2.

#### **2.4.2 Hybrid green composites**

Hybrid composites are defined as composites produced by the incorporation of two or more fibers in a matrix (John & Thomas, 2008). The combination of two or more plant fibers in a biodegradable matrix is known as a hybrid green composite. The high mechanical properties of these composites have made them popular in material design, as they overcome traditional disadvantages. To obtain the desired properties of hybrid green composites, the compatibility between constituent fibers and the matrix is the prime concern. For optimal hybrid green composites, the plant fibers should be highly strained compatible (Sreekala, George, Kumaran, & Thomas, 2002), determined by a MFA (Barnett & Bonham, 2004; Bledzki et al., 1996). Fiber with low MFA typically produces high strength and stiffness.

Composites containing few types of fiber are targeted to improve composites properties rather than single type fiber composites. The various fiber-reinforced composites may resist load by compensating for low properties of individual fibers reinforcement (Kistaiah, Kiran, Reddy, & Rao, 2014; Sathishkumar et al., 2014). For instance, high elongation of coir fibers improves failure strain level required to break (Dong & Davies, 2014a; Zhang, Chaisombat, He, & Wang, 2012), while high strength and modulus in kenaf and bamboo incorporate between the fibers to improve strength properties of components. Hybridization in material design has been a popular method

for plant/synthetic fiber-reinforced polymer composites to overcome the disadvantages of moisture absorption and poor adhesion with matrices to obtain high mechanical properties of the composites (Alex & Retnam, 2014; Almeida, Amico, Botelho, & Amado, 2013; Satyanarayana et al., 2009). Nevertheless, there are a few studies based on the combination of different kinds of plant fiber-reinforced biodegradable polymer in hybrid green composites instead of hybrid plant fibers/synthetic-based composites (Ramesh et al., 2013; Naheed Saba, Tahir, & Jawaid, 2014; Sathishkumar et al., 2014; Sudhir et al., 2014; Zhang et al., 2013).

Table 2.2 Comparison of tensile and flexural strengths of single green composites/hybrid composites (plant/synthetic fiber).

Composites plant/ synthetic fiber	Tensile strength (MPa)	Young's modulus (GPa)	Strain at break (%)	Flexural strength (MPa)	Flexural modulus (GPa)	Ref.
Kenaf/PLA (40wt.%)	-	-	-	98	7	(Han, Karevan, Bhuiyan, Park, & Kalaitzidou, 2012)
Kenaf/PLA (40wt.%)	-	-	-	115	6.64	(Han, Karevan, Sim, et al., 2012)
Kenaf/PLA (40wt.%)	-	-	-	40	6	(Masud S. Huda et al., 2008)
Kenaf/PLA (40wt.%)	-	-	-	-	-	(Akil et al., 2011)
Kenaf/PLA (70vol.%)	223	24	-	254	23	(Ochi, 2008)
Kenaf/PLLA (70vol.%)	60	6.4	2	-	-	(Akil et al., 2011)
Kenaf/PP (40wt.%)	44	-	-	72	-	(Akil et al., 2011)
Coir/PP (40wt.%)	10	-	-	28	-	(Akil et al., 2011)
Coir/PLA (20wt.%)	5	1.5	0.7	25	3.2	(Y. Dong et al., 2014)
Coir/PLA (30wt.%)	18	0.4	3.4	20	1.4	(Y. Dong et al., 2014)
Bamboo/CP-300 starch-based (50wt.%)	45	-	-	58	-	(Takagi & Ichihara, 2004)
Flax/PLA (65wt.%)	332	45.6	-	-	-	(McGregor et al., 2010)
Flax/PLA (65wt.%)	165	22	-	-	-	McGregor et al., 2010)
Flax/PLA (~40wt.%)	257	18	-	194	13	(Duhovic et al., 2009)
Flax/glass fiber/phenolic	393-450	40	1.0	-	-	(Zhang et al., 2013)
Jute/sisal/epoxy	40	1.6	5	88.33	3.5	(Sudhir et al., 2014)
Sisal/glass fiber/epoxy	69	-	-	-	-	(Ramesh et al., 2013)
Jute/glass fiber/epoxy	63	-	-	-	-	(Ramesh et al., 2013)



# CHAPTER THREE

## Experimental

### 3.1 Introduction

We used biodegradable PLA polymer and three different plant fibers (kenaf, bamboo, and coir fibers) to investigate the mechanical properties of fiber bundles, and their single and hybrid composites. Each type of fiber-reinforced PLA composite had similar total fiber content, which is up to 70 wt.%. These composites were fabricated by a hot-press molding method. We carried out tensile tests on fiber bundles and, tensile, flexural, and impact tests on single and hybrid green composites and observed the effects on the composites' microstructures through optical and scanning electron microscopy. The composites were examined for mechanical properties to compare with single composites, plant/glass fiber composites and conventional materials. The damping loss factor was obtained from Center Impedance Method, evaluating their loss factor to compare with that of glass fiber reinforced composites. A water absorption test was also conducted to evaluate the weight change due to immersion in water.

### 3.2 Materials

#### 3.2.1 Matrix polylactic acid (PLA)

A dispersion-type PLA resin (PL 2000) with fine particle (4.5 $\mu$ m average diameter) was used in this study and was supplied by Miyoshi Oil & Fat Co. Ltd., Japan. The melting point of the PLA is 167 to 170°C, with D-lactide and L-lactide content of 10

and 90 wt.% respectively and its density is 1.23 g/cm<sup>3</sup>. The melt flow index of PLA is 10 g/min at 210°C test temperature and 2.16 kg test load.

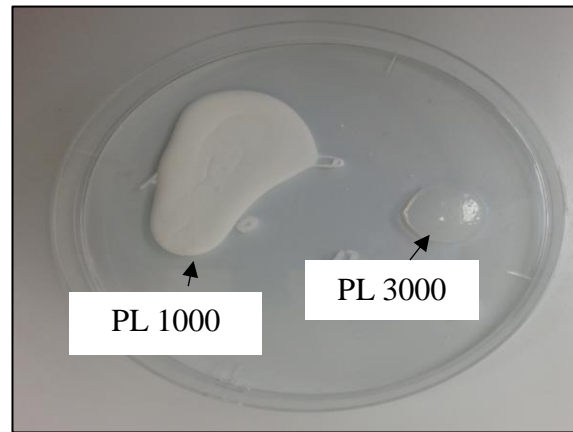


Figure 3.1 PLA mixture of PL 1000 and PL 3000.

### 3.2.2 Plant fibers

Coir fibers were obtained from Kamenoko-Tawashi Nishio-Shoten Co., Ltd. Japan. The bamboo culms were freshly harvested in Anan City, Tokushima, Japan, and bamboo fibers were extracted by a steam explosion method. Kenaf cultivated in Kelantan, Malaysia was supplied by Lembaga Kenaf dan Tembakau Negara (LKTN). All fibers were used in as-received condition, namely without any surface treatment. Each fiber type was measured in similar weight percentage based on the total weight of fiber of hybrid composites. The cross-sectional area of the fiber bundles was assumed as circular since all the cross-sections were approximately circular (Figs. 3.2). The diameter of each fiber bundle was measured at three different positions on both sides of the fiber as mentioned (Cai et al., 2015; Yusoff, Takagi, & Nakagaito, 2016). A digital microscope equipped with a 300x magnification lens (VHX-600, Keyence Co. Ltd., Japan) was used to measure the diameter of the fibers.

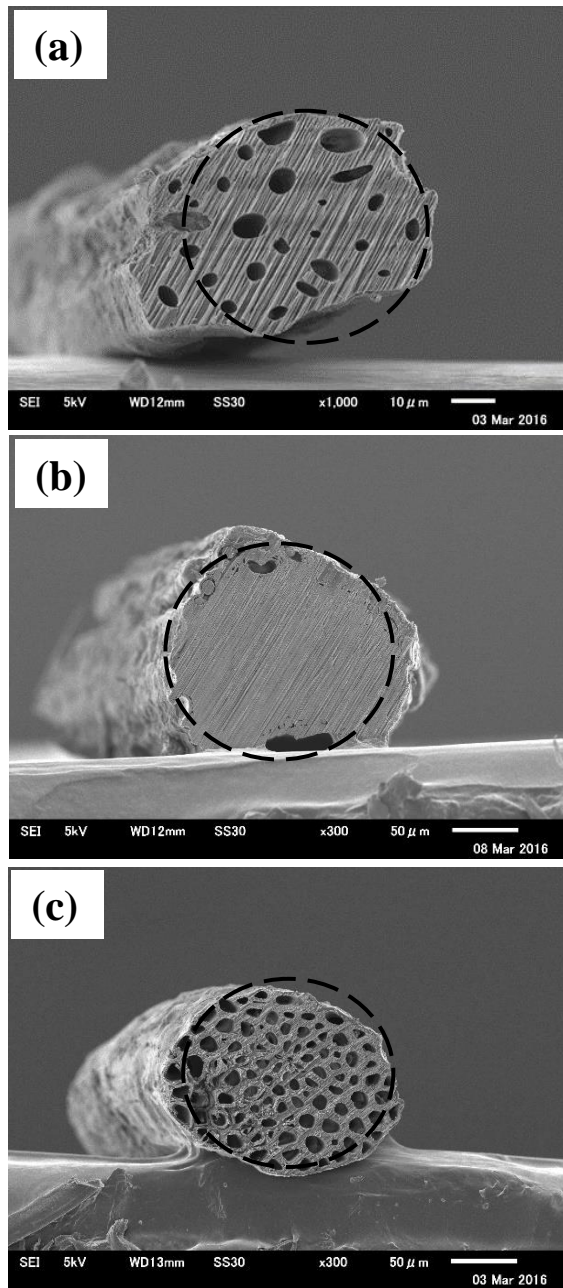


Figure 3.2 Cross-sectional views of (a) kenaf, (b) bamboo and (c) coir fiber bundles.

### 3.3 Methods

#### 3.3.1 Density measurement

In the first study, the density of the fibers was measured by using an electronic densimeter (MD-300S, Alfa Mirage Co. Ltd. Japan), and then a water pycnometer. Fig. 3.3 and Fig. 3.4 show the tools for the density measuring. Figure 3.5 illustrates the step of density measuring. The fibers were cut into approximately 40 mm lengths and the fiber bunches were put on the machine. The density, weight, and the volume were recorded based on the electronic densimeter software, based on Archimedes principle equation for determining the fibers' density.

As for the pycnometer method, it was done according to D 854-02 by means of a water pycnometer (Method B-oven dried specimen). The tests were conducted with cutting the fibers into 2-5 mm length, approximately 10-15 pieces of 100 mm length of fiber bundles being cut for one density test. And these fibers' particles were dried in an oven (105°C) for 1 hour. For the density measurement, firstly, the mass of the empty pycnometer was measured. Secondly, it was measured continuously with filled water, measured with particles fiber bundles and the mass of pycnometer filled with water and particles fiber bundles. For 20 minutes, the vacuum was used when the pycnometer and particles fiber bundles filled with water, leading to the water filling into the lumen and removing the trapped air between the fibers to avoid false results. The pycnometer was filled 1/3 full with the sample material to measure the density according to the equation (3.1):

$$\rho_S = \frac{(m_2 - m_0)}{(m_1 - m_0) - (m_3 - m_2)} \times \rho_w \quad (3.1)$$

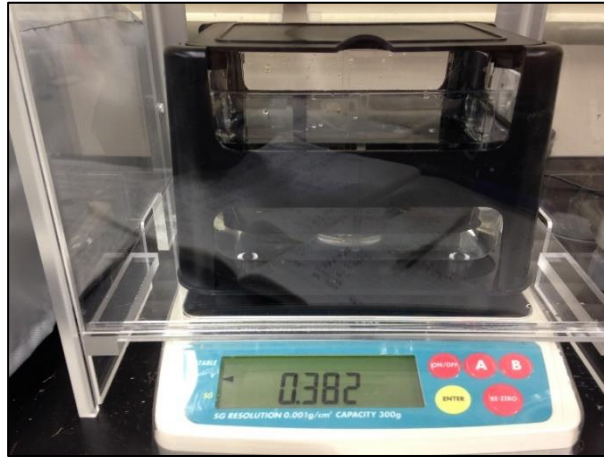


Figure 3.3 Electronic densimeter.



Figure 3.4 Pycnometer.

where,  $m_0$  is the mass of the empty pycnometer,  $m_1$  is the mass of the pycnometer filled with water,  $m_2$  is the mass of the pycnometer with the solids,  $m_3$  is the mass of the pycnometer with the solids and filled with water,  $\rho_w$  is the density of water.

This is the measurement of cell wall density since the measurement of  $m_2$  only cell wall was measured as the lumen was filled with air. The lumen for  $m_3$  is filled with water because the measurement procedure used a vacuum to remove the trapped air between

the fibers to avoid false results. This vacuum also leads to the water filling the lumen. The density also was measured based on the equation (3.2):

$$\rho = \frac{m}{V}, \quad (3.2)$$

where  $\rho$ ,  $m$ , and  $V$  are density, mass and volume of a composite specimen.

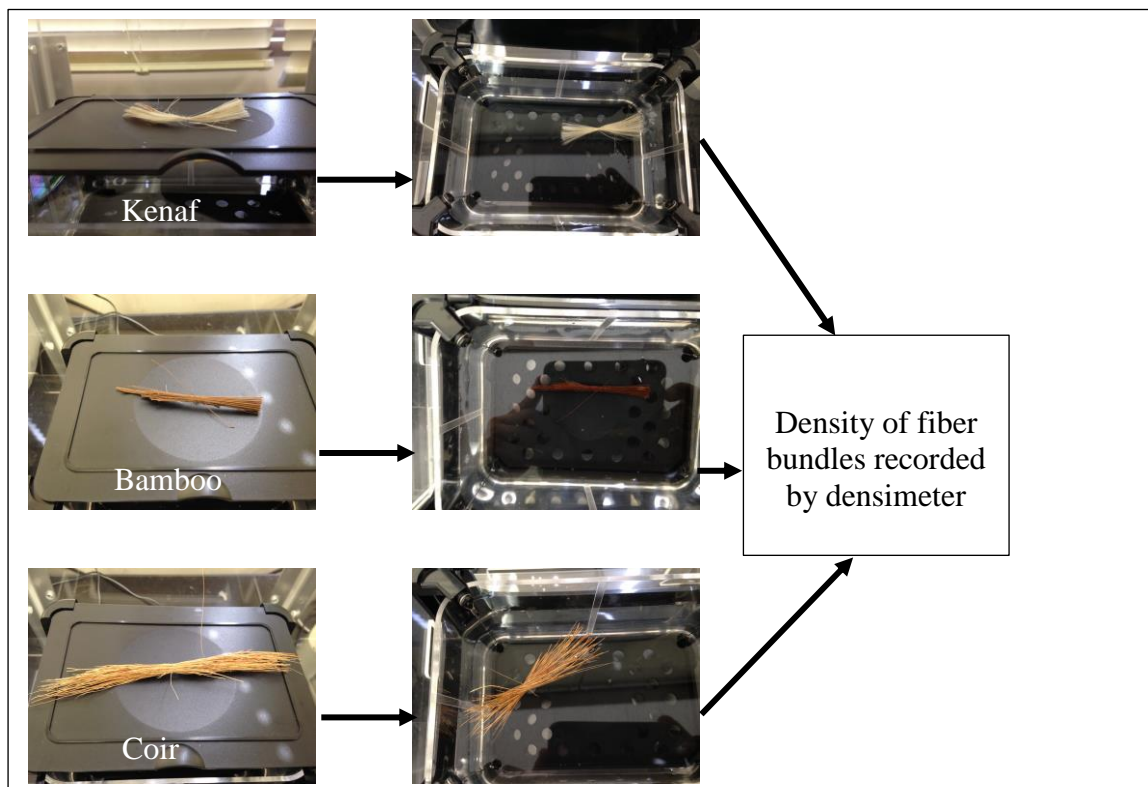


Figure 3.5 Step to determine the density using densimeter.

### 3.3.2 Fiber cleaning

Simple cleaning by using water retting, brush and wet cloth was used to remove the particles on the outer surface of fibers. This is only to increase the adhesion of matrix with fibers, especially in the case of bamboo fibers which have many impure substances on their surfaces.

### 3.3.3 Single fiber bundles tensile testing

To investigate the initial properties of plant fibers, untreated separated long fiber bundles were used. Tensile tests of fiber bundles were conducted to estimate the mechanical properties of composites to decide the stacking configuration of hybrid green composites. This information was based on the high correlation between the properties of single fibers and fiber bundles, as well as the accuracy fracture behavior as reported by R'Mili et al. (R'Mili et al, 2008). A paperboard was used to perform the tensile test. A fiber bundle was attached to the paperboard frame as shown in Fig. 3.6. Bamboo and coir fibers were randomly and gently withdrawn from bunches of fibers, while kenaf fiber was withdrawn and combed gently to separate the fiber bundles. Fibers were cut approximately 45 mm long and then glued to the paperboard frame to remain straight and to prevent moving during gripping. Each of the tensile specimens was tested on a 5567 Universal Testing Machine (Instron Corporation, U.S.A.). Tensile strength, Young's modulus and strain at break were obtained from stress-strain curves recorded during tensile tests.

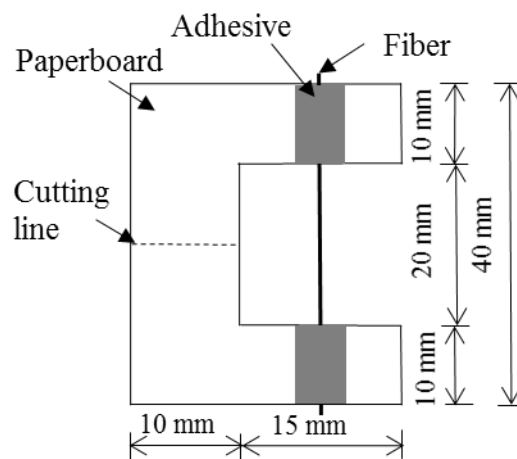


Figure 3.6 Schematic of paperboard frame for single fiber testing.

### 3.3.4 Fabrication of monolithic green composites and hybrid green composites

Since several types of composites were tested in this research, the stacking patterns and the reinforcement types are referred to by the fibers' names (K=kenaf, B=bamboo, and C=coir). Monolithic green composites were prepared from a single type of plant fibers. Firstly, the single green composites were fabricated to evaluate the properties of individual fibers, which was believed to influence the theoretical estimation of strength and modulus, using the rule of mixtures (ROM) and the hybrid rule of mixtures (HROM). The single composites types (Fig. 3.7) were tested as follows:

- (a) bamboo/PLA,
- (b) coir/PLA and
- (c) kenaf/PLA

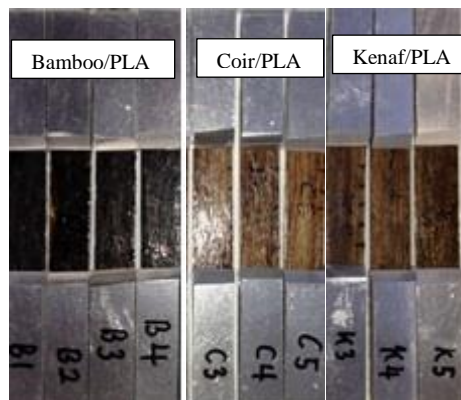


Figure 3.7 Monolithic green composites.

Secondly, several types of hybrid green composites were prepared. One was comprised of three kinds of fiber types, at 60wt.% fiber content for the initial properties of hybrid composites.

- (a) KCCK/PLA
- (b) BCCB/PLA
- (c) KBCCBK/PLA



Finally, the other hybrid was comprised of three kinds of fibers in the composite as follows:

- (a) KBC/PLA (from 50 to 70wt.% of fiber content)
- (b) CBK/PLA (from 50 to 70wt.% of fiber content)

Basically, a similar fabrication process was used for both single and hybrid green composites as illustrated in Fig. 3.8. Except for the drying process of fibers/PLA. They were done in three different conditions which are at room temperature, 30°C and 50°C under a vacuum condition. Apart from that, the fibers used were different, according to stacking sequences of the composites. The nomenclature and processing parameters of composite samples are listed in Table 3.3.

For all composites types, the untreated coir, bamboo and kenaf fibers were used for the fabrication. Firstly, each type of fiber was cut into 120 mm-long pieces and the weight was measured in order to insure equal weights based on the total fiber content in a composite. In contrast, for the mold of 200 mm x10 mm size, 220 mm of fibers were cut and the dried at 50°C and a vacuum condition was applied.

Initially, all fibers were cleaned to remove the non-cellulosic substances on the fibers' surfaces. For bamboo fibers, a wet cloth and brush were applied. Then, the fibers were impregnated with the PLA suspension (equally mixed with PL-3000 and PL-1000). Secondly, they were dried at room temperature for 24 hours. Prior to hot-pressing, the fibers and PLA were further dried in an oven for 1 hour at 105°C. Finally, a hot-pressing method was used to mold the hybrid green composites. The hot-pressing was heated by setting the temperatures at 210°C and 185°C for the upper and the lower plates, respectively. The mold was placed onto the hot pressing until the temperature reached

170°C, then the temperature was maintained for ten minutes, and a pressure of 10 MPa was applied for 5 minutes. The mold was finally removed from the hot-pressing machine and allowed to cool down to room temperature under low pressures.

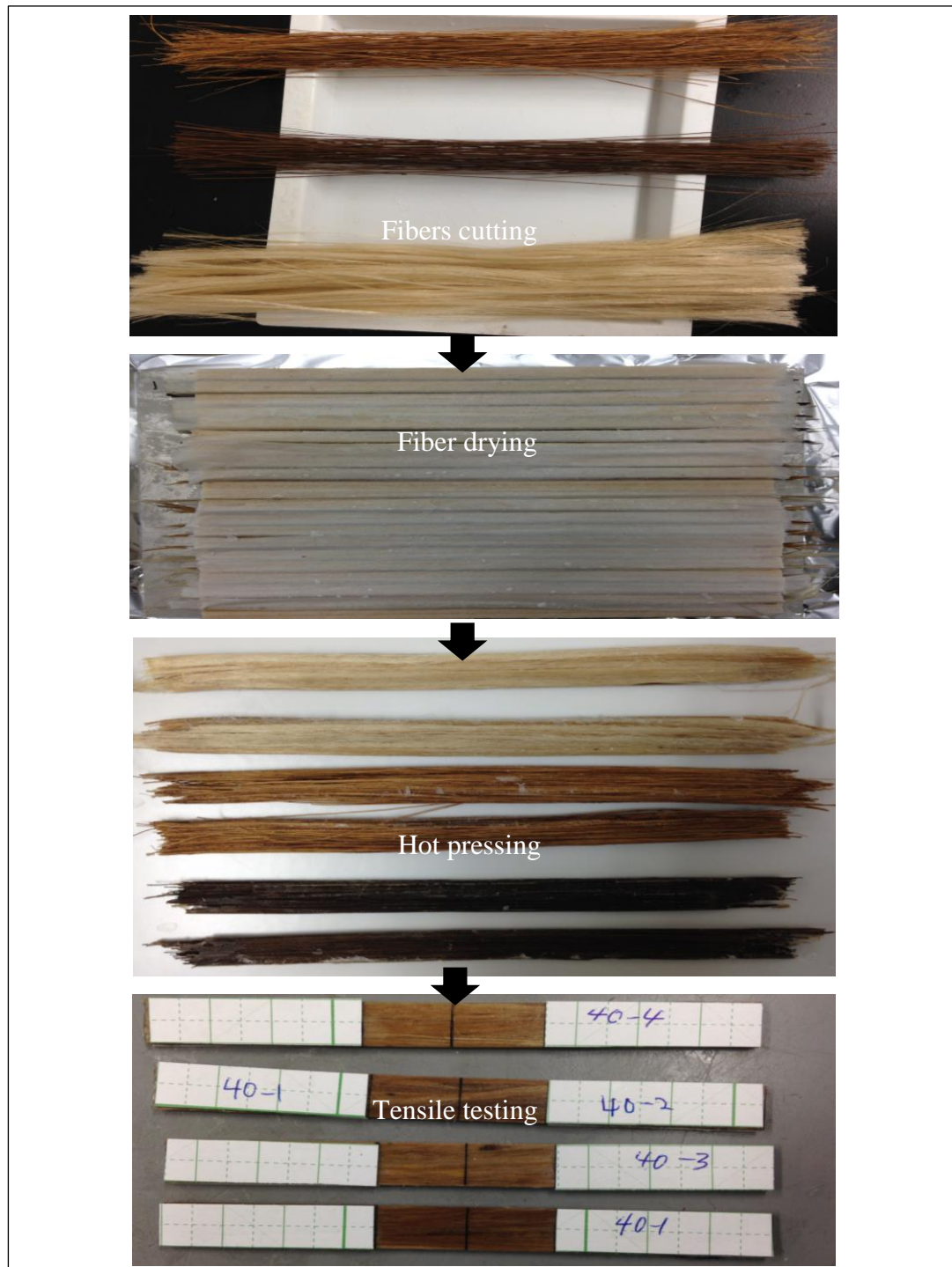


Figure 3.8 Fabrication of hybrid green composites.

Table 3.3 Nomenclature and processing parameters used for various composite sample testing.

Abbreviation	Composites	Processing parameters
Kenaf/PLA	Kenaf fiber-reinforced PLA composites	Drying temperature : 30°C Mold size: 100 x 10 x 1.6±0.2 mm
Bamboo/PLA	Bamboo fiber-reinforced PLA composites	Drying temperature : 30°C Mold size: 100 x 10 x 1.6±0.2 mm
Coir/PLA	Coir fiber-reinforced PLA composites	Drying temperature : 30°C Mold size: 100 x 10 x 1.6±0.2 mm
KCCK/PLA	Kenaf and coir fiber-reinforced PLA hybrid green composites	Drying temperature: room temperature Mold size: 200 x 10 x 2±0.2 mm
BCCB/PLA	Bamboo and coir fiber-reinforced PLA hybrid green composites	Drying temperature : room temperature Mold size: 200 x 10 x 2±0.2 mm
KBCCBK/PLA	Kenaf, bamboo and coir fiber-reinforced PLA hybrid green composites	Drying temperature : (1) room temperature (2) 50°C at vacuum Mold size: 200 x 10 x 2.2±0.3 mm
CBK/PLA	Coir, bamboo and kenaf fiber-reinforced PLA composites	Drying temperature : 30°C Mold size (mm): 100 x 10 x 2±0.2 mm
KBC/PLA	Kenaf, bamboo and coir fiber-reinforced PLA hybrid green composites	Drying temperature : 30°C Mold size (mm): 100 x 10 x 2±0.3 mm

### 3.3.5 Mechanical testing of composites

#### 3.3.5.1 Tensile testing

The tensile strength, strain at break, and Young's modulus of the samples were determined by a tensile software, Instron Bluehill 2, version 2.33 (Instron Corporation, U.S.A.). The initial linear slope of tensile stress-strain curve presented the Young's modulus of the materials while maximum stress and strain were obtained from the point where materials fail (Gosline et al., 1999).

### 3.3.5.2 Tensile toughness

The tensile toughness was calculated from underneath the area of stress-strain curves, which presented energy absorption to break per unit volume of the materials. The rectangular composites were gripped at a 30 mm gauge length and the crosshead speed was set at 1.0 mm/min. All the parameters were obtained by averaging five experimental runs.

Rule of mixtures (ROM) and the hybrid rule of mixtures (HROM) were used to estimate the elasticity (Young's modulus) and tensile strength of hybrid green composites. The rule of mixtures (Hull, 1981) for monolithic green composites were derived from equation (3.3).

$$E_c = E_f V_f + E_m V_m \quad (3.3)$$

$$V_f = \frac{\frac{w_f}{\rho_f}}{\frac{w_f}{\rho_f} + \frac{w_m}{\rho_m}} \quad (3.4)$$

and since

$$V_m = 1 - V_f \quad (3.5)$$

$$E_c = E_f V_f + E_m (1 - V_f) \quad (3.6)$$

where  $E, V, w$ , and  $\rho$  are the Young's modulus, volume fraction, weight fraction and density, respectively. The subscripts  $c, f$  and  $m$  refer to composite, fiber and matrix, respectively.

Likewise, the Young's modulus of hybrid green composite consisting of three types of fibers and matrix was determined by equation (3.7):

$$E_H = E_c V_{fc} + E_b V_{fb} + E_k V_{fk} + E_m (1 - V_{fc} - V_{fb} - V_{fk}) \quad (3.7)$$

It is also appropriate to express the stress on a laminate parallel to fibers as the following equation (3.8):

$$\sigma_H = \sigma_c V_{fc} + \sigma_b V_{fb} + \sigma_k V_{fk} + \sigma_m (1 - V_{fc} - V_{fb} - V_{fk}) \quad (3.8)$$

where  $\sigma_H$  is tensile strength of hybrid green composite,  $\sigma_m$  is the tensile strength of matrix at the composite's strain at break,  $\sigma_c, \sigma_b$  and  $\sigma_k$  are tensile stress of coir, bamboo and kenaf fibers at the composite's strain at break. While  $V_{fc}, V_{fb}$  and  $V_{fk}$  are the volume fractions of coir, bamboo and kenaf fibers, respectively.

Similar to the rule of mixtures consideration, this expression should also assume that fibers are distributed throughout the matrix, a perfect bonding between matrix and fibers and the load applied is parallel to the fibers' direction.

### 3.3.5.3 Flexural testing

Three point flexural tests were carried out based on Japanese Industrial Standard (JIS K7017:1999) to determine the maximum stress induced in the outermost fibers. Span length was 40 mm based on the ratio of 20 of span length and the thickness (L/h). Flexural strength was calculated from equation (3.9).

$$\sigma = \frac{3FL}{2bh^2} \quad (3.9)$$

where F is the maximum load (N); L is the distance between the supports or span (mm); b is the width (mm) and h is the thickness (mm) of the specimen.

### 3.3.5.4 Izod impact testing

The Izod impact strength was measured according to JIS K7110 with a modified cross section of the test specimen of 10 mm x 2 mm with 80 mm long. Notched specimens

had a 2 mm deep 45 degree notch angle at distance 40 mm from the top. The average value of impact strength of the fractured specimens were valuated based on equation (3.10).

$$\text{Impact strength} = \frac{W}{hb_N} \times 10^3 \quad (3.10)$$

where,  $W$  = absorb energy (joule), and  $hb_N$  = cross section of notched specimen ( $\text{mm}^2$ )

The impact energy,  $W$  is determined by the equation (3.11).

$$W = R \left[ (\cos\beta - \cos\alpha') - (\cos\alpha' - \cos\alpha) \left( \frac{(\alpha+\beta)}{(\alpha+\alpha')} \right) \right] \quad (3.11)$$

where  $R$ ,  $\beta$ ,  $\alpha$  and  $\alpha'$  are the height of hammer to break specimen, final point of swing angle to break the specimen, initial point of swing hammer without specimen, final point of swing hammer without specimen, respectively.

### 3.3.5.5 Water absorption testing

The test was performed based on D 570-98 standard. The size of 20 mm x 20 mm of test samples was prepared. The cut section of the specimens were painted. From each sample, at least 5 specimens were immersed in water at 50 °C for 2 hours and 48 hours. Weight increased was calculated from equation (3.12).

$$\text{Weight change} = \frac{\text{Wet weight} - \text{Initial weight}}{\text{Initial weight}} \times 100 \quad (3.12)$$

### 3.3.5.6 Loss factor

Plant fiber is promising for noise barrier and acoustic absorption (Zhu, Kim, Wang, & Wu, 2014). For reduction of noise, the damping materials which have viscoelasticity characteristics are useful in automotive and other industry products. A system is naturally loose energy due to heat loss during physical deformation and as a consequences of friction with air. In an oscillating system, loss of energy is known as damping ( $\eta$ ). Mass and stiffness of materials are responsible for the loss of energy from the system. Unlike mass and stiffness, damping is hard to predict. The most common techniques to measure the loss factor are Oberst method, flexural vibration test and resonant beam technique (Carfagni, Lenzi, & Pierini, 1994).

The damping can be expressed by different parameters, such as damping ratio ( $\zeta$ ) and loss factor ( $\eta$ ). These parameters can be obtained from the dynamic mechanical analyzer, according to Japanese Industrial Standards, JIS K 7391. Center impedance method or Center Excitation Method was also used which is easy to measure requiring only the standard equipment as Fig. 3.9. This method can measure the frequency and damping ratio directly and the experiment can be repeated. The size of specimens was approximately 200 mm x 10 mm with the thickness of approximately 2.2 mm. They were glued and placed in the center and the evaluation was based on first mode frequency.

Several definitions of  $\eta$  are feasible (Carfagni et al., 1994; Crane & Gillespie, 1989; Gelfuso, Thomazini, de Souza, & de Lima, 2014), one of those is the energy ratio of damping energy loss per radian with peak strain energy associated to the vibration as follows (Carfagni et al., 1994):

$$\eta = \frac{D}{2 \pi W} \quad (3.13)$$

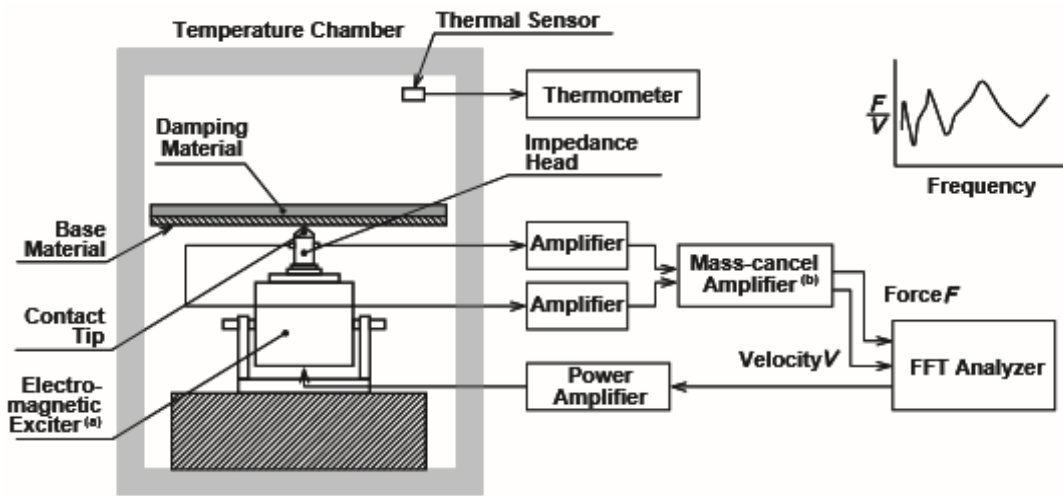


Figure 3.9 Central Excitation Method (JIS K7391).

Vibration system can be classified into three categories by their nominal maximum frequency: (1) low frequency (50 to 60 Hz), (2) intermediate frequency (500 to 900 Hz), and (3) wide band (2000 to 5000 Hz) (Curtis, Tinling, & Abstein, 1971). The loss factor can be determined directly from this experiment's software. Also, at first resonance frequency, the Young's modulus can be obtained.



# CHAPTER FOUR

## Result and discussion-Part 1-Fibers and green composites characterization

### 4.1 Introduction

The lightweight density of composites is the most preferable in engineering production rather than their strength in a certain application, such as automotive parts, products' housing, and accessories, for their dynamic moving and easy handling. For the preliminary study, this chapter presents the characteristics of:

- Physical properties of individual fiber bundles.
- The density of fiber bundles and their composites, and PLA matrix.
- Tensile properties of:
  - Fiber bundles,
  - Neat PLA,
  - Bamboo/PLA,
  - Coir/PLA, and
  - Kenaf/PLA.
- Surface morphology of fiber bundles and green composites.

### 4.2 Physical properties of fiber bundles

#### 4.2.1 Surface structures

The internal structure of the fiber cell walls provides stability to the composites since failures are governed by fracture of the cell walls in fiber bundles (Kulkarni,

Satyanarayana, Sukumaran, & Rohatgi, 1981). They are not usable as fibers alone and are used to impregnate a polymer matrix. Bonding formation occurs at wetting and flow of the polymer matrix into the fiber pores, and ends up in the cell wall, polymerization or the reaction with the cell wall components in the cell promotes an interphase region (Frihart, 2006). On the other hand, lumen exists in the secondary layer of the cell wall interlocking the fiber cell walls with the matrix, promoting cohesion (Khalil et al., 2006), as existing in kenaf and coir fibers as shown in Fig. 4.1. These kinds of surfaces may help fiber bundles to attract matrix PLA that will increase the brittleness of a matrix.

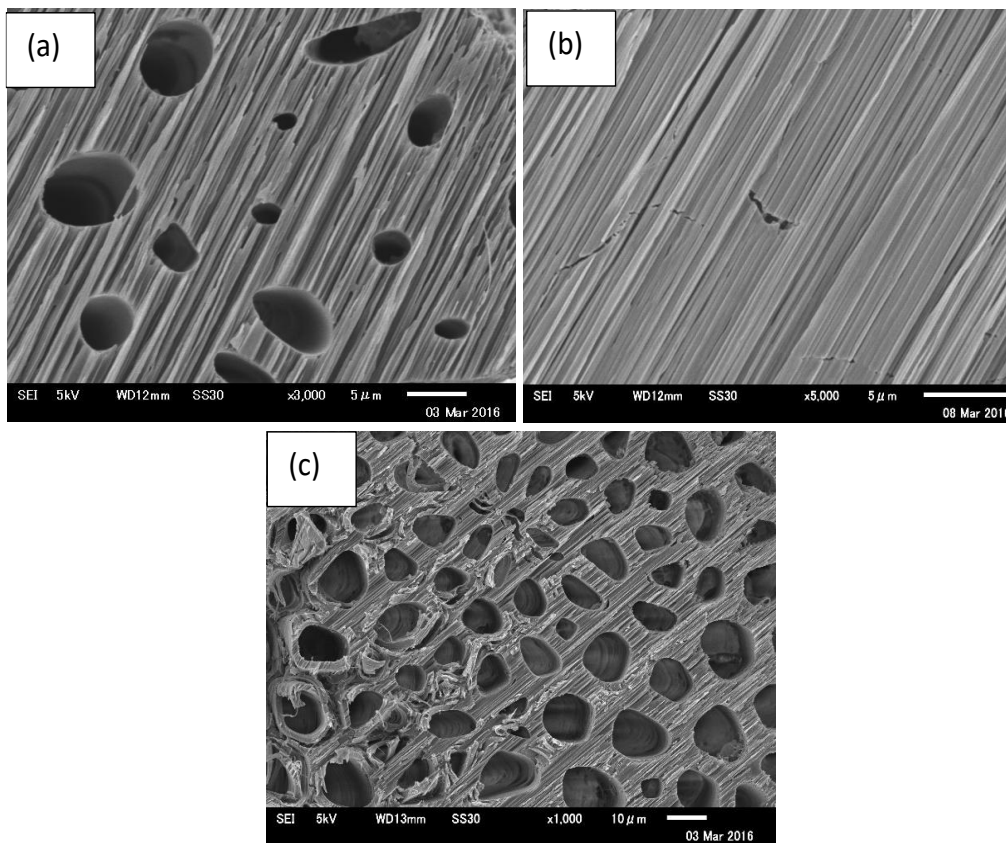


Figure 4.1 Cross-sectional views of (a) kenaf, (b) bamboo, and (c) coir fiber bundles.

#### 4.2.2. Density of kenaf, coir and bamboo fiber bundles

The density of each fiber bundle was measured by using electronic densimeter and water pycnometer due to the safety, low cost and ease of use, however, it was reported to produce an inaccurate result (Truong, Zhong, Boyko, & Alcock, 2009). Table 4.1 indicates that coir has the lowest density, followed by kenaf and bamboo as measured by densimeter. Coir fiber was floating in the water, thus it has to be measured using the water pycnometer to obtain the exact value for further study. Based on the water pycnometer method, kenaf had the highest density among bamboo and coir fibers. This is the effect of kenaf fibers being able to absorb air from the surroundings. Since a vacuum was used during density measuring, water penetrated into the cell walls, thus increasing density.

Table 4.1 Density of kenaf, bamboo and coir fiber bundles.

Fiber bundles	Densimeter (g/cm <sup>3</sup> )	Water pycnometer (g/cm <sup>3</sup> )	Literature (g/cm <sup>3</sup> )
Kenaf	1.18±0.11	1.64±0.3	0.62 (Yousif et al., 2012)
Bamboo	1.23±0.04	1.47±0.3	1.44-1.56 (Li et al., 2010)
Coir	0.85±0.03	1.2±0.24 1.30±0.1	1.15-1.37 (Ali, 2011) 1.25 (Kistaiah et al., 2014)

#### 4.2.3 Density of PLA, monolithic and hybrid green composites

The density of the composites was calculated based on a theoretical calculation mentioned in the previous section (equation 3.2), which is the ratio of mass and volume of the final composite produced after hot-pressing. The specimen size of 100 mm x 10 mm x 1 mm was measured to calculate the density.

Table 4.2 shows the appearance density of specimens of single composites and hybrid green composites based on 60 wt.% fiber content. In the case of single composites, coir/PLA has the lowest density of the composites, influenced by the hollow microstructure of the fiber cell walls. A higher density was obtained in kenaf/PLA, followed by bamboo/PLA. Neat PLA has the highest density compared to all the composites. Based on our observation, kenaf/PLA has the capability to adhere to the PLA matrix better than coir and bamboo fibers. PLA acts as glue to kenaf fibers. But bamboo hardly performed with PLA as it is rich in chemical compounds on surfaces and was squeezed out during PLA melting in hot pressing. Overall, all weights of the single and hybrid green composites are lightweight density, approximately 1 g/cm<sup>3</sup>, with the dimension of 100 mm x 10 mm x 1 mm.

Table 4.2 Characteristics of single and hybrid green composites measured by the theoretical.

Materials	Length (mm)	Thickness (mm)	Width (mm)	Ave. Weight (g)	Density (g/cm <sup>3</sup> ) $\rho = \frac{m}{V}$
PLA	100	1	10	1.4	1.23
Bamboo/PLA	100	0.92	10.12	1.1	1.16
Coir/PLA	100	1.03	10.1	1.03	1
Kenaf/PLA	100	0.96	10.09	1.1	1.19
KBCCBK /PLA	100	1.2	10.1	1.1	1

Table 4.3 displays composites' densities obtained from the software of Electronic Densimeter (MD-300S), which aimed to provide the precise value of composite densities. A similar result was obtained from coir/PLA that remained at the lowest density among other composites.

Table 4.3 Characteristics of single and hybrid green composites measured by Electronic Densimeter (MD-300S).

Composite (60wt.%)	Moisture content (%)	Weight (g)	Volume (cm <sup>3</sup> )	Density (g/cm <sup>3</sup> ) Electronic Densimeter
Bamboo/PLA	1.93	0.03	0.035	1.15
Coir/PLA	8.77	0.05	0.065	0.79
Kenaf/PLA	9.95	0.055	0.065	0.905
KBCCBK/PLA	-	0.11	0.09	0.99

Comparing both methods, approximately 21-29% higher density was measured using the theoretical equation than the electronic densimeter. This is because of many voids presented in PLA composites and the hollow structures of coir and kenaf fiber bundles were not measured theoretically.

Table 4.4 Comparison of PLA, single and hybrid green composites density based on the ratio of mass and specimen volume and Electronic densitometer (MD-300S).

Density/ Composites	$\rho = \frac{m}{V}$	Electronic Densimeter (MD-300S)	Difference
PLA	1.23	0.95	28%
Bamboo/PLA	1.16	1.15	1%
Coir/PLA	1	0.79	21%
Kenaf/PLA	1.19	0.905	29%
KBCCBK/PLA	1	0.99	1%

### 4.3 Tensile properties

#### 4.3.1 Fiber bundles of coir, bamboo and kenaf

The maximum stress-strain curves of the selected three types of plant fiber bundles are presented in Fig. 4.2. Bamboo fiber bundles achieved the highest tensile strength, approximately 440 MPa, followed by kenaf fiber bundles with approximately 400 MPa, while coir fiber bundles showed the lowest strength of 44 MPa. However, coir fiber bundles depicted higher strain at break compared to bamboo and kenaf fiber bundles. As shown in Fig. 4.3, the average Young's modulus (stiffness) of fiber bundles after tensile testing is high; both bamboo and kenaf fiber bundles have much higher Young's modulus than coir fiber bundles, which are 20 GPa and 22 GPa, respectively. The high Young's moduli of these fibers contribute to produce better stiffness in the green composites, while coir fiber has low modulus and high strain has potential to produce high elongation and toughness. These characteristics affect the flexural properties of the composites as well.

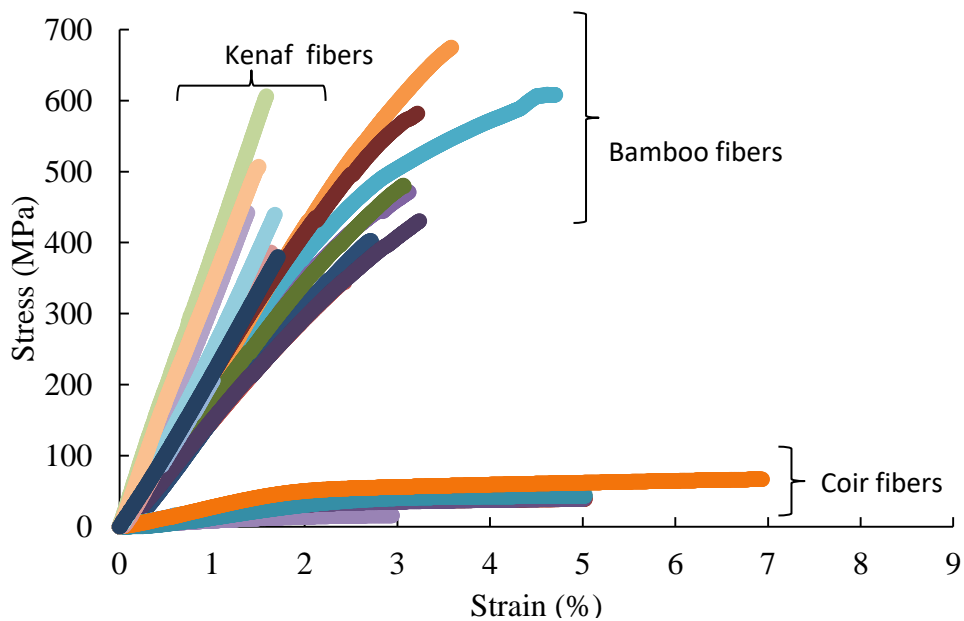


Figure 4.2 Typical tensile stress-strain curves of selected fiber bundles.

The tensile properties of the fiber bundles are related to the MFA of these fiber bundles as shown in the previous Table 2.1. It is well known that the fiber's stiffness decreases with the increase of the MFA, however, it linearly increases with cellulose content (Bledzki, Sperber, & Faruk, 2002). Coir fiber had the lowest stiffness between bamboo and kenaf fibers as it has low MFA in the cell walls. Its inherent property of high elongation is assumed to add some extra properties of the new composite such as to be used in hybrid green composite that may produce high deformability. Table 4.5 gives the average tensile strength of fiber bundles.

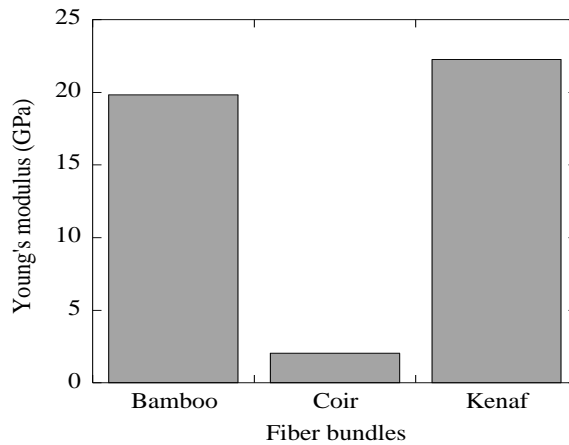


Figure 4.3 Stiffness of bamboo, coir and kenaf fiber bundles.

Table 4.5 Average tensile strength of bamboo, coir and kenaf fiber bundles.

Properties	Fibers		
	Bamboo	Coir	Kenaf
Diameter ( $\mu\text{m}$ )	480 $\pm$ 60	380 $\pm$ 90	100 $\pm$ 21
Tensile strength (MPa)	490 $\pm$ 90	44 $\pm$ 8	280 $\pm$ 90
Young's modulus (GPa)	20 $\pm$ 4	2.0 $\pm$ 0.3	22 $\pm$ 6
Strain at break (%)	2.8 $\pm$ 0.7	4.5 $\pm$ 0.8	1.29 $\pm$ 0.20

### 4.3.2 PLA matrix

Since PLA colloidal suspension was mixed with PL 1000 and PL 3000, the effect of the stirring process was compared. Without the stirring process, the higher tensile strength and Young's modulus were obtained. In contrast, with the applied stirring process, the tensile strength and Young's modulus decreased. However, it produced a higher strain property compared to the PLA without the stirring process. Table 4.6 and Table 4.7 show the details of the PLA matrix without stirring, and with stirring process. Both properties were used for estimation of strength's properties based on the theoretical of ROM and HROM.

Table 4.6 Average tensile strength of PLA matrix (without stirring condition).

Properties	Neat PLA
Tensile strength (MPa)	25±4
Young's modulus (GPa)	1.68±0.25
Strain at break (%)	2.2±0.7

Table 4.7 Properties of PLA matrix (stirring condition).

Properties	PLA-2000 (PL1000 + PL3000)
Tensile Strength (MPa)	6.3 ±0.2
Strain at break (%)	20±12
Young's modulus (GPa)	0.4±0.1
Impact strength (kJ/m <sup>2</sup> )	2±0.6
Flexural strength (MPa)	3±0.3
Flexural modulus (GPa)	0.07±0.01

In terms of PLA properties such as the effect of pressure and temperature of the hot pressing mold, PLA almost maintained its tensile strength and stiffness, approximately achieved at 25 MPa and 2 GPa respectively. Figure 4.4 illustrates its



properties based on temperature and pressure (with or without pressure). This is based on the unstirred condition of PLA colloidal suspension.

According to this result, it was estimated that the properties of PLA have minimal effect on the pressure of hot pressing temperature when a similar type of PLA is used. However stirring condition has an effect on the tensile properties of PLA since PLA particles in water were not dissolved evenly in colloidal suspension matrix during mixing with PL 1000 and PL 3000 due to the high viscosity of PL 3000. It was found that at up to 190 °C, PLA flows out to the mold’s surfaces, resulting in low matrix content and turns into high fiber content. Higher fiber (low matrix) content eases water absorption and increases the possibility of fiber agglomeration as well as increases porosity in the composites (Pickering & Aruan Efendy, 2016).

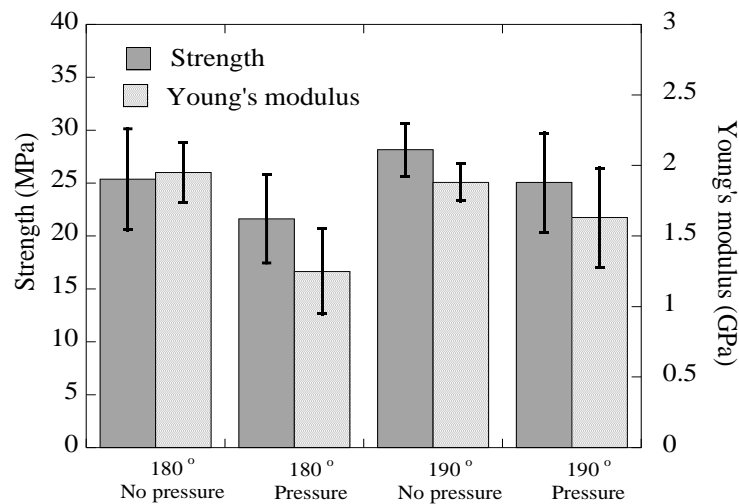


Figure 4.4 Tensile strength of PLA after hot pressing fabrication by means of temperature and pressure.

### **4.3.3 Monolithic green composites**

Monolithic green composites were used to estimate the compatibility between the one type of fibers and matrix. This is also beneficial to estimate the theoretical strength and stiffness of the composites by ROM (Mansor, Sapuan, Zainudin, Nuraini, & Hambali, 2013). The thickness of the composites was fixed, at approximately 100 mm x 10 mm x 1.6 mm (tolerance  $\pm 0.02$ ). PLA with stirring condition was used in this fabrication.

#### **4.3.3.1 Coir fiber-reinforced PLA composites (Coir/PLA)**

As shown in Fig. 4.5, the strength of coir/PLA was constant, accounting for 44 to 55 MPa, and the Young's modulus achieved up to 2 GPa with up to 70wt.% fiber content. Kulkarni et al. (1981) reported that there were no significant changes in coir fiber between the retted and unretted fibers. In their studies, there was a slight decrease in stiffness with the increased diameter of the fibers. Based on the similar observation, cell walls' fracture and uncoiling of MFA were the cause of the initial failure of fibers. In relation to the fiber content, the increase of coir fiber reinforcement had a slight effect on tensile properties of the composites.

Referring to Fig. 4.6, Coir/PLA had 3% of strain at break up to 70wt.% fiber content. This value is lower than those of bamboo and kenaf fiber composites. In fact coir has high strain ability when referring to its fiber bundles. The fabrication process, such as fibers' drying condition, matrix selection and fibers' cross section and dispersion may have an effect in decreasing the mechanical properties (Pickering et al., 2016).

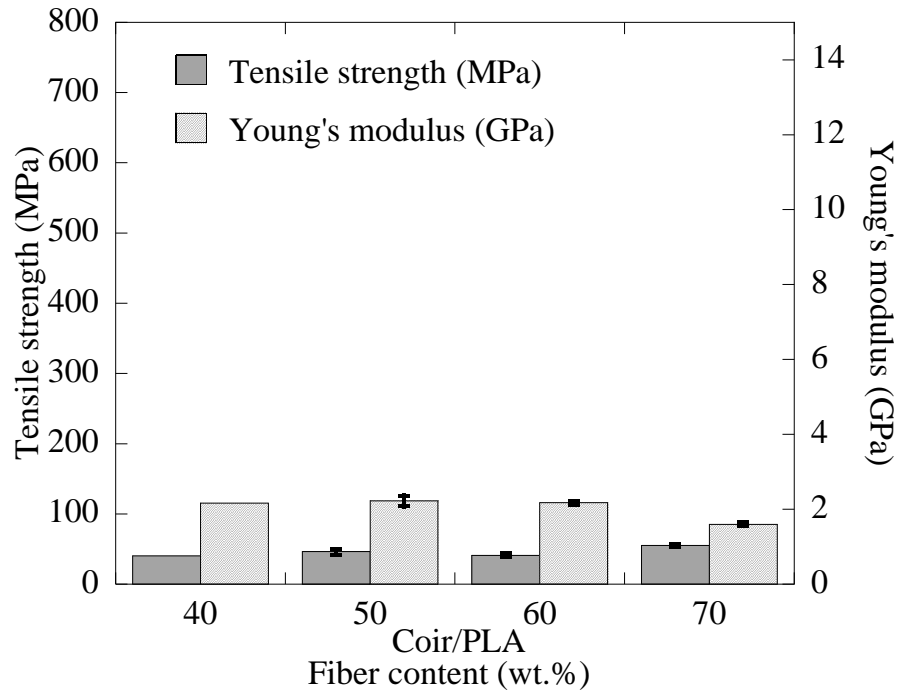


Figure 4.5 Tensile strength of coir fiber reinforced PLA as a function of fiber content.

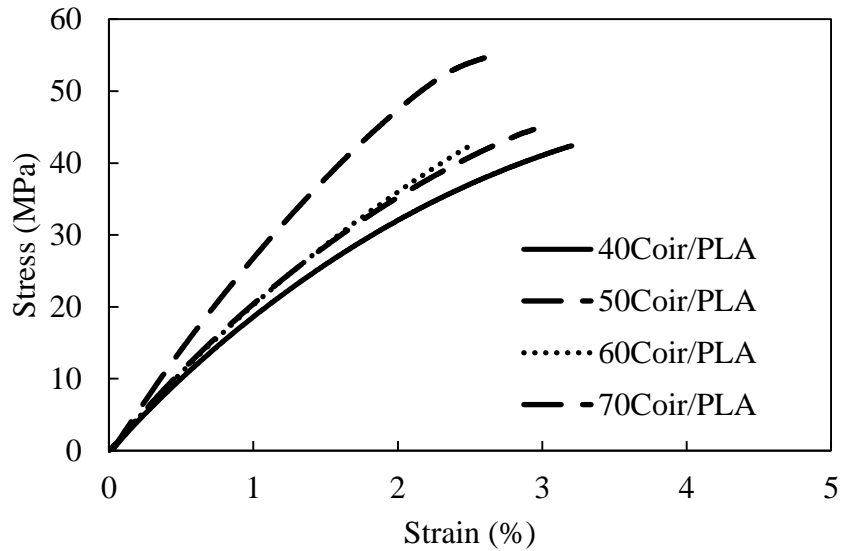


Figure 4.6 Typical stress-strain curves of coir fiber reinforced PLA at 40, 50, 60 and 70 wt.% fiber content.

#### **4.3.3.2 Bamboo fiber-reinforced PLA composites (Bamboo/PLA)**

Bamboo/PLA strength properties are presented in Fig. 4.7. It is noticeable that bamboo fiber composites had unstable properties with up to 50 wt.% fiber content. Bamboo/PLA had high and constant stiffness, approximately 9 GPa up to 70 wt.% fiber contents. Tensile strength increased with fiber content with a slight decrease in strength at 50 wt.% fiber content before it continued to increase in strength. This may be affected by bamboo fiber that was hardly bonding during fabrication, at both pre-pregs and in the hot pressing condition. The untreated bamboo fibers contain compound substances that were squeezed out along with PLA during hot-pressing, leaving mostly pure bamboo fiber in the composites. Therefore, at the content of bamboo fibers lower than the PLA matrix content, the fibers were unable to embed with the matrix, and were not evenly distributed between the fibers and the matrix. Consequently, it affected the tensile properties. The PLA matrix was also mixed with substances on bamboo fiber surfaces instead adhering to fibers.

However, via the compressing molding method, the substances on bamboo fibers' surfaces were beneficial to increase the thermal stability, and bamboo fillers were reported to improved strengths (Okubo, Fujii, & Yamashita, 2005). Adding bamboo fiber as a filler with PLA has increased bending strength and enhanced thermal stability (Tokoro et al., 2008). While the substance compound leads to imperfect bonding, it has also improved the thermal stability by the using bamboo fillers as a solution.

According to Fig. 4.8, a high strain obtained from higher bamboo fiber content, at 60 and 70 wt.% fiber content, it is likely to have a similar strength. A small crack is

noticeable from nonlinear stress-strain curves, which means a weak interfacial bonding between bamboo fibers and matrix.

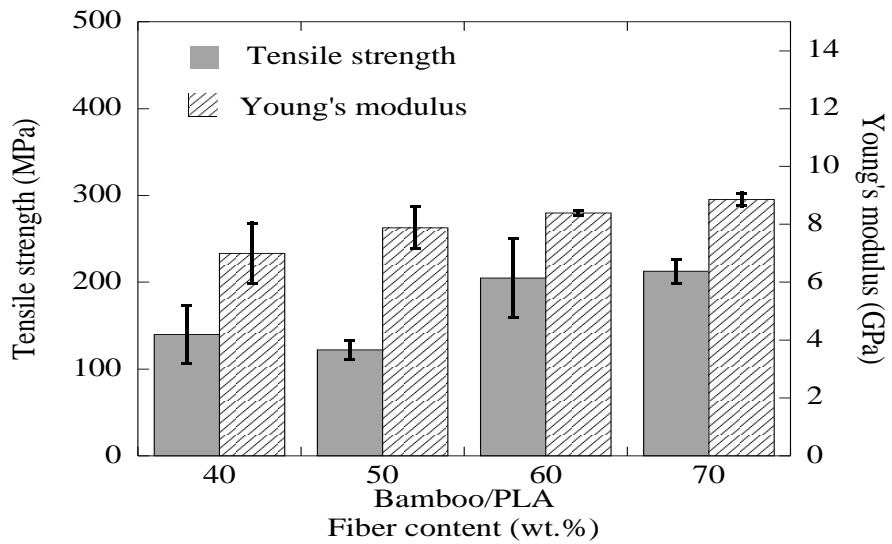


Fig. 4.7 Tensile properties of bamboo fiber reinforced PLA as a function of fiber content.

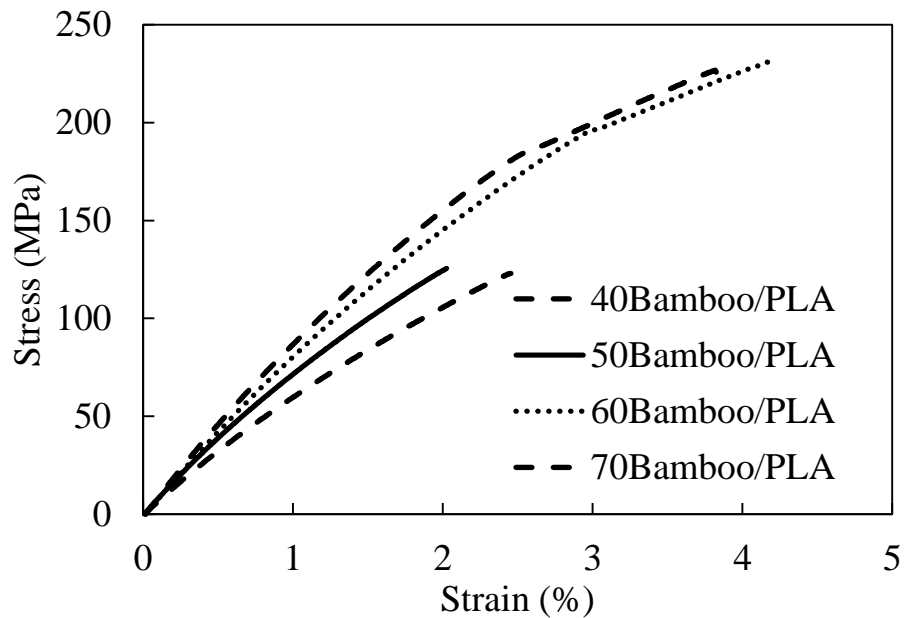


Figure 4.8 Typical stress-strain curves of bamboo fibers reinforced PLA at 40, 50, 60 and 70 wt.% fiber content.

### 4.3.3.3 Kenaf fiber-reinforced PLA composites (Kenaf/PLA)

Tensile properties of kenaf/PLA show the increase in strength and stiffness with fiber content up to 70 wt.% as shown in Fig. 4.9. The compatibility between matrix and fiber yields high properties of the composites. As shown in Fig. 4.10, kenaf/PLA has almost 3% to 5% strain property which increased with fiber content. Kenaf/PLA has a strong adhesion with PLA, thus leading to higher mechanical properties compared to coir and bamboo fiber reinforced PLA. In addition, a small cross-sectional area of kenaf fiber propagates the reinforcement efficiency of the composites.

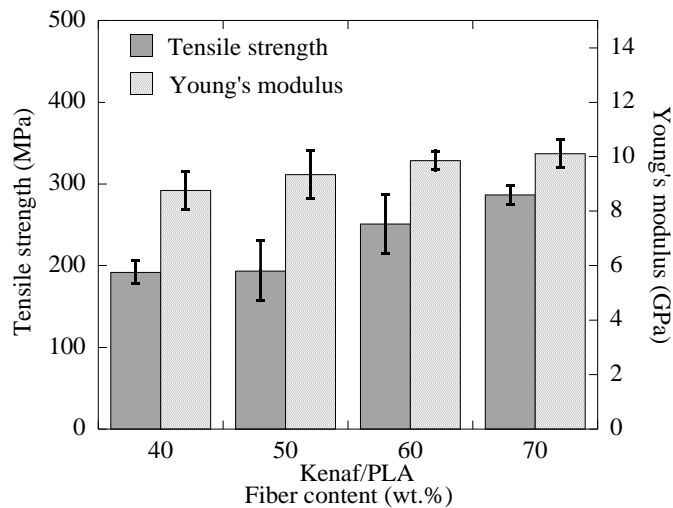


Figure 4.9 Tensile properties of kenaf fibers reinforced PLA as a function of fiber content.

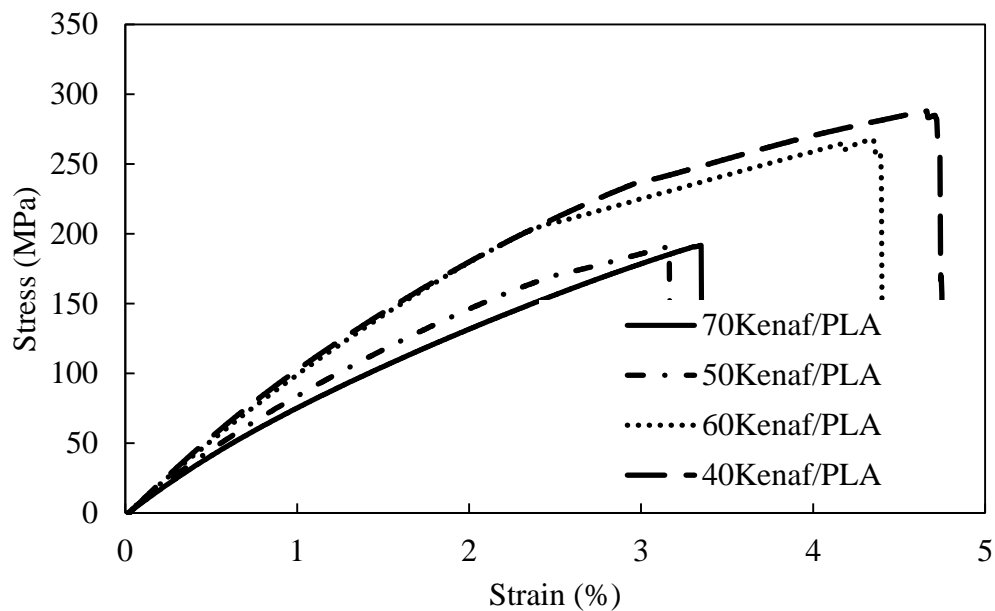


Figure 4.10 Typical stress-strain curves of kenaf fiber-reinforced PLA at 40, 50, 60 and 70 wt.% fiber content.

#### 4.4 Comparison properties of monolithic green composites

Kenaf/PLA shows the highest tensile strength and stiffness compared to coir/PLA and bamboo/PLA up to 70wt.%, presenting the compatibility of kenaf and PLA in the composites. Kenaf fiber was reported to produce a strong bond between PLA matrix (Masud S. Huda et al., 2008). As shown in Fig. 4.11, tensile strength increases gradually with the increasing fiber content for kenaf/PLA and bamboo/PLA. Meanwhile coir/PLA showed a minimal increase with fiber content up to 70wt.%. It may be depicted that the coir fiber content did not contribute to the composites' strength, but may affect strain properties. However, bamboo and kenaf fibers relied on fiber content for their individual composites.

Kenaf and bamboo fibers are likely affecting the strength properties of composites. In contrast, the increment of coir fiber reinforcement in composites would not affect the strength but decreased its strength at 25% coir fiber content (Tran et al., 2014).

Based on this assumption, the hybrid content of kenaf and bamboo fibers may produce high strength composites, depicted by the high properties of individual fiber bundles and single composites, while coir fiber aims to increase fracture energy and flexural toughness (Yan, Chouw, Huang, & Kasal, 2016).

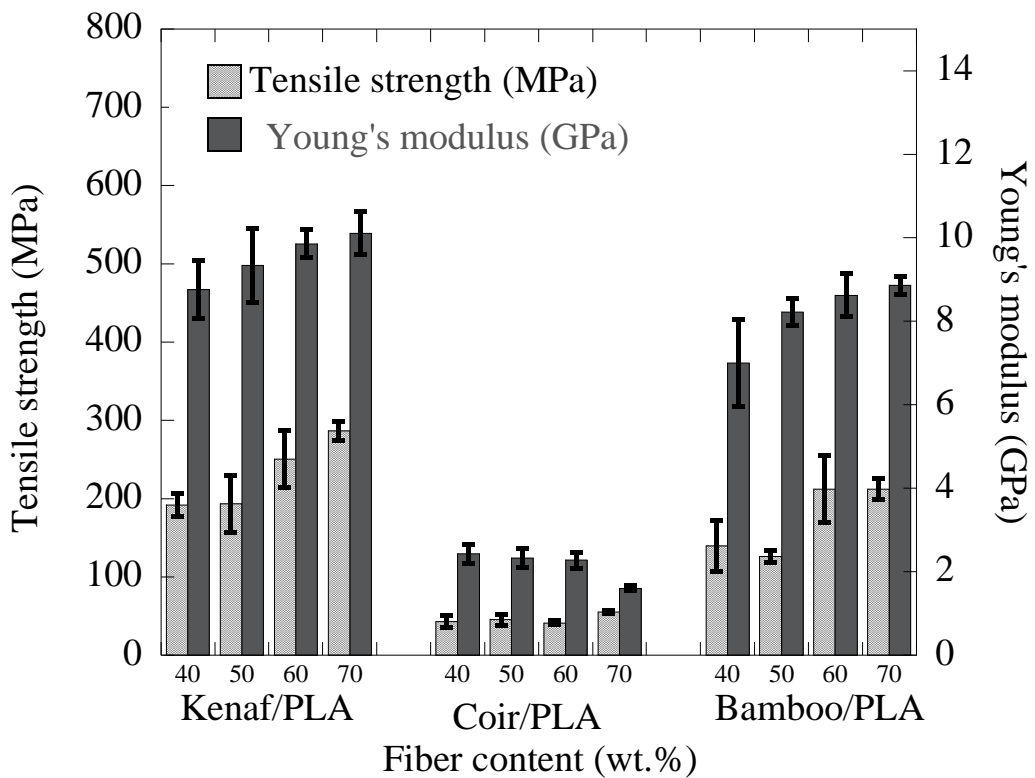


Figure 4.11 Comparison tensile properties of monolithic green composites reinforced PLA as a function of fiber content.



#### 4.5 Prediction of tensile properties by rule of mixtures (ROM)

As shown in Fig. 4.12, the increasing trend of composite strength was also estimated by ROM, equation (3.6). The volume fraction of fiber was determined by weight fraction using equation (4.1).

$$V_1 = \frac{W_1/\rho_1}{W_1/\rho_1 + W_2/\rho_2 + W_3/\rho_3 \dots} = \frac{W_1/\rho_1}{V_s} \quad (4.1)$$

where  $V_1$  is the volume fraction of one type of fiber in a composite,  $\rho$ , and  $W$  are the density and weight fraction of the components respectively. The subscripts of 1, 2, 3 and etc. are the total components in the composite or can be substituted by  $V_s$ , the sample volume ( $\text{cm}^3$ ). With the density values of 1.64, 1.47 and 1.3  $\text{g/cm}^3$  for kenaf, bamboo and coir fibers, respectively, and of elastic modulus of 22, 20, and 2.04 GPa for kenaf, bamboo, coir fibers, respectively, the ROM can be calculated. The elastic modulus of PLA is estimated to be 1.95 GPa.

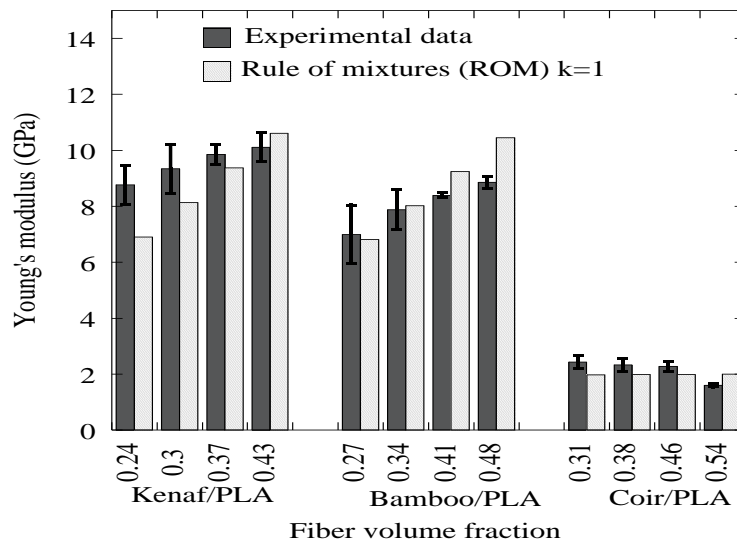


Figure 4.12 Comparison with experimental data of monolithic green composites and theoretical ROM.

The comparison between single green composites and theoretical values of ROM is shown in Fig. 4.12. At first, the fiber weight content (wt.%) was converted to volume fraction. Even though the initial fibers' weight content in each composite was equal (40, 50, 60 and 70 wt.%), after converting, there was a slight difference in each composite fraction which referred to their individual fibers' density, ranging from 0.24 to 0.54 volume fractions. The figures show that the composites depicted the increasing trend up to 70wt.% fiber content for both experimental and ROM data (Davallo, Pasdar, & Mohseni, 2010; Sergio Neves Monteiro et al., 2011). However, the experimental value and theoretical values were slightly different. The estimated values of both kenaf/PLA and coir/PLA were lower than experimental data in up to 60 wt.% fiber content. The values of ROM for bamboo/PLA were higher than that of experimental value with up to 70 wt.%. In contrast, at 50 wt.%, the experimental data agreed with the estimated value from ROM. The reason for the mismatch values was the effectiveness factor ( $k$ ) of the fiber reinforcement, which was difficult to assume, and the inaccurate values of densities also tend to be in disagreement with the theoretical rule of mixtures. Factors such as layering patent and the intermingling of fibers and various aspect ratios may influence the properties and strain compatibility of one fiber type leading to a decrease in the elastic properties (Zhang et al., 2013). The imprecise density may produce gaps between theoretical and experimental values.

#### **4.6 Surface morphology**

Scanning electron microscopy and optical microscopy were used to observe their microstructural failures.

#### 4.6.1 Coir fiber-reinforced PLA composites (Coir/PLA)

As can be seen in Fig. 4.13, coir fiber cell walls were fractured during the tensile strength test which led to the failure of the composites. The interphase between coir and PLA matrix was strong as no void was revealed in the images. However, PLA matrix was not evenly distributed around the fibers. The viscosity of PLA influenced the flow of matrix and matrix penetration among fibers.

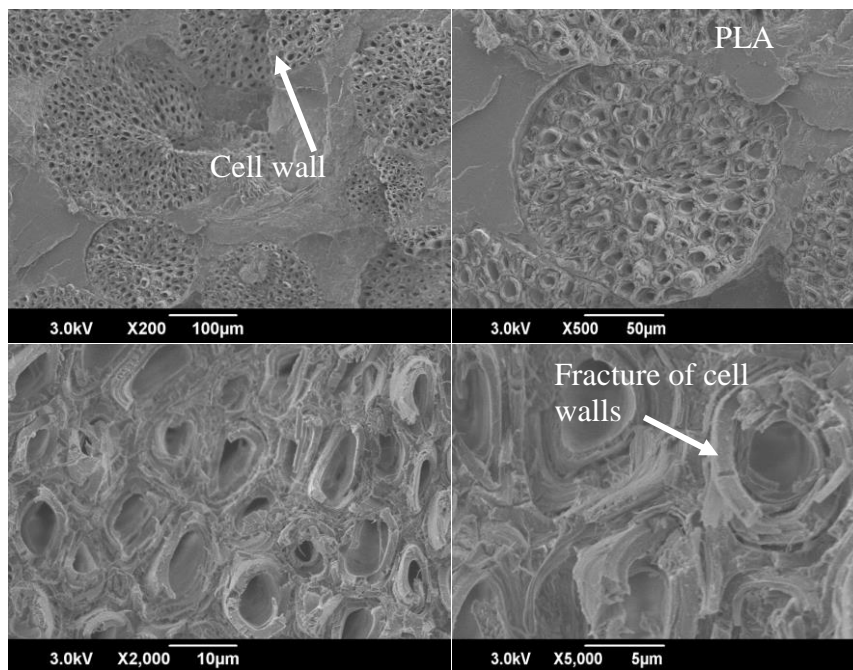


Figure 4.13 SEM images of coir fiber fracture in cell walls under tensile loads.

#### 4.6.2 Bamboo fiber-reinforced PLA composites (Bamboo/PLA)

As for bamboo fiber, a pulled out mechanism can be observed on composites' fracture surfaces. The weak interfacial bonding between bamboo fiber and matrix, yield the pull out of the fiber from the composites. A further wetting and cleaning of fibers, as well as chemical treatment through mercerization and silane modification, will help to

improve the interphase behavior of composites. The pulled-out bamboo fibers can be seen in Fig. 4.14.

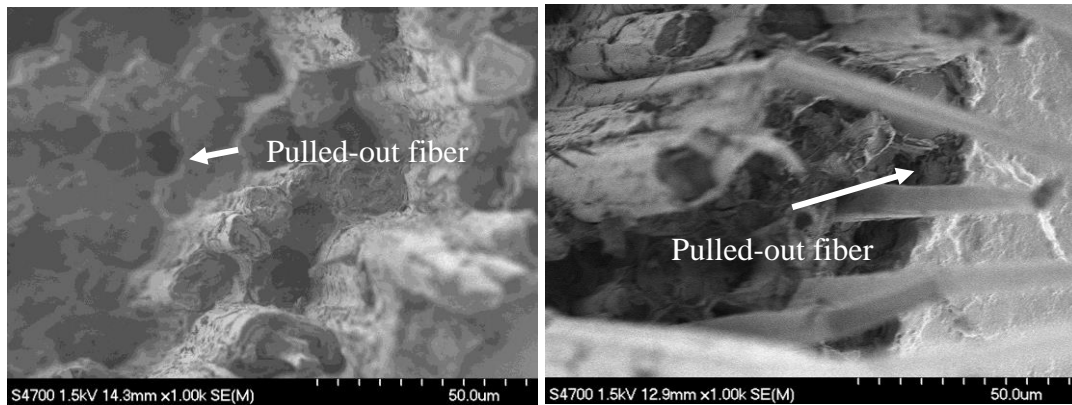


Figure 4.14 SEM images of bamboo fiber pull out under tensile load.

#### 4.6.3 Kenaf fiber-reinforced PLA composites (Kenaf/PLA)

Kenaf/PLA was found to show better interfacial behavior with PLA matrix. A strong bond can be seen in Fig. 4.15 in composites surfaces. The foreign particles (substance) surrounding the matrix were also noticed since kenaf fiber has many dissimilarities of fiber length taken from bast fibers.

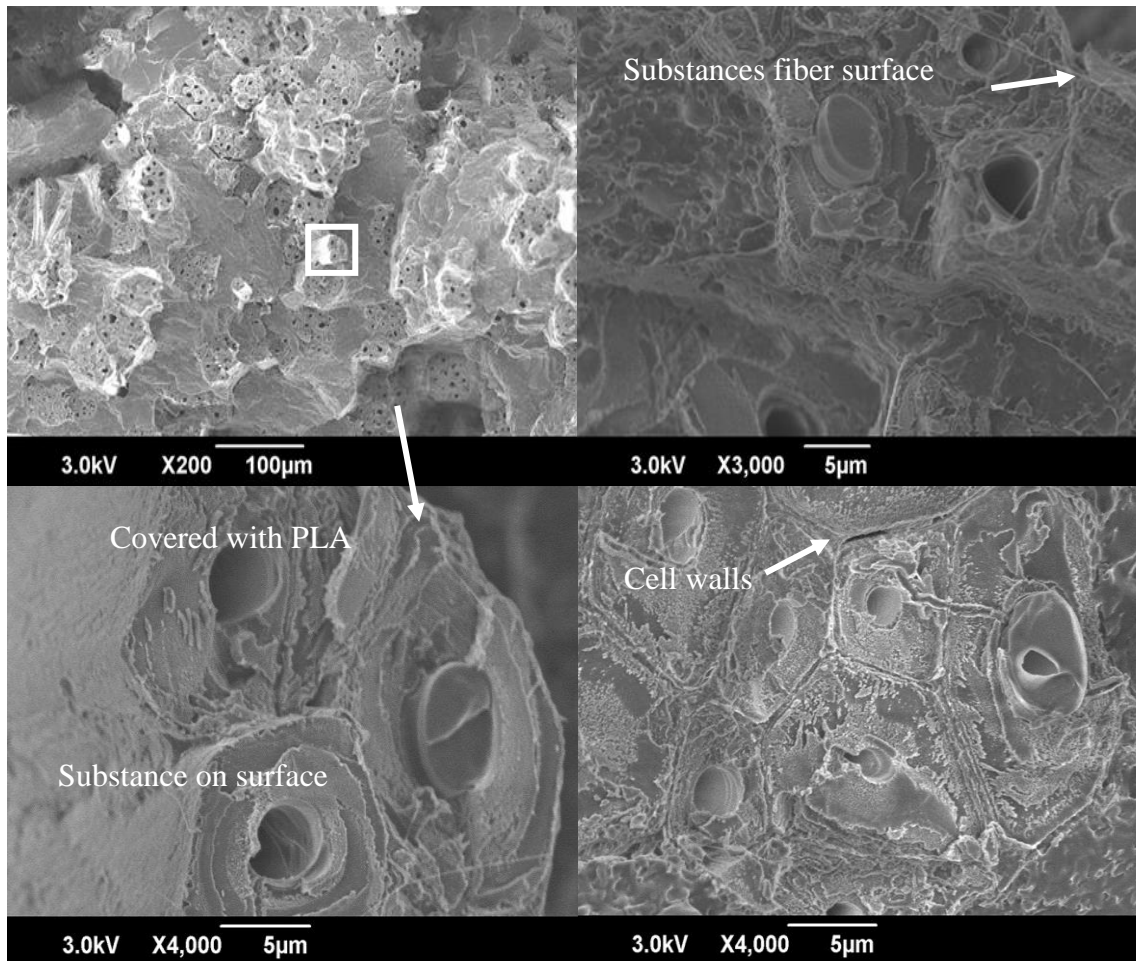


Figure 4.15 SEM images of kenaf fiber fracture in cell walls under tensile load.

# CHAPTER FIVE

## Result and discussion-Part II – Hybrid green composites characterization

### 5.1 Introduction

For the optimum combination of hybrid green composites, the prime concern is to comprise composites with both high strain and strength fibers as well as compatible with PLA matrix. To obtain the desired properties, the selection of plant fibers and their stacking sequences would determine the end properties of the composites. This chapter presents the results of:

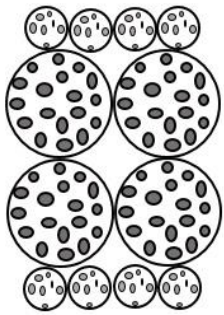
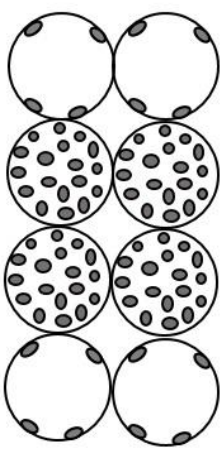
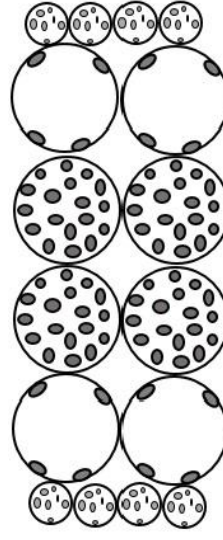

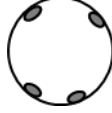

- Tensile and flexural properties of the three kinds of hybrid green composites
  - KCKK/PLA,
  - BCCB/PLA and
  - KBCCBK/PLA.
- Comparison of experiments with tensile strength theoretical hybrid rule of mixtures
- Surface morphology of hybrid green composites

### 5.2 Stacking sequence of hybrid green composites-high modulus fiber in the outer layer

The three different stacking designs were prepared to investigate the mechanical properties of hybrid green composites reinforced with two and three kinds of fibers. Basically, these stacking sequences were based on high modulus fiber in the outer layer

and low modulus and high strain fiber in the middle layer. The stacking patterns are referred to by the fibers' name (K=kenaf, B=bamboo and C=coir). The composite were: kenaf and coir fibers (KCCK/PLA), bamboo and coir (BCCB/PLA), and kenaf, bamboo and coir (KBCCBK/PLA). Each type of fiber-reinforced PLA composite had similar weight percentages (60:40) fiber to matrix content. Table 5.1 illustrates the stacking sequence of these composites.

Table 5.1 Model of hybrid green composites with different stacking configurations before hot-pressing.

Designations	KCCK/PLA	BCCB/PLA	KBCCBK/PLA
Number of layer	4	4	6
Stacking sequence			
Thickness (mm)	1.90±0.09	1.80±0.08	1.9±0.3
	 Kenaf fiber;	 Bamboo fiber;	 Coir fiber

Based on individual fiber bundle property (see section 4.3.1) and for the optimum hybrid properties in the composites, the high modulus kenaf and bamboo fibers should be placed both on top and bottom layers as suggested by Sreekala et al., (2002) and Ticoalu

et al., (2010) while the highly ductile and deformable coir fibers are positioned in the middle layer. Selecting the two higher Young's modulus kenaf and bamboo out of the three fibers on the top and bottom layers would produce greater flexural modulus since the outer layers receive a greater proportion of applied loads (Hull, 1981). Based on this assumption, kenaf and bamboo may produce high strength composites based on their high individual fiber strength, while coir aims to increase fracture energy and flexural toughness (Yan et al., 2016). The KCCK/PLA and BCCB/PLA composites are comprised of 4 layers, whereas KBCCBK/PLA composites consisted of 6 layers.

### **5.3 Mechanical properties of hybrid green composites reinforced PLA**

#### **5.3.1 Tensile properties of KCCK/PLA, BCCB/PLA and KBCCBK/PLA**

The hybridization of kenaf, coir and bamboo fibers in the composites show that the tensile and elastic modulus were improved when compared to neat PLA matrix. Figure 5.1 and Table 5.2 give the averages of their tensile properties. Comparing stacking sequences, the maximum tensile strength and Young's modulus were, when combining the three types of fibers in the form of KBCCBK/PLA composite, approximately 187 MPa and 7.5 GPa respectively. It was approximately 20 and 78% higher than BCCB/PLA and KCCK/PLA in tensile strength, while Young's modulus was approximately 2 and 25% higher than KCCK/PLA and BCCB/PLA respectively. The lowest tensile strength of 105 MPa was produced by kenaf/coir (KCCK/PLA), however this combination produced a high elastic modulus of 7.3 GPa similar to the elastic modulus of KBCCBK/PLA.



A similar trend can be seen in both KBCCBK/PLA and KCCK/PLA, while the stress-strain curve of BCCB/PLA shows a non-linear (internal fiber-matrix failure) behavior before starting to elongate further until reaching the composite's failure (Fig. 5.2). Untreated bamboo fiber was the reason for internal fiber-matrix failure mode. Since the chemical compound of its surface led to ineffective interfacial bonding with the matrix (Kang, Park, & Kim, 2014), treatment with alkaline is favourable to clean the fibers to improve strength of BCCB/PLA (Yan et al., 2016). It is believed that the multi fiber system (hybrid) compensating of low strength of coir, low elongation of kenaf fiber and high strength of bamboo produce high tensile properties of composites as evident in stress-strain curves of KBCCBK/PLA and BCCB/PLA.

Based on our observation, the impurities of bamboo fiber led to fiber-matrix interfacial failure and decreased the stiffness of the composites as reported in Huda et al. (2008). In addition, the small cross section of kenaf and the compatibility of kenaf and coir with matrix produced high stiffness due to the highly packed fibers and matrix.

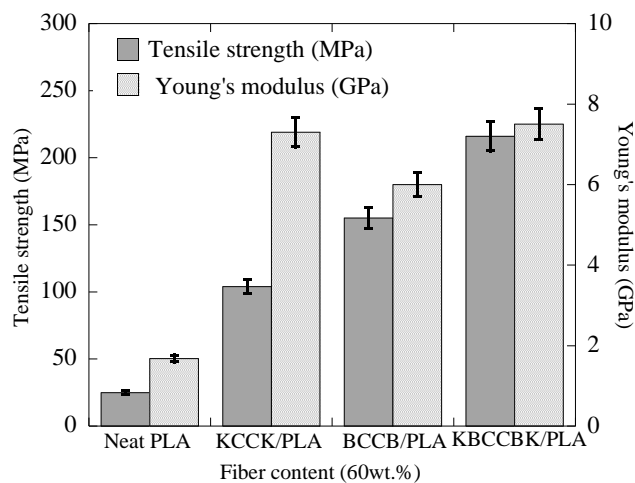


Figure 5.1 Tensile strength and Young's modulus of hybrid green composites.

Table 5.2 Tensile properties of PLA, KCCK/PLA, BCCB/PLA and KBCCBK/PLA.

Hybrid green composites	Tensile strength (MPa)	Young's modulus (GPa)	Strain at break (%)
Neat PLA	25±4	1.68±0.25	2.2±0.7
KCCK/PLA (60wt. %)	104±9	7.3±0.4	1.7±0.8
BCCB/PLA (60wt. %)	155±8	6.0±0.3	3.83±0.20
KBCCBK/PLA (60wt. %)	190±30	8±0.4	5.3±0.3

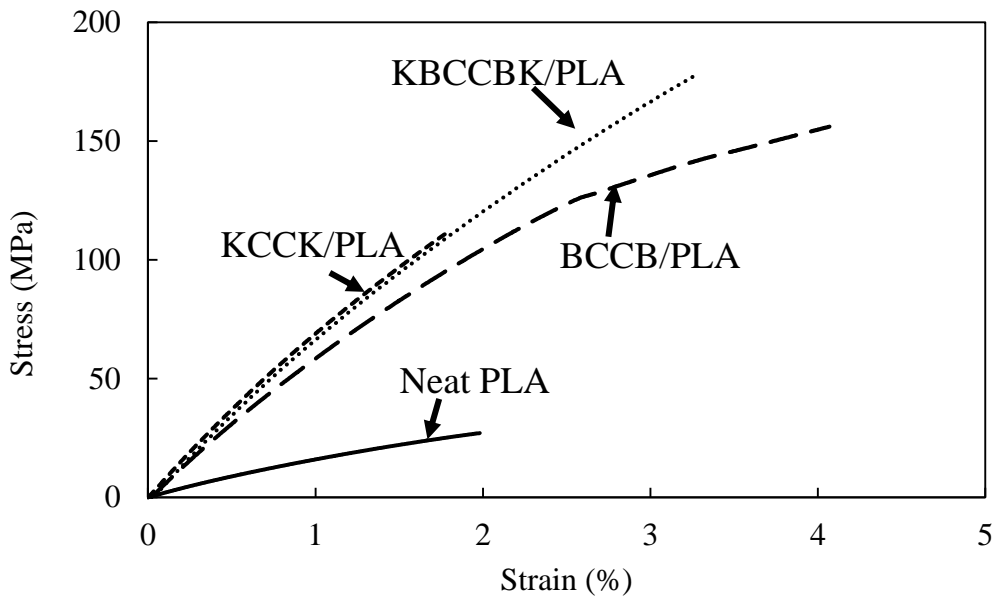


Figure 5.2 Typical stress-strain curves of hybrid green composites and neat PLA matrix.

### 5.3.2 Tensile toughness

The energy to break per unit volume (toughness) was estimated by the total area under the composites' tensile stress-strain curve (Gordon, 1978). As shown in Fig. 5.3, the toughness of hybrid green composites achieved  $0.97 \text{ MJ/m}^3$ ,  $3.54 \text{ MJ/m}^3$  and  $4.45 \text{ MJ/m}^3$  for KCCK/PLA, BCCB/PLA and KBCCBK/PLA, respectively. As a comparison, the toughness of the hybrid green composites and selected conventional material were examined. Despite the lower toughness than that of carbon fiber, the toughness of hybrid green composites is similar to that of high-tensile steel. As mentioned by Nam et al.,

(2011), coir fiber content should be limited to 25% to produce composites with high toughness. Since KBCCBK/PLA contained a similar fraction of fiber (20 wt.% ) in the composite, coir fiber helped produce high toughness. The structure of coir fibers, with high MFA (30-49°) contributes to higher toughness than those with small MFA in the fiber cell wall (Baley, 2002; Bledzki & Gassan, 1999). Contrary to that, higher content of coir fiber (more than 25%) in other combinations, KCCK/PLA and BCCB/PLA resulted in low toughness, though they displayed high elastic modulus that deliver acceptable performance for structural applications (N. Saba, Paridah, & Jawaid, 2015; Shah, 2013). Therefore, these kinds of hybrid stacking configurations produce optimum properties for unidirectional plant fiber-reinforced composites, through the integration of layer properties over repeating fiber/matrix patterns to attain effective elastic modulus, which is essential in various applications.

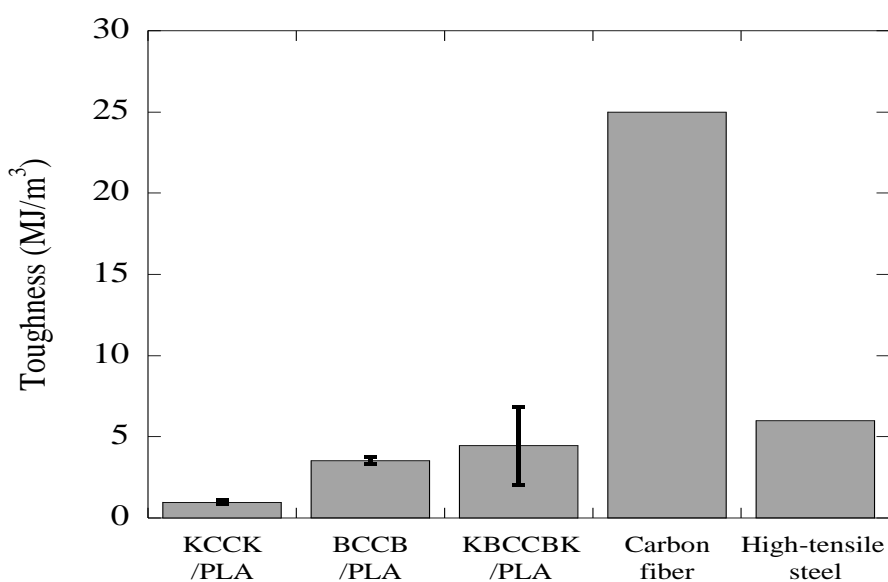


Figure 5.3 Toughness of hybrid green composites and selected materials.

### 5.3.3 Comparison between experimental and theoretical (HROM) tensile properties

The mechanical performance of composites can be predicted by a simple hybrid rule of mixtures (HROM) as mentioned in section 4.5. As for the types of composites, both tensile and elastic modulus were improved significantly compared to neat PLA matrix and agreed well with the hybrid rule of mixtures (Fig. 5.4 and Fig. 5.5).

As shown in Fig. 5.5, a small difference in tensile strength of KCCK/PLA and KBCCBK/PLA, indicated by experimental values that were 9 and 15% higher than the hybrid rule of mixtures, respectively. On the other hand, Young's modulus of KBCCBK/PLA and BCCB/PLA were 20 and 13% lower than estimated values of the hybrid rule of mixtures, respectively (Fig. 5.6).

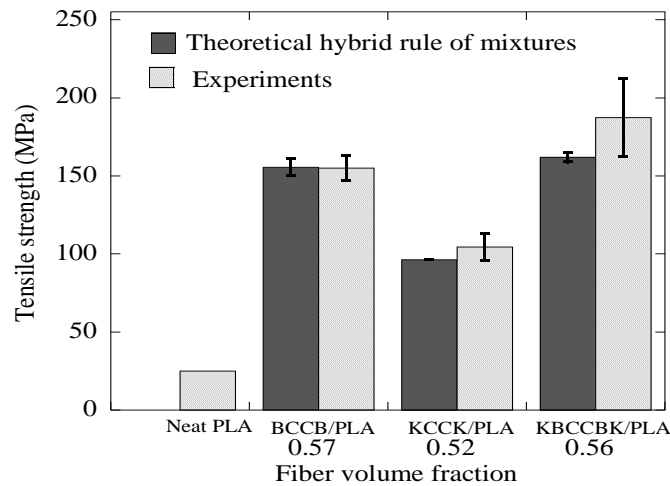


Figure 5.4 Tensile strength from experiment and theoretical hybrid rule of mixtures.

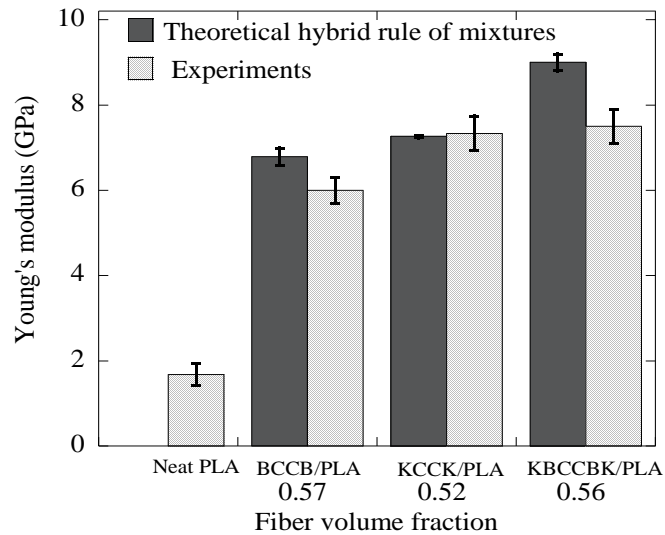


Figure 5.5 Young's modulus from experiment and theoretical hybrid rule of mixtures.

### 5.3.4 Flexural properties

Figure 5.6 shows the stacking configuration of kenaf-coir fibers (KCCK/PLA) exhibited the highest flexural modulus of 15 GPa, approximately 70% higher than other stacking combinations. At 60 wt.% fiber content, KCCK/PLA was comparable to the property of flax/PLA (40 wt.%) composites reported by Duhovic et al. (2009) and McGregor et al. (2010). The function of kenaf fibers as a rigid filler was assumed to enhance the stiffness of polymer matrix (Avella et al., 2008) and its strong interaction with PLA matrix (Masud S. Huda et al., 2008; Nishino et al., 2003). Flexural modulus of both KBCCBK/PLA and BCCB/PLA achieved approximately 9 GPa. In contrast, flexural strengths of KBCCBK/PLA and BCCB/PLA were higher than that of KCCK/PLA. The stronger kenaf and bamboo fibers in the outer layers might have influenced the superior mechanical properties of hybrid composites as reported in literature (Ticoalu et al., 2010).

Consequently, bamboo and kenaf fibers play a major role in increasing the strength of hybrid green composites, but coir fibers offset the low elongation of bamboo and kenaf fibers. Therefore, kenaf and bamboo fiber-reinforced composites can substitute for plywood or particle board in furniture applications.

On the other hand, flexural failures are governed by fracture of the cell walls themselves in fiber bundles (Kulkarni et al., 1981). The internal structure of the fiber cell walls provides stability to composites. Based on wood bonding formation, the wetting and flow of adhesive (polymer matrix) into the fiber pores ends up in the cell wall, and the reaction with the cell wall components, or polymerization in the cell, promotes an interphase region (Frihart, 2006). Lumen connected in the secondary layer of the cell wall, promoting cohesion by interlocking the fiber cell walls (Khalil et al., 2006) with the matrix, as existing in kenaf and coir fibers. A strong interaction between the fiber cell walls and matrix correlates to high flexural properties of composites (Almeida et al., 2013). Thus, the inclusion of bamboo and kenaf fibers compensates for the low strength of coir fibers by bearing the tensile load, while coir fiber is capable of high elongation that contributes to higher toughness of the hybrid green composites.

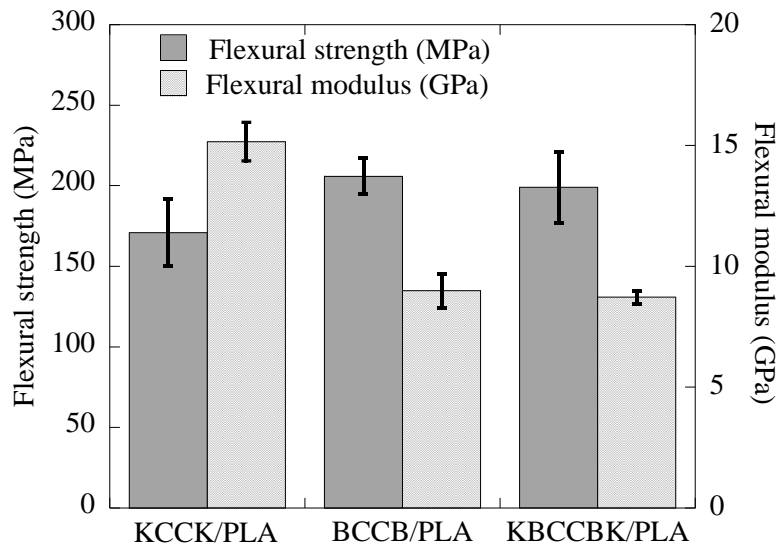


Figure 5.6 Flexural strength and flexural modulus of hybrid green composites.

### 5.3.5 Comparison properties of hybrid and various green composites

Single kenaf, bamboo, and coir-reinforced PLA composites have been discussed greatly and produced relatively low strength compared to hybrid green composites (Pickering et al., 2016; Sathishkumar et al., 2014; Yusoff et al., 2016). The fabrication method tends to reduce their strength properties, such as fibers' orientation, length of fiber and size of specimens. Comparing to other single fiber type biodegradable composite materials, such as flax/Bioceta, flax/Sconacell A, flax/Mater-Bi, the hybrid green composites are far better in tensile strength and in elastic modulus, except for flax/Mater-Bi which is slightly higher in elastic modulus (Bledzki & Gassan, 1999). In addition, these properties are relatively good to make the green composites applicable as interior construction material where exposure to environment is limited (Charlet et al., 2007).

Comparing with plant and glass fiber-reinforced polymer composites (GFRP), hybrid green composites show comparable tensile strength to flax/GFRP/phenolic resin

(Zhang et al., 2013). On the other hand, hybrid composites of jute/sisal/epoxy (Sudhir et al., 2014), sisal/GFRP/epoxy and jute/GFRP/epoxy (Ramesh et al., 2013) were far lower in tensile strength than hybrid green composites (Table 5.3). As well as tensile strength, the flexural strength and flexural modulus of hybrid green composites showed far higher values compared to those of jute/sisal/epoxy, and single fiber/PLA composites

Table 5.3 Strengths of selected hybrid green composites.

Hybrid green composites/ plant fibers/synthetic	Tensile strength (MPa)	Young's modulus (GPa)	Strain at break (%)	Flexural strength (MPa)	Flexural modulus (GPa)	Ref.
Flax/glass fiber/phenolic	393-450	40	1.0	-	-	(Zhang et al., 2013)
Jute/sisal/epoxy	40	1.6	5	88.33	3.5	(Sudhir et al., 2014)
Sisal/GFRP	69	-	-	-	-	(Ramesh et al., 2013)
Jute/GFRP	63	-	-	-	-	(Ramesh et al., 2013)

In order to pursue the biodegradable, green composites, matrix resin of polylactic acid (PLA) is frequently used to reinforce plant fibers. PLA can be reconverted and naturally decomposed to satisfy the environmental impact. Compared to polypropylene (PP), PLA not only exhibits higher modulus but also higher storage modulus and flexural properties (Han, Karevan, Sim, et al., 2012) as well as high mechanical and thermal properties (Masud S. Huda et al., 2008; Nishino et al., 2003) which is comparable to polystyrene (Suryanegara et al., 2009). Green composite of single fiber type-reinforced PLA have been reported about extensively, though with much variability in strength properties (Akil et al., 2011; Y. Dong et al., 2014; Han, Karevan, Sim, et al., 2012; Han, Karevan, Bhuiyan, et al., 2012; Ochi, 2008; N. Saba et al., 2015; Sukmawan, Takagi, &



Nakagaito, 2016; Takagi & Ichihara, 2004). The variability of properties may be due to the size of testing samples, fiber orientation, fiber content, fiber treatment, fiber length and fabrication method.

#### **5.4 Surface morphology of hybrid green composites**

Optical microscopy and scanning electron microscopy were used to observe the interfaces between PLA matrix and fibers as well as to clarify the failure mechanism of hybrid green composites. Among the three fibers, kenaf is hardly discernible in the matrix PLA, showing better adhesion than coir and bamboo fibers.

##### **5.4.1 BCCB/PLA**

Figure 5.7 shows the optical images of fracture surfaces of fiber according to their stacking sequences. It shows a poor bonding between fibers (coir and bamboo) and matrix PLA due to insufficient matrix in the composite sequence. The rich chemical compound (impurities) of untreated bamboo fibers lead to low matrix penetration between the fibers, high porosity and weak interlocking, which brought about the initial failure of composites as evident in tensile stress-strain curve of BCCB (Fig. 5.2). As can be seen in the SEM images (Fig.5.8), bamboo cell walls have impurities on their surfaces which are incompatible with PLA, causing fiber pull out and splitting during tensile loads. The function of the matrix to bind the fibers and transmit the stress to the fibers had failed due to loss of the matrix that was squeezed out during hot pressing.



Figure 5.7 Optical microscopy images of fracture surfaces of untreated bamboo and coir fibers embedded in PLA in BCCB/PLA.

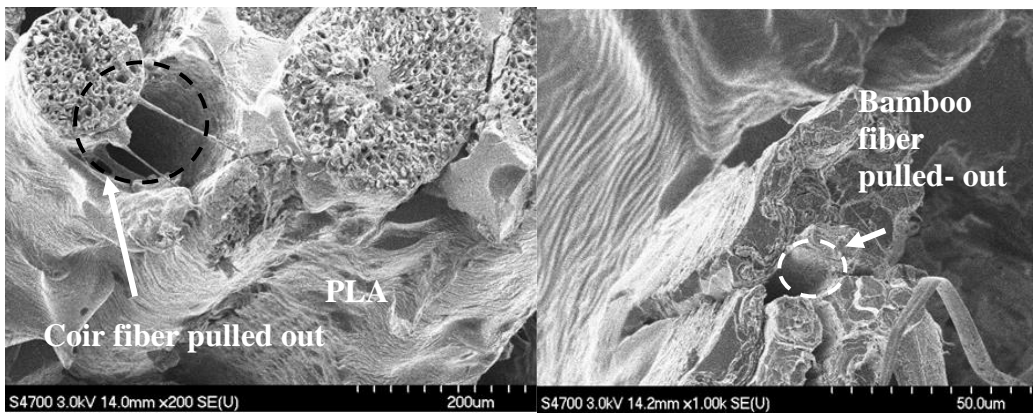


Figure 5.8 SEM images of BCCB/PLA composites showing pull out of (a) coir fibers and (b) bamboo fibers.

#### 5.4.2 KCCK/PLA

In contrast, Figure 5.9 and 5.10 show a highly packed KCCK composite which was influenced by a small cross section of kenaf fibers and fewer impurities, allowing the matrix to penetrate better. Compared to BCCB/PLA, this effect produced a higher elastic modulus of KCCK/PLA as evident in tensile properties than the other two hybrid green composites. The hollow structures (lumen) of coir fibers may decrease the volume of the composite which attributes to the low tensile strength. It is noticeable that the dissimilar

dimension (cross sectional) of coir fibers in the composites decreases the compaction in the composites, producing porosity and affecting the low strength of the composites.

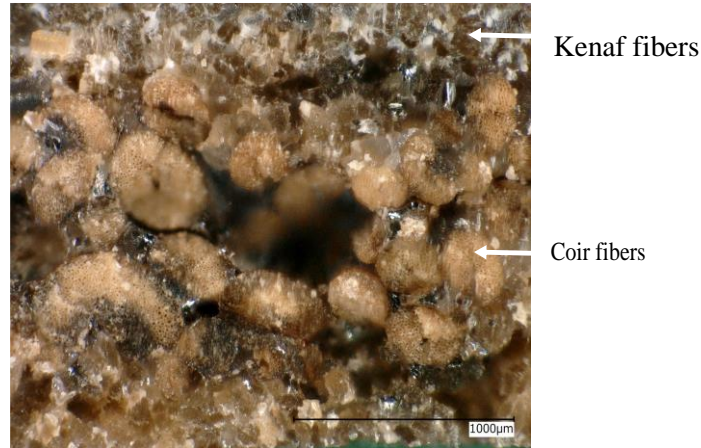


Figure 5.9 Optical microscopy images of KCCK/PLA composites showing fracture of coir and kenaf fibers

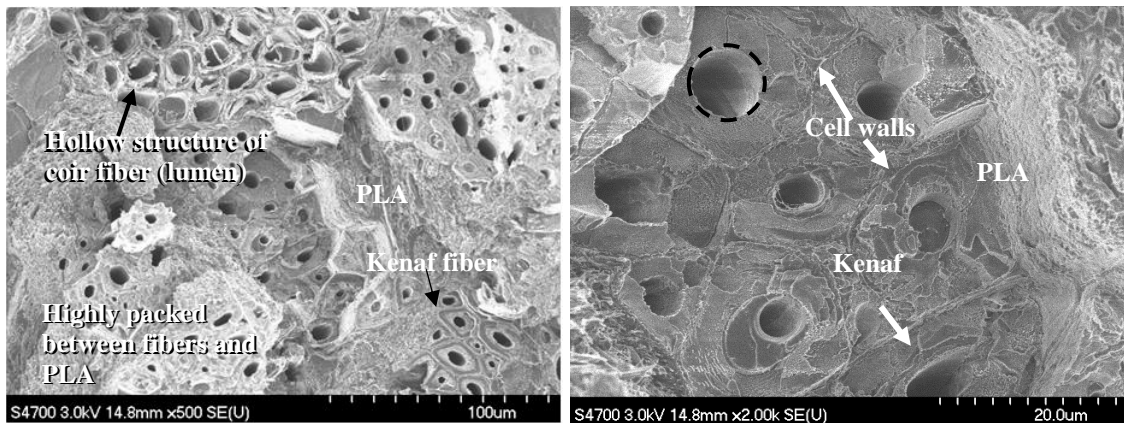


Figure 5.10 SEM images of KCCK/PLA composites showing (a) fracture surface of coir and kenaf fibers and (b) cell wall of kenaf fiber and PLA matrix.

### 5.4.3 KBCCBK/PLA

The existence of untreated bamboo fibers in composites, KBCCBK stacking sequence illustrates a similar failure mechanism with BCCB/PLA composites, which is poor interfacial between fibers and matrix, where pull out of bamboo is noticeable, decreasing its strength (Fig. 5.11).



Figure 5.11 Optical microscopy images of fracture surfaces of untreated kenaf, bamboo and coir fibers embedded in PLA in KBCCBK/PLA.

Penetration of the PLA matrix into the lumens may occur via the porous structure of plant cell walls, consequently creating cohesion in cell walls to resist loads (Frihart, 2006). However, poor wetting of coir and bamboo fibers in PLA matrix resulted in de-cohesion of fiber cell walls, consequently leading to fiber pull out in BCCB/PLA and KBCCBK/PLA as can be seen in Figs. 5.8 and 5.12. However, kenaf fibers were well adhered and dispersed in the PLA matrix, thus delivering high strength to the composites (Figs. 5.12 (e-f)). Likewise with wood surfaces, this microscopic wetting of fibers and flow of the matrix is one step of a bond formation in the composites' interface (Frihart, 2006). In addition, kenaf fibers stimulate the nucleation of PLA to have higher impact strength and toughness and firmly adhere to the matrix (Avella et al., 2008; Han, Karevan, Bhuiyan, et al., 2012) since the fibers act as a reinforcement phase, preventing stress concentration in critical areas.

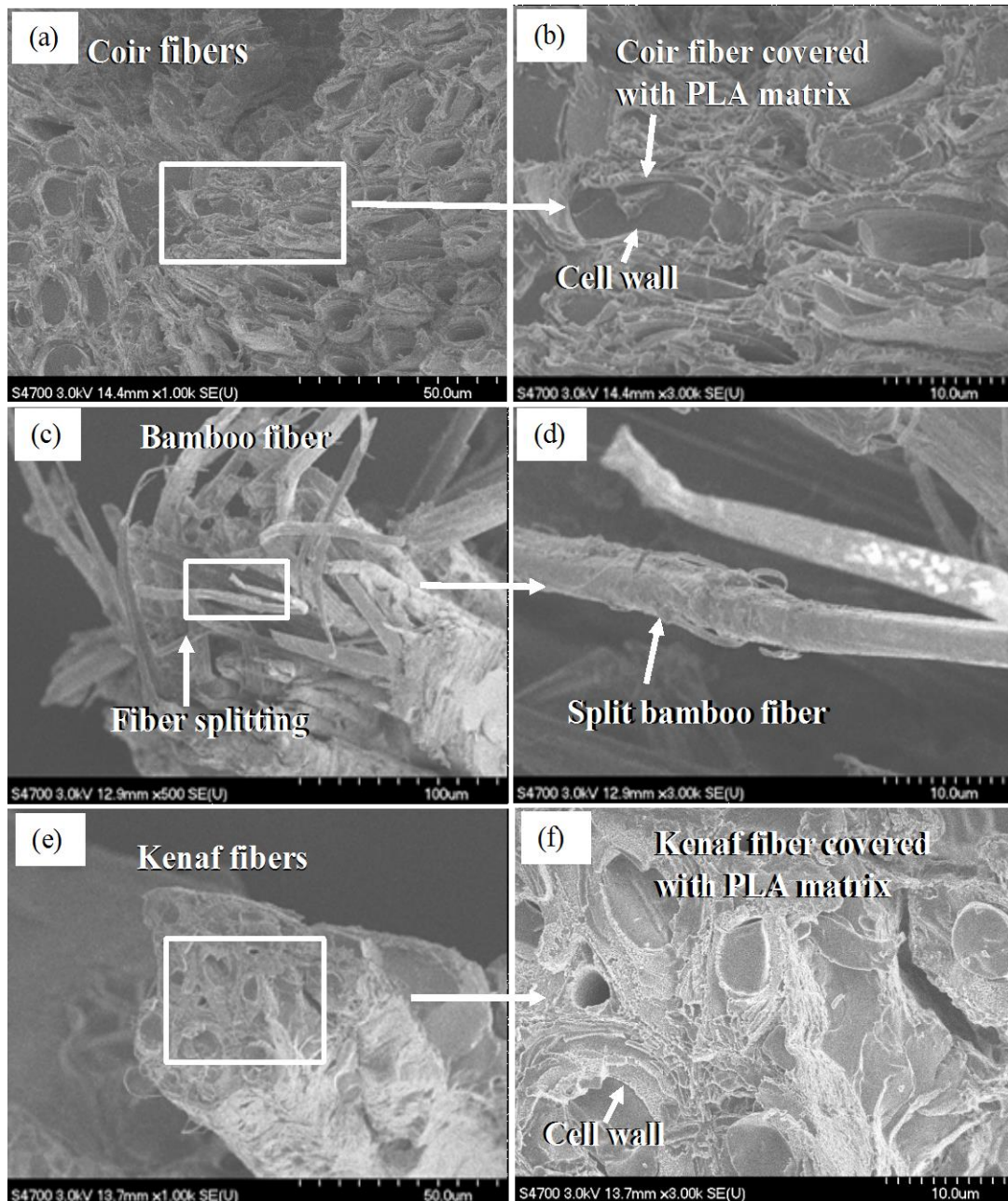


Figure 5.12 SEM images of (a)-(b) coir fibers, (c)-(d) bamboo fibers and (e)-(f) kenaf fibers in KBCCBK/PLA composites.

In conclusion, these stacking patterns provided well adhered interfaces between the layers of the composites, evidenced by smooth stress-strain curves (Fig. 5.2), except for BCCB/PLA where a premature failure occurred due to interface failure. The interface splitting crack of fiber bundles in composites was non-existent until reaching fiber failure

based on three dominating damage characteristics which are interface splitting crack, matrix shear crack and fiber failures (Yan, Chouw, & Jayaraman, 2014). Therefore, the design configurations and reinforced fiber selection are highly compatible with PLA matrix, producing composites with high mechanical properties. Generally, the greater interfacial bonding allows stress to be transferred between the matrix and the reinforcing fibers, to yield higher strength and stiffness (Piggott, 1987). Through effective wetting of the fibers and the matrix, a strong interfacial adhesion may be generated. Otherwise poor interfacial interaction leads to internal strain, porosity, and environmental degradation (Bledzki et al., 1996). Hence, an appropriate selection of fiber and polymer matrix is vital to achieve optimum compatibility properties of fiber and matrix. Further physical and chemical treatments should be carried out for better adhesion between hybrid constituents.

# CHAPTER SIX

## Result and discussion-Part III – Stacking sequences of hybrid green composites characterization

### 6.1 Introduction

Fabrication method and various fiber contents show various strength properties of the composites. Fibers' alignments as well as stacking sequences influence the composites' final properties. Typically, the high modulus fibers may produce high properties of the composites. Higher fiber content reinforced in composites also produces higher flexural and tensile strength, while too high fiber loading decreases its flexural strength. In addition, the viscoelasticity of plant fiber has a higher damping capacity compared to conventional fibers. This chapter presents the results of:

- Tensile, flexural and impact properties of two sequences composites:
  - CBK/PLA- low modulus fibers in the outer layers
  - KBC/PLA-high modulus fibers in the outer layers
- Comparison of experiments with tensile strength theoretical hybrid rule of mixtures
- Surface morphology of the hybrid green composites
- Loss factor of KBCCBK/PLA

### 6.2 Mechanical properties of coir, bamboo and kenaf hybrid green composites reinforced PLA

Two kinds of symmetrical stacking designs were prepared. One type of composite was reinforced with high modulus fibers in the outer layers (kenaf-bamboo-coir/PLA)

and the other with low modulus and high strain fibers in the middle layer (coir-bamboo-kenaf/PLA). Placing a high modulus in the outer layers (such as kenaf and bamboo fibers) and a high strain fiber (such as coir) in the middle layer, and vice versa was estimated to produce a unique property. These stacking designs of fiber layers were prepared to compare the effectiveness of low modulus and high strain fibers in the outer layer (coir-bamboo-kenaf/PLA) with the high modulus in the outer layers (Fig.6.1). Tensile, flexural and impact tests were conducted to investigate their mechanical properties with total fiber content varying from 50 to 70 wt.%.

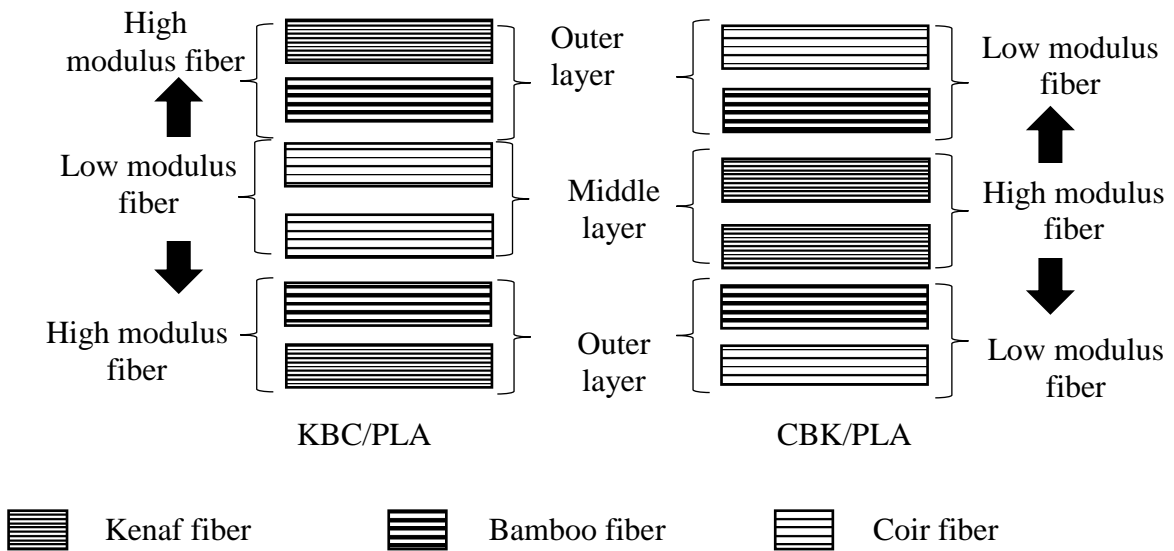


Figure 6.1 Stacking sequences of hybrid green composites of KBC/PLA and CBK/PLA.

### 6.2.1 Kenaf-bamboo-coir/PLA-high modulus fiber in the outer layer (KBC/PLA)

Since high modulus fiber in the outer layer produced high strength in the previous section (Chapter Five), the high modulus fibers of kenaf-bamboo-coir/PLA in the outer layers were evaluated. The stacking composite of KBCCBK/PLA was similar with KBC/PLA, and was reported in the section 5.2. However, some methods were changed



in this experiment, such as the drying condition, and a long stem and white bast kenaf fibers were used to reinforce the composites. Also the various contents were discussed in this section, ranging from 50-70 wt.%.

### **6.2.1.1 Tensile properties**

The maximum tensile strength and modulus of KBC/PLA were 158 MPa and 6.7 GPa, respectively. This stacking sequence showed a small standard deviation of the specimen, representing the uniformity of fiber dispersion and a few voids all over the composites (Fig. 6.2). The minimal increase of tensile properties was approximately 8% in up to 70 wt.% fiber content. The inherent properties of the high modulus fiber in the outer layer (bamboo and kenaf fibers) as reinforcement were likely to maintain its strengths with fiber content. Bamboo fiber was estimated to bear the tensile loads. While kenaf fiber was effective to increase adhesion at interfaces between matrix and fibers (Huda, Drzal, Misra, & Mohanty, 2006). As well as tensile strength, a small increase in Young's modulus was due to high modulus fibers in the outer layers which bamboo is capable of resisting axial and tensile loads (Fei et al., 2014). With up to 70wt/% fiber content, only 3% of elastic modulus was increased in this stacking sequence. This may be the result of the high strength of outer layer fibers' interaction to resist the tensile load, thus maintaining its strength throughout the fiber content. It was estimated that the high modulus of bamboo fibers induced strength more than coir fibers in the middle layer of this stacking sequence. Therefore, an increase in fiber content may maintain the hybrid properties due to the presence of high modulus fibers in the composites, like bamboo and kenaf fibers.

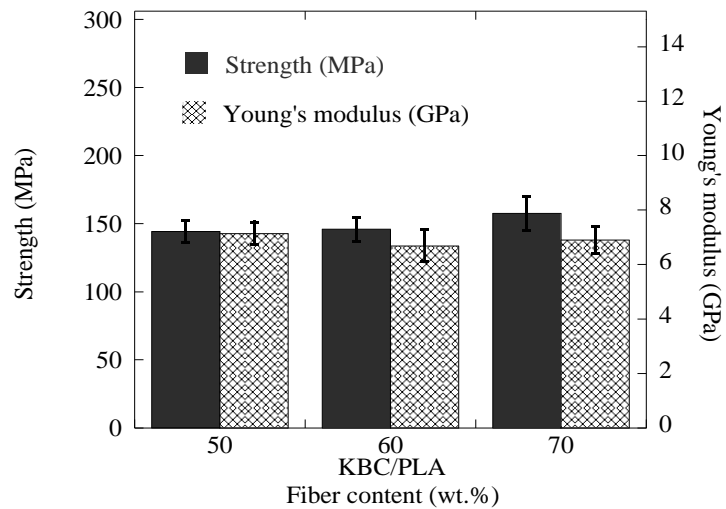


Figure 6.2 Tensile properties of KBC/PLA with fiber content.

### 6.2.1.2 Flexural properties

As predicted, Fig. 6.3 shows the stacking sequence of KBC/PLA achieved a higher flexural strength of 209 MPa at 60 wt.% fiber content. And it decreased slightly to 198 MPa at 70 wt.% fiber content. Typically, at higher fiber content in a composite, the matrix is insufficient which creates the porosity in the composites (Pickering & Aruan Efendy, 2016). Thus, the flexural properties in the composites are reduced. The similar trend of flexural modulus can be seen at 60 wt.% fiber content and achieved the highest, 15 GPa, and slightly decreased to 13 GPa at 70 wt.% fiber content. A large standard deviation at 70 wt.% fiber content represents the weak bonding between the fibers' network due to higher fibers content, generating high porosity in the composites as well. The inclusion of air and water may exist in the composite during fabrication, providing various strengths of the composites (Bourmaud, Duigou, Gourier, & Baley, 2016).

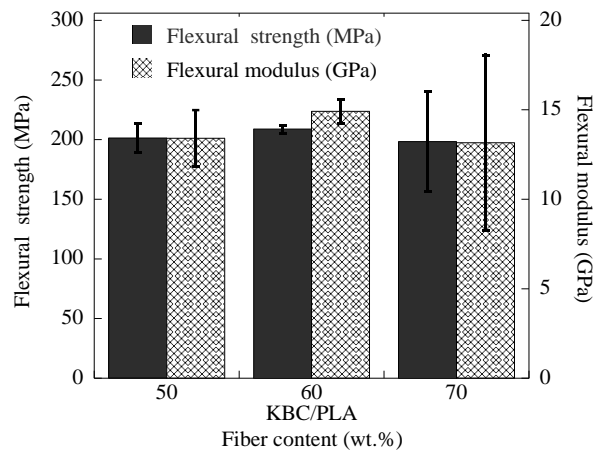


Figure 6.3 Flexural properties of KBC/PLA with fiber content.

### 6.2.1.3 Impact properties

Figure 6.4 illustrates the impact strength of hybrid composites which were considerably higher than that of neat PLA (2 kJ/m<sup>2</sup>), approximately one-twelfth. As well as tensile properties, the impact strength was increased with fiber content, from 28 to 51 kJ/m<sup>2</sup>.

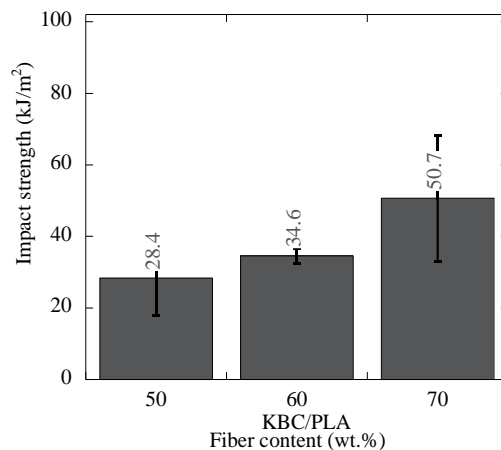


Figure 6.4 Impact strength of KBC/PLA with fiber content.

## 6.2.2 Coir-bamboo-kenaf/PLA-low modulus fiber in the outer layer (CBK/PLA)

The stacking effect of hybrid green composites is difficult to examine. Even though the fibers should have the high modulus and high strain for the optimum hybrid combination (Sreekala et al., 2002), the layer arrangement is one factor that may improve the composite properties. By replacing the sequences of the composite layers, it would be possible to understand the effectiveness of each layer in the composites.

### 6.2.2.1 Tensile properties

There was a minimal increase in tensile strength and modulus of CBK/PLA with up to 70 wt.%. The maximum tensile strength of 158 MPa was achieved and Young's modulus of 6.5 GPa was obtained in this stacking sequence (Fig. 6.5).

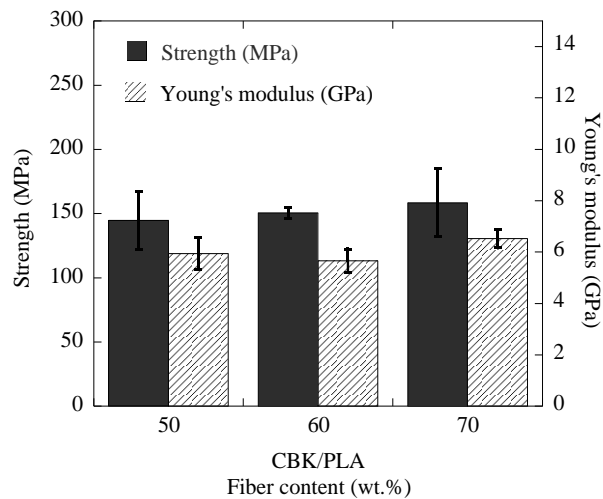


Figure 6.5 Tensile properties of CBK/PLA with fiber content.

### 6.2.1.2 Flexural properties

As seen in Fig. 6.6, the flexural strength of low modulus fiber in the outer layer (CBK/PLA) had increased up to 60 wt.%, from 100 MPa and 148 MPa. However, at high content of 70 wt.%, the flexural strength had reduced. The weak interaction between the high content of fibers may be the reason for the reduction in strength. In addition, the agglomeration of fiber occurred when the content of fibers in the composites affected the flexural properties. At 60wt. %, the maximum flexural modulus was achieved, approximately 7.2 GPa, while slightly decreasing to 6.7 GPa at 70 wt.%.

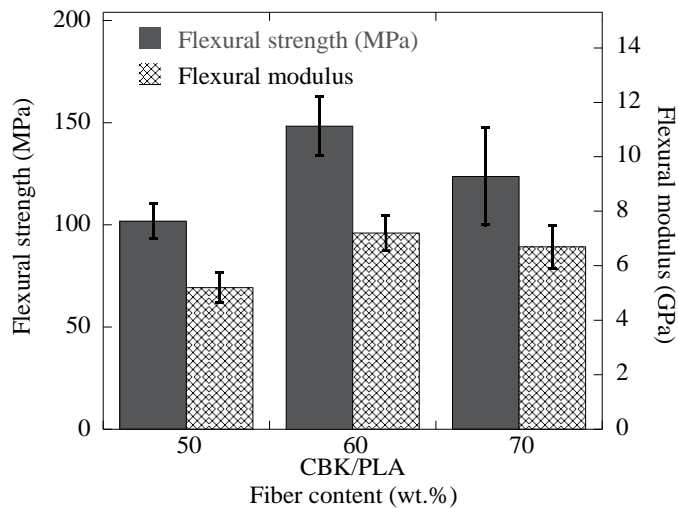


Figure 6.6 Flexural properties of CBK/PLA with fiber content.

### 6.2.2.3 Impact properties

The impact strength was improved with fiber content, ranging from 28 to 61 kJ/m<sup>2</sup>. As mentioned before, the agglomeration of fiber in the composites may be the reason for large standard deviation. Coir fiber has considerable fiber dimension and deformability which affects the fibers' orientation. Apart from that, a low interface between the layers

tends to decrease the impact strength. According to this stacking sequence, at up to 70 wt.% the impact strength of this stacking hybrid green composite is still applicable in structural applications.

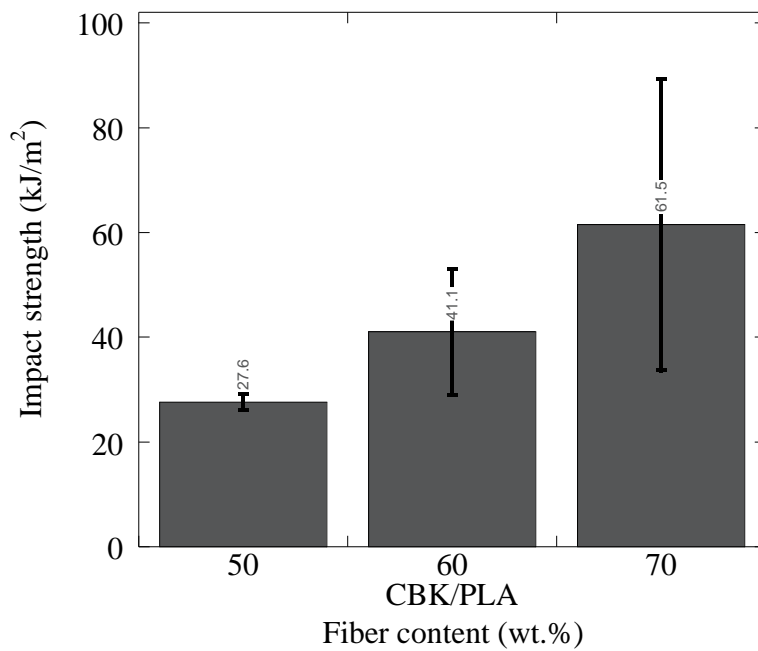


Figure 6.7 Impact strength of CBK/PLA with fiber content.

### **6.3 Comparison of experimental and theoretical tensile strengths of KBC/PLA and CBK/PLA**

#### **6.3.1 Tensile properties**

Both tensile modulus and tensile strength for the two stacking sequences increase minimally up to 70 wt.% fiber content (Figs. 6.8 and 6.9). Tensile modulus of KBC/PLA at 50 wt.% was higher than CBK/PLA, approximately up to 20% (7.14 GPa) higher than CBK composites (6 GPa), influenced by high modulus in the outer layer. However, the tensile modulus of both stacking sequences were varying, decreasing at 70 wt.% fiber content. This may be related to poor bonding between the matrix and fiber since impurities of the untreated fiber surface reduced the stiffness of the composites (Masud S. Huda et al., 2008), particularly in the case of bamboo fibers. For further study, the surface treatment was recommended to clean inorganic substances from the fibers as they tend to decrease the quality and durability of green composites (Lertwattanakruk & Suntijitto, 2015).

The hybrid rule of mixtures estimated the increasing trend in tensile modulus with fiber content (Mansor et al., 2013). The experimental values were lower than estimated. From the observation of composite surface, a poor interfacial bonding between fiber and matrix was believed to reduce strength properties.

Compared to neat PLA, both strength properties of hybrid composite improved significantly, approximately up to 530% in tensile strength and up to 260% in tensile modulus. It is observed that the stacking sequence was not significantly influenced by the tensile strength and there was a small effect in the tensile modulus if compared to similar content fiber reinforcement. According to Fig. 6.10, the stacking sequence CBK/PLA

with coir fiber in the outer layer (low modulus and high strain) had higher strain properties than KBC/PLA, which contained a high modulus of kenaf and bamboo in the outer layers. This may be attributed to the low modulus of coir fiber bearing the increasing loads preventing pre-failure of the composites.

Referring to a previous study (Yusoff et al., 2016), kenaf fibers fracture first before transferring loads to fiber networks. When one fiber fractures, the composite's performance may be affected, therefore, a non-linearity was seen in the hybrid composites. Apart from that, some misalignment of fibers during preparation led to rotational locking, preventing MFA realignment of the cellulose, and producing high stiffness of composites (Bourmaud et al., 2016). Consequently, bamboo and kenaf fibers play a major role to increase the strength of hybrid green composites, but coir fibers offset the low elongation of bamboo and kenaf fibers. It was found that 50-70 wt.% fiber content, and the equal ratio of these fiber types is the optimum fiber content in this hybrid combination since the small difference in each total fiber content affects the tensile properties. The strength properties are likely to drop if fiber content above 70% is used (Campbell, 2010).



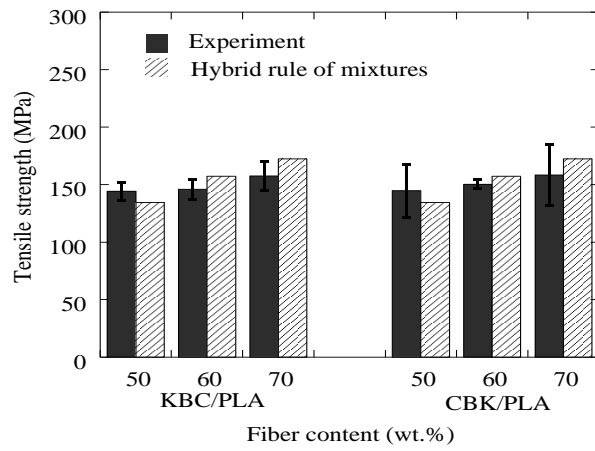


Figure 6.8 Tensile strength from experiment and calculation based on hybrid rule of mixtures.

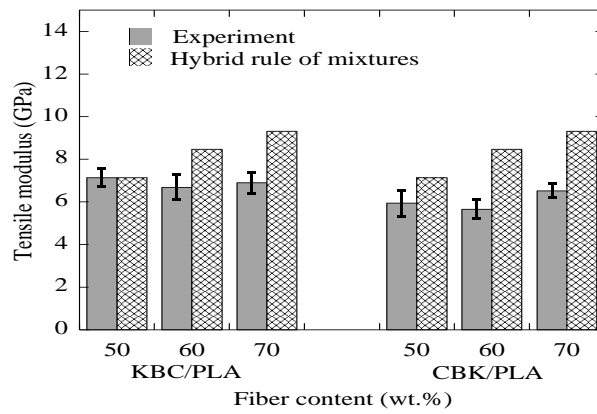


Figure 6.9 Tensile modulus from experiment and calculation based on hybrid rule of mixtures.

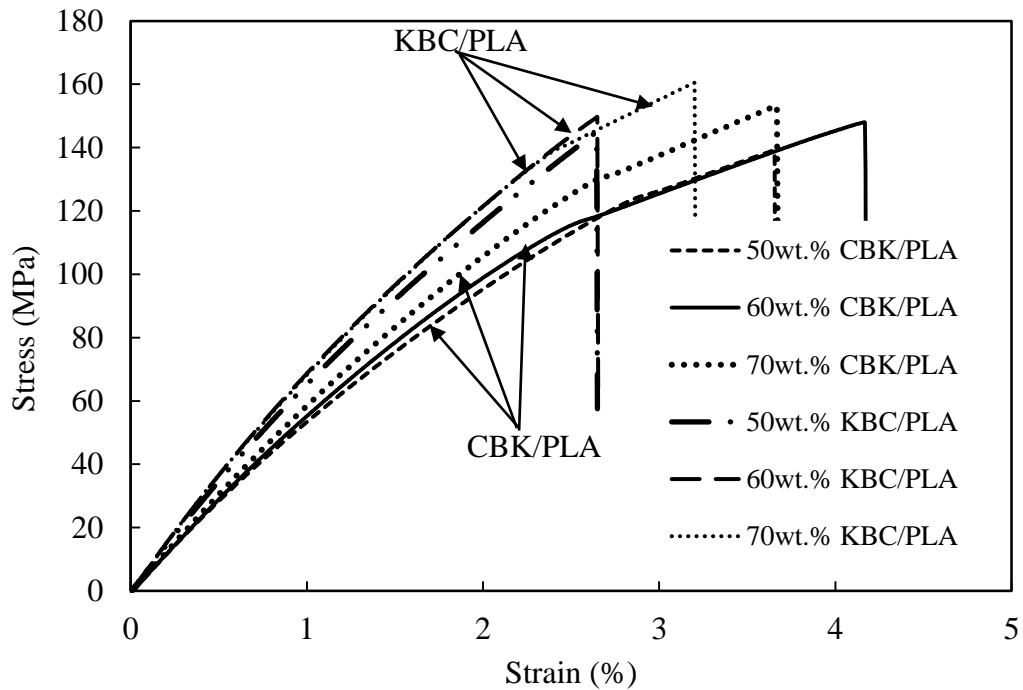


Figure 6.10 Typical stress-strain curves of hybrid green composites.

### 6.3.2 Flexural properties

The stacking sequence of KBC/PLA resulted in higher flexural strength and flexural modulus than CBK/PLA (Fig. 6.11). Neat PLA had relatively low flexural strength and modulus, measuring 2.5 MPa and 0.6 GPa, respectively. Similar to tensile properties, at 60 wt. % fiber content, both composites achieved the highest flexural strength and modulus. However, the different values from 50 to 70 wt.% fiber content were minimal particularly for KBC/PLA. CBK/PLA was assumed to have lower properties due to the low modulus of coir fibers in the outer layer's failure to withstand bending loads. Apart from that, a lower dispersion of coir fiber than kenaf fiber in the outer layers affected load transferred from matrix to fibers. In addition, richer lumens and thinner cell walls of coir fiber reduce its fiber volume (Yusoff et al., 2016), accounting for 22 to 30% fiber porosity (Tran et al., 2014). Also, the greater cross sectional area of

coir fibers was the reason for low fiber dispersion, prevented from perfect compaction during hot pressing compared to the smaller one, consequently yielding to low strength and stiffness (S.N. Monteiro et al., 2011). Apart from that, the presence of impurities in the bamboo fiber in the second outer layer of CBK/PLA promoted poor bonding between fibers and matrix. The foreign compounds (chemical composition) could yield to a low degree of low compaction (Graupner & Müssig, 2011). Air inclusion could occur and be regularly present in the fibers and matrix (Graupner & Müssig, 2011; Mitra, 2014). As evident in the fracture images (Fig. 6.15) and Fig.6.16), surfaces around the bamboo fibers had huge porosity in the composites. Higher fiber content increases air inclusion (Graupner & Müssig, 2011) thus decreasing the strength properties, particularly as a result of untreated fibers and small effect of modulus properties. The porosity seemed to have less effect on the stiffness of composites, but affected water absorption. Also, the increase in fiber content (70 wt.%) produced a considerable variation of flexural strength due to the low inclusion of matrix between fibers.

Considering the stacking effect, the flexural strength of KBC/PLA and CBK/PLA achieved up to 201 and 102 MPa at 50 wt.%, approximately 49% of KBC/PLA higher than CBK/PLA. The flexural modulus of CBK/PLA varies between those fiber contents, accounting for 5-7 GPa. A high modulus fiber in the outer layer of up to 14 GPa was achieved at 60 wt.% and decreased slightly to 13 GPa at a higher fiber content (70 wt.%). The decreasing trend was discussed by many researchers when the high reinforcement tended to lower the penetration of the matrix to the fibers, thus the poor adhesion of both decreased strength properties. KBC/PLA was assumed to describe the function of high modulus kenaf fibers in the outer layer as a rigid filler to enhance the stiffness of polymer matrix (Avella et al., 2008) and has promising usage for flexural strength (Graupner &

Müssig, 2011; N. Saba et al., 2015). Along with a better surface appearance, a high dispersion of kenaf fiber in PLA produced high flexural properties of KBC/PLA.

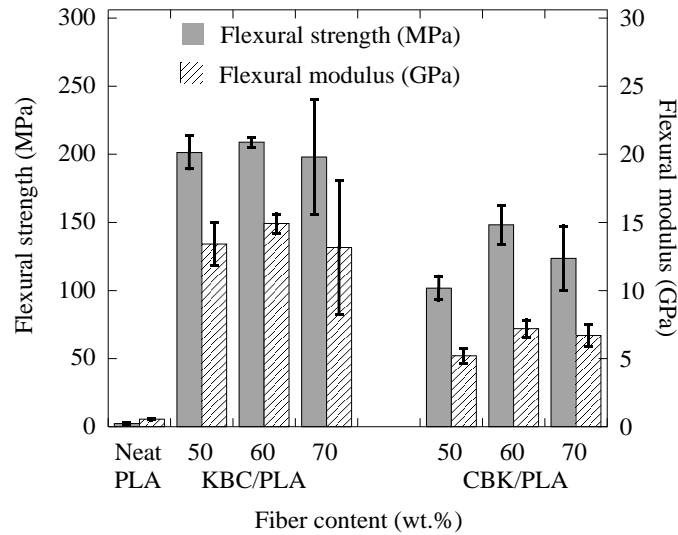


Figure 6.11 Flexural properties of experimental and calculation based on hybrid rule of mixtures.

Compared to literature (Yusoff et al., 2016), the flexural strength and flexural modulus of hybrid green composites shows far higher values compared to those of single fiber plant and hybrid plant/synthetic fiber composites. Some data was obtained to compare with experimental values. It is observed that the stacking sequence of high modulus fiber in the outer layer produced better flexural strength as reported in literature (Ticoalu et al., 2010). Therefore, kenaf and bamboo fiber-reinforced composites are promising as substitutes for plywood or particle board in furniture applications. Flexural failures are governed by fracture of the cell walls themselves in fiber bundles (Kulkarni et al., 1981). The internal structure of the fiber cell walls provides stability to the composites. Lumen, which exists in kenaf and coir fibers, connected to the secondary layer of the cell wall, promoting cohesion by possibly enhancing the mechanical interlock

(Elsaid, Dawood, Seracino, & Bobko, 2011). KBC/PLA produced the strong interaction between the kenaf fiber cell walls and matrix due to the high absorbability of kenaf fiber which correlates to high flexural properties of composites (Almeida et al., 2013; Elsaid et al., 2011). The inclusion of kenaf compensates for the low strength of coir fibers by bearing the bending loads, while coir fiber is capable of high elongation that contributes to higher toughness of the hybrid green composites. Therefore, the stacking sequence has an effect on flexural strength and the flexural modulus of hybrid green composites since internal stress of the high modulus fiber in the outermost layer was equally divided (Peltola, Pääkkönen, Jetsu, & Heinemann, 2014). According to Dong and Davies (C. Dong & Davies, 2014b), flexural strength increases with the span-to depth ratio, at a span ratio greater than 32, flexural becomes stable. Similarly, flexural strength improves with fiber content. These properties are applicable for interior construction materials where exposure to environment is limited (Charlet et al., 2007).

### **6.3.3 Comparison of impact strength of KBC/PLA and CBK/PLA and various green composites**

Both CBK/PLA and KBC/PLA showed an increase in impact strength with fiber content. Up to 70 wt.% fiber content, up to 51 kJ/m<sup>2</sup> and 62 kJ/m<sup>2</sup> were produced by KBC/PLA and CBK/PLA, respectively. It shows that the stacking effect has some effect on impact strength: about 21% higher in CBK/PLA than KBC/PLA. According to Fig.6.13, these hybrid green composites were higher in impact strength than the plant synthetic-based, and could replace synthetic fibers. According to literature (Graupner & Müssig, 2011; Mohammad Jawaid, Abdul Khalil, Bhat, & Abu Baker, 2011; Saw, Akhtar,

Yadav, & Singh, 2014), plant-based and plant-synthetic based composites had varying impact strengths, ranging from 6.5 to 52 kJ/m<sup>2</sup> (Fig. 6.13). Lyocell/PLA was reported to have the highest impact strength (Graupner & Müssig, 2011) compared to other combinations. For plant-synthetic based composites, palm oil empty fruit/jute-reinforced epoxy composites produced 19.47 kJ/m<sup>2</sup> (Mohammad Jawaid et al., 2011). However, a single composite consisting of 40wt. % (Graupner & Müssig, 2011) was higher than hybrid-synthetic fiber. Therefore, instead of using synthetic-based fiber composites, using plant-based composites in production induced high strength by increasing fiber content. The impact strength in the composites was based on crack initiation energy and crack propagation. Crack initiation energy was produced by notched specimens, and the crack propagation was created by a load that exceeded the propagation energy, which made the composite fail completely (Bernard et al., 2011).

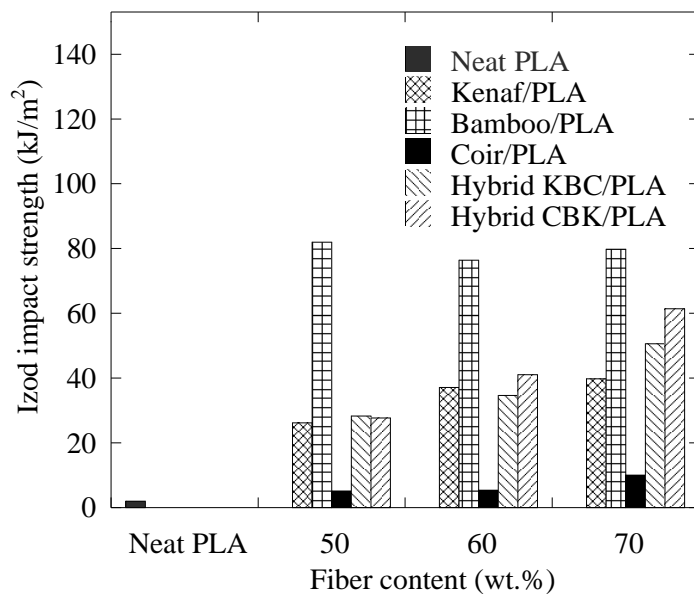


Figure 6.12 Impact strength of hybrid green composites and their single composites.

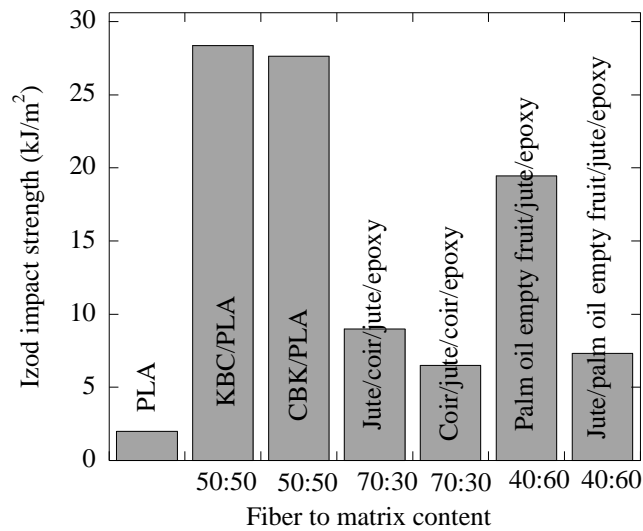


Figure 6.13 Impact strength of KBC/PLA, CBK/PLA and selected synthetic-plant composites.

#### 6.4 Water absorption

The main concern of plant-based composites is their hydrophilic properties due to large amount of hydroxyl groups in cellulose (Verma, Gope, Shandilya, Gupta, & Maheshwari, 2013). This is the main disadvantage of green composite production which affects the interface and strength due to high moisture absorption and water swelling. It is important to examine the swelling factors for wide applications. For plant based composites, the influential factors are fiber content, voids, matrix viscosity, temperature and humid condition (Saw et al., 2014). Swelling with water will increase the weight of the composites. The result of percentage weight change after 2 hours and 48 hours for KBC/PLA and CBK/PLA versus fiber content is presented in Fig. 6.14. Clearly, the swelling rate increased with increased fiber content. In the first two hours, a minimal increase in KBC/PLA of up to 70 wt.% fiber content occurred.

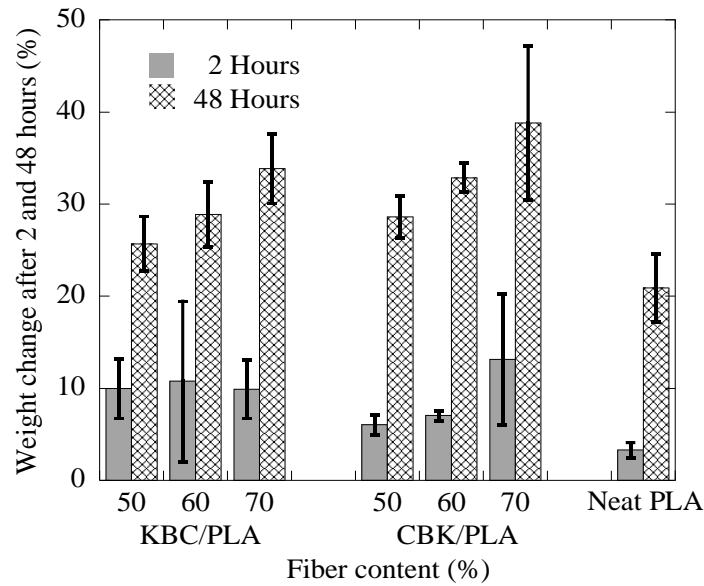


Figure 6.14 Water absorption of KBC/PLA, CBK/PLA and PLA matrix at 50°C temperatures.

In contrast, CBK/PLA increased gradually and higher than KBC/PLA up to 70 wt.% fiber content. After 48 hours of submerging in water, weight increases of as high as 34 % and 39% up to 70 wt. % were achieved for KBC/PLA and CBK/PLA, respectively. Compared to neat PLA, both composites were higher in water absorption, approximately 70-90% higher than PLA. High fiber content provides a fiber network where water easily diffuses in the composites. During hot pressing, the surface nature and dissimilar cross-sections of coir fiber were the reason for the low degree of compaction and low fiber dispersion (Peltola et al., 2014). Therefore, gaps produced between the fiber networks made it easy for water to penetrate in CBK/PLA. Coir fiber in the outer layer was also the reason for high thickness obtained from trilayer coir/jute/coir and jute/coir/jute composites (Saw et al., 2014). In addition, high water absorption was due to pure coir fiber in the composites. Compared to KBC/PLA, these composites were covered with



small cross-sections of kenaf fiber where the gaps were not obvious as coir fiber in the outer layer. The compatibility of kenaf and PLA resulted in better dispersion (Masud S. Huda et al., 2008), and better surface finishing than coir fiber. Coir fibers' agglomeration was more apparent in CBK/PLA than in the KBC/PLA sequence. The swelling property increases linearly with fiber content. The role of low dispersion of fibers contributed to a high swelling rate. The more water was absorbed, transferred stress between fiber and the matrix was decreased (Pérez-Fonseca et al., 2014).

## **6.5 Morphology of hybrid green composites**

To observe the interfaces between PLA matrix and fibers, field emission scanning electron microscopy was used to clarify the failure mechanism of the composites. Figures 6.15 and 6.16 show the images of KBC/PLA and CBK/PLA surface failures, respectively. As shown in the figures, the similar fracture behaviour of both composites surfaces after tensile strength. Among the three fibers, kenaf fiber has a smaller cross section area than coir and bamboo fibers. It was observed that the use of bamboo fiber was the reason for a poor interfacial between fiber networks and matrix as evidenced by voids (holes) surrounding the bamboo fibers, meaning the bamboo had split as well as pulled out of the matrix (Fig. 6.15 (a)-(b)). Despite poor interfacial strength, it could be that the fiber had shorter than critical lengths (Pickering & Aruan Efendy, 2016), which depends on bond strength (Hull, 1981). In addition, the untreated bamboo fiber had reduced the strength of matrix to bond with fiber networks, consequently it was unable to transfer stress to the fibers during tensile stress. As for the presence of lumens in fiber cell walls, they could provide interlocking behaviour in cell walls and the matrix of coir and kenaf

fibers (Khalil et al., 2006). However, with the high viscosity of the PLA matrix, ineffective wetting of fiber may produce de-cohesion of fiber cell walls and fracture in the composites. Therefore, a few lumens were covered with matrix, yielding to a small increase in strength and stiffness. Generally, the penetration of the matrix into the lumens may occur via the porous structure of plant cell walls (Frihart, 2006). By placing coir fiber in the outer layer, it is observed that coir only acts as a filler rather than a reinforcement in polymer. Furthermore, the finished surface with coir fiber in the outer layer produced uneven fiber dispersion in the composites. The dissimilarity of cross-section areas along the fibers produced gaps between fiber networks and layers. In addition, kenaf fibers stimulate the nucleation of PLA to have higher impact strength and toughness and firmly adhere in the matrix (Avella et al., 2008; Han, Karevan, Bhuiyan, et al., 2012) since the fibers act as a reinforcement phase, preventing stress concentration in critical areas. Though PLA is effective in splitting the bamboo fiber (Peltola et al., 2014), bamboo cell walls have impurities on their surfaces which are incompatible with PLA, leading to fibers pulling out and splitting during tensile loads (Figs. 6.15 (b) and 6.16 (c)). The function of the matrix to bind the fibers and transmit the stress to the fibers had failed due to poor adhesion with the matrix. However, kenaf fibers were well adhered and well dispersed in the PLA matrix during hot pressing compaction, thus delivering high strength to the composites and lower water absorption (Fig. 6.16).

In terms of fiber selection and stacking sequence, the composites may perform high tensile properties. Generally, a good interfacial bonding allows stresses to be transferred between the matrix and the reinforcing fibers, to yield higher strength and stiffness. Through effective wetting of the fibers and the matrix, a strong interfacial adhesion may be generated (Bourmaud et al., 2016). Otherwise poor interfacial interaction leads to

internal strain, porosity, and environmental degradation (Bledzki et al., 1996). Hence, an appropriate selection of fiber and polymer matrix is vital to achieve optimum compatibility of fiber and matrix. Further physical and chemical treatments should be carried out for better adhesion between hybrid constituents.

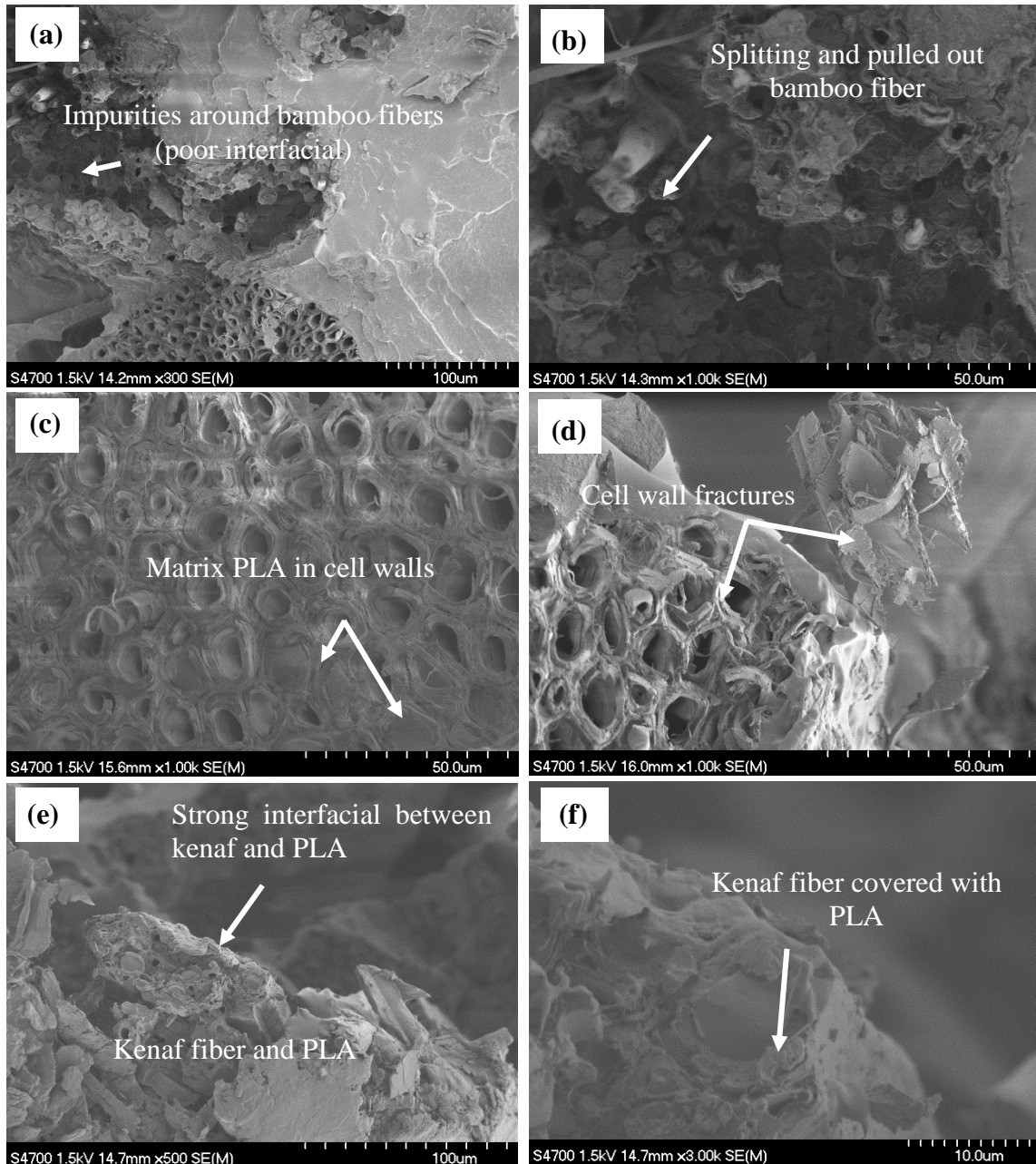


Figure 6.15 SEM images of fiber fracture (a)-(b) bamboo (c)-(d) coir and (e)–(f) kenaf in KBC/PLA.

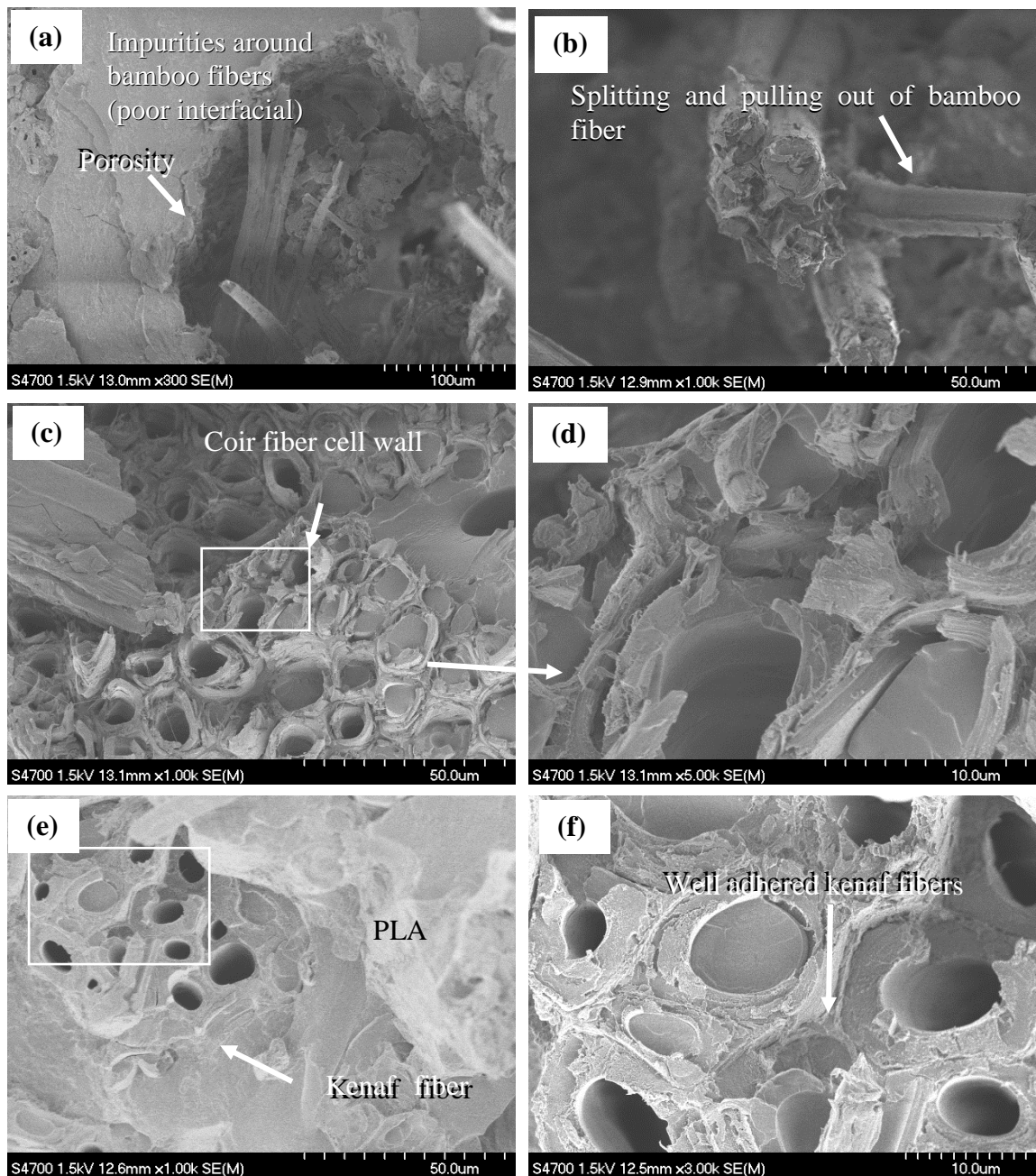


Figure 6.16 SEM images of fiber fracture (a)-(b) bamboo (c)-(d) coir and (e)-(f) kenaf in CBK/PLA.

## 6.6 Loss factor properties of KBCCBK/PLA

Vibrating structure dissipates and radiates energy in the form of kinematical and potential energies which were stored by mass and elasticity of the materials, respectively. The energy dissipates from internal leads to the conversion of mechanical energy into thermal energy (Iwaniec, 2003). To determine the energy dissipation (energy losses), loss factor ( $\eta$ ) can be measured experimentally by using Central Excitation Method (JIS K7391).

Figures 6.17 show the reduction of the loss factor from 0.00751 to 0.00695 up to 80 wt.% fiber content, accounting for a 7.5% decrease, while the frequency increased 22% up to 80 wt.% fiber content, ranging from 175 to 213 Hz (22%). The highest loss factor was produced at 60 wt.%, whereas at higher fiber content, the highest frequency was obtained. Compared to carbon fiber-reinforced composites (CFRP), the damping loss factor of this hybrid composite was 590% higher than CFRP (0.001) (Fujimoto, 1994).

In general, to control prolonged severe vibrations, fiber's orientation, thickness and suitable fibers are factors to be considered before designing the composites. The viscoelastic property of plant fiber is a potential material when desiring a higher loss factor than that produced from synthetic fibers. It was reported that the viscoelastic material used as dissipation energy elements decreases the vibration level of rigid structures (Gelfuso et al., 2014).

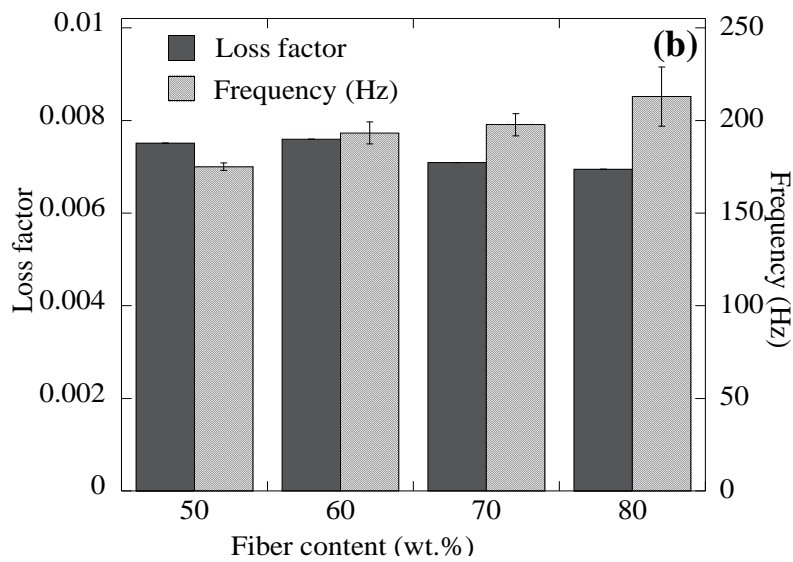
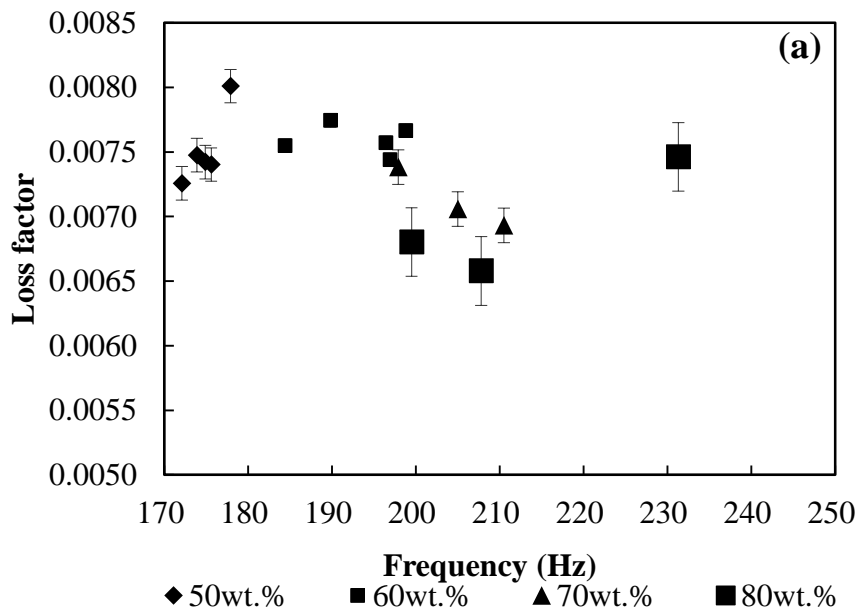


Figure 6.17 Comparison of (a) loss factor of testing specimens (b) average loss factor of KBCBK/PLA with fiber content.

## **6.7 Comparisons of loss factor properties between KBCCBK/PLA and synthetic composites**

It was reported that synthetic fibers, such as Kevlar are better for damping property than glass and graphite (Hoa & Ouellette, 1984). The unidirectional graphite tape/epoxy was mentioned to have a lower loss factor (0.0042) than that of unidirectional Kevlar fabric/epoxy (0.0134). In terms of thickness, loss factor had a minimal dependence on specimens' thickness when comparing with 8 and 20 stacking ply of graphite/epoxy (Table 6.1), whereas, it had 50 % less than that of 8 and 20 stacking ply specimens (Crane & Gillespie, 1989). Therefore, the thicker and stiffer synthetic fibers do not always represent a higher damping capacity than all plant fibers. Referring to Table 6.1, hybrid green composites (KBCCBK/PLA) have considerably high damping properties. Plant fiber reinforcement presents intermediate behavior between pure elastic and plastic deformation. During damping vibration, not all energy is converted into deformation. Some energy is used to resist internal friction of the material, which is called "energy dissipation effect" (Gelfuso et al., 2014). The inherent property of viscoelastic plant fiber is the advantage to reduced vibration effect that is applicable to reduce undesired vibration effect. This result provides wider applications of plant fibers as the alternative to solve the indispensability of non-degradable synthetic composites, which lead to environmental loadings where high strength is not of primary importance.

Table 6.1 Comparison properties of KBCCBK/PLA and selected synthetic composites.

Composites	Length (mm)	Thick-ness (mm)	First resonant frequency (Hz)	Loss factors ( $\eta$ ) $10^{-4}$	Damping ( $\zeta$ )	Refs.
KBCCBK/PLA (50wt.%)	198	2.2-2.6	175	75	0.375	Present study
KBCCBK/PLA (60wt.%)	198	2.4-2.6	193	76	0.379	Present study
KBCCBK/PLA (70wt.%)	198	2.2-2.5	198	71	0.354	Present study
KBCCBK/PLA (80wt.%)	198	2.3-2.6	213	69	0.347	Present study
Graphite tape/epoxy (air)	200	1.1	62	99		(Hoa & Ouellette, 1984)
Graphite tape/epoxy (vacuum)	200		63	42		(Hoa & Ouellette, 1984)
Graphite/Epoxy $[0^0]$	254	1.02	21	21		(Crane & Gillespie, 1989)
Graphite/Epoxy $[0^0]_8$	254	-	21	17		(Crane & Gillespie, 1989)
Graphite/ epoxy $[0^0]_{20}$	184	-	100	18-22		(Crane & Gillespie, 1989)
S2 Glass/epoxy	224	-	38	54-66		(Crane & Gillespie, 1989)
S2 Glass/epoxy	203	-	46	57-59		(Crane & Gillespie, 1989)
Glass fiber mat/PP	200-300	2-3	10	-	0.014-0.042	(Landro & Lorenzi, 2009)
Kenaf/hemp/flax/PP	200-300	2-3	10		0.015-0.038	(Landro & Lorenzi, 2009)



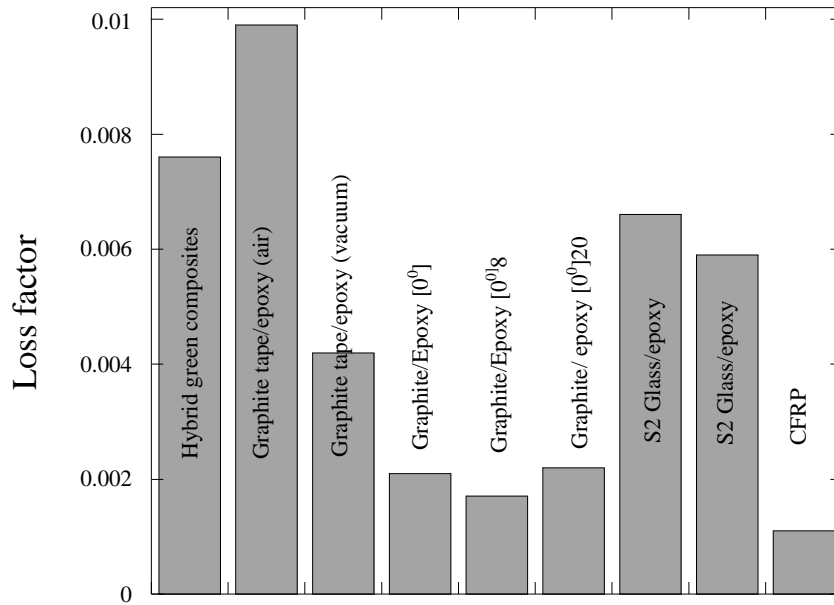


Figure 6.18 Loss factor of hybrid green composites and selected synthetic composites.

## 6.8 Loss factor properties of monolithic green composites

The unique property of a plant fiber-based composite is its potential to yield a higher loss factor than that of synthetic composites. Lumen in the plant cell allows for embracing a more diversified mode to attenuate sound wave energy, which is useful for sound absorption (Zhu et al., 2014). The green composites, such as coir fiber composites are suitable for automotive and non-structural components, when high mechanical properties are not very important.

This result is to reveal the damping loss factor of one type of plant fiber-reinforced PLA. We focused on measuring the monolithic green composites which are kenaf/PLA, bamboo/PLA and coir/PLA for their own damping loss factor property for future

improvement in hybridization. Table 6.2 shows the result of each kind of plant fiber-reinforced PLA. It can be seen that coir/PLA displayed the higher loss factor than kenaf/PLA and bamboo/PLA. The deformability of coir fiber produces the highest loss factor among all the composites. Moreover, coir fiber-reinforced polypropylene (coir/PP) tested by dynamic testing was reported to produce higher loss factor compared to this study (Gelfuso et al., 2014). The similar report mentioned that the balance between the elastic and viscous phase, and the damping behaviour of the material influence the loss factor. Also, the damping mechanism is strongly dependent on temperature and has limited damping properties at high frequency.

As a consequence of hybrid design, the loss factor has increased 10% compared to monolithic composites, kenaf and bamboo reinforced-PLA. Despite a small increment in the loss factor, this hybrid green composite shows higher strength properties than that of coir fiber reinforced-PLA. Thus, it is essential to combine those fibers in fabricating hybrid green composites instead of one fiber type reinforcement that may exhibit low property.

Table 6.2 Comparison properties of monolithic green composites and coir/PP composites.

Composites	Length (mm)	Thick-ness (mm)	First resonant frequency (Hz)	Loss factors ( $\eta$ ) $10^{-4}$	Damping ( $\zeta$ )	Refs.
Coir/PLA (60 wt.%)	194	2.26	93	174	0.868	Present study
Kenaf/PLA (60 wt.%)	197	2.06	178.92	72	0.35	Present study
Bamboo/PLA (60 wt.%)	197	1.74	142.23	72	0.35	Present study
Coir/PP (5-30 %)	160	3	20-25	500-580	0.025-0.03	(Gelfuso et al., 2014)

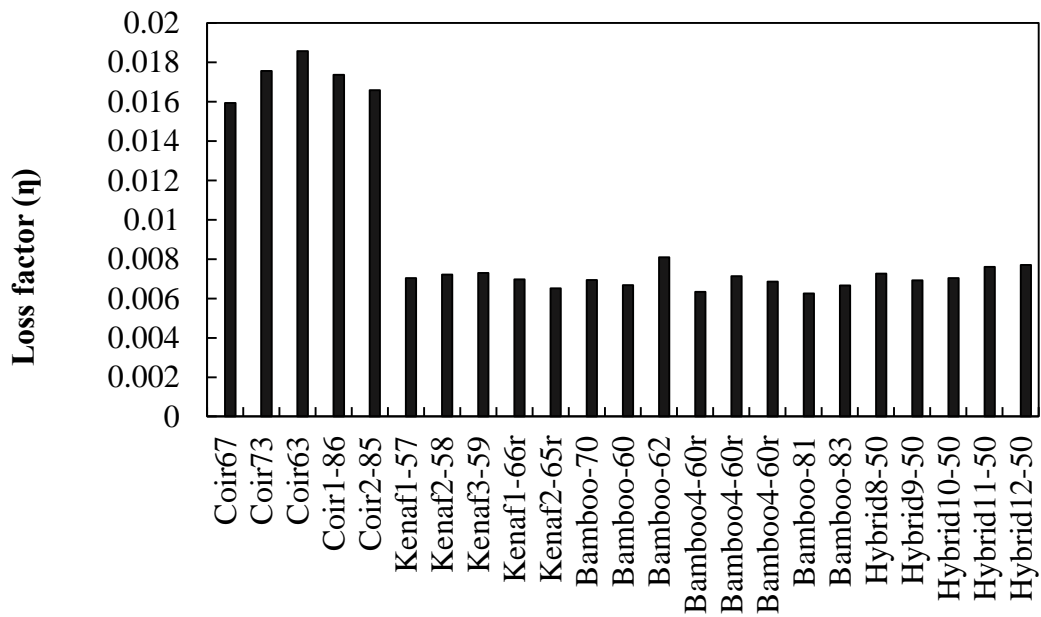


Figure 6.19 Loss factor of monolithic green composites and hybrid green composites.

# CHAPTER SEVEN

## Conclusion

### 7.1 Introduction

Plant fibers are the most promising renewable sources for new composite production. The fibers can be reinforced to produce various consumer products due to an acceptable performance, process-ability, unlimited resources at low cost, and environmental friendliness. In addition, the natural fibers are less dependent on petroleum-derived products. A few stacking sequences of composites have been produced in this study. It is found that the typical factors affecting the composite properties are fibers-matrix interfacial bonding, fibers' stacking sequences, fiber content, processing temperatures, and fibers' alignments. The hybrid green composites provide fully eco-designed materials to increase the inherited low mechanical properties of one fiber, compensated by other fibers.

### 7.2 Monolithic green composites

In terms of monolithic green composites, kenaf/PLA had higher tensile properties than bamboo/PLA and coir/PLA. Even though bamboo has higher tensile properties based on fiber bundles tensile test, the weak interfacial bonding between bamboo and PLA was the reason for lower strength than kenaf/PLA, whereas coir/PLA had a better elongation property than those of bamboo/PLA and kenaf/PLA. It was found that coir fiber

reinforced-PLA had no significant increase in stiffness at these amounts of fiber content compared to the kenaf and bamboo fiber reinforced-PLA.

### **7.3. Hybrid green composites**

#### **7.3.1 High modulus fibers in the outer layer (KBCCBK/PLA, KCCK/PLA and BCCB/PLA)**

Compared to all types of hybrid green composites, a higher tensile strength and elastic modulus were obtained from a combination of kenaf, bamboo and coir fibers (KBCCBK/PLA and KBC/PLA). This is a consequence of kenaf and bamboo fibers having higher mechanical properties, preventing failure of the composites. High flexural modulus was obtained in KCCK/PLA as a result of the good bonding of kenaf-PLA and the deformable nature of coir fiber.

#### **7.3.2 High and low modulus fibers in the outer layer (KBC/PLA and CBK/PLA)**

While the delamination of high modulus fibers (kenaf) in the outer layers produced higher flexural properties, the delamination of low modulus fibers (coir) in the outer layers decreased in flexural strength. With similar fiber content, tensile strength had a small effect on stacking sequences, either placing high and low modulus in the middle layer or high modulus in the outer layers. However, it had a high impact on the flexural properties of the composite. Higher flexural strength and flexural modulus were obtained from the combination of high modulus fibers (KBC/PLA). In addition, the placing of low modulus coir fiber (CBK/PLA) produced higher strain property. Coir was estimated to promote elongation of hybrid green composites and counteract the inherited properties of

low elongation of bamboo and kenaf fibers in the composites. It was found that the low modulus of plant fiber and high modulus fiber provides a barrier to impact load, and prevents flexural loads respectively. This may be suitable for front parts of vehicles to reduce impact loads.

### **7.3.3 Loss factor of KBCCBK/PLA**

Higher damping loss factor was obtained from this hybrid green composite. The value is higher than that of glass fiber reinforced composites. It has potential to replace GFRP and CFRP with plant fiber composites for vibration and noise reduction, such as in the automobiles and interior parts for the safety and durability of the products. It may resist load impact in most applications, structural and non-structural. The advantages of green composites of low density, low-cost production, and high specific strength, as well as ecofriendliness, may reduce the impact of high loading of land fillings due to the non-degradability of synthetics composites.

## REFERENCES

- Abdul Khalil, H. P. S., Bhat, I. U. H., Jawaid, M., Zaidon, A., Hermawan, D., & Hadi, Y. S. (2012). Bamboo fibre reinforced biocomposites: A review. *Materials and Design*, 42, 353–368.
- Akil, H. M., Omar, M. F., Mazuki, A. A. M., Safiee, S. M., Ishak, Z. A. M., & Abu Bakar, A. (2011). Kenaf fiber reinforced composites: A review. *Materials and Design*, 32(8–9), 4107–4121.
- Alex, S., & Retnam, S. . J. (2014). A review on degradable hybrid natural fibre. In *International Journal of Design and Manufacturing Technology (IJDMT)* (Vol. 5, pp. 137–141). Ernakulam, 30-31 December.
- Ali, M. (2011). Coconut fibre: A versatile material and its applications in engineering. *Journal of Civil Engineering and Construction Technology*, 2(9), 189–197.
- Almeida, J. H. S., Amico, S. C., Botelho, E. C., & Amado, F. D. R. (2013). Hybridization effect on the mechanical properties of curaua/glass fiber composites. *Composites Part B: Engineering*, 55, 492–497.
- André, A. (2006). *Fibres for strengthening of timber structures*. Luleå University of Technology.
- Andreas, M. (Ed.). (2007). *Concise Encyclopedia of Composite Materials* (2nd ed.). Oxford, UK: Elsevier.
- Ashby, M. F. (2008). The CES EduPack Database of Natural and Man-Made Materials. In *Cambridge University* (Vol. 1, pp. 1–26). Retrieved from Cambridge
- Assamoi, B., & Lawryshyn, Y. (2012). The environmental comparison of landfilling vs. incineration of MSW accounting for waste diversion. *Waste Management*, 32(5), 1019–1030.
- Avella, M., Bogoeva-Gaceva, G., Buzarovska, A., Emauela Errico, M., Gentile, G., & Grozdanov, A. (2008). Poly(lactic acid)-Based Biocomposites reinforced with Kenaf fibres. *Journal of Applied Polymer Science*, 108, 3542–3551.
- Azwa, Z. N., Yousif, B. F., Manalo, A. C., & Karunasena, W. (2013). A review on the degradability of polymeric composites based on natural fibres. *Materials and Design*, 47, 424–442.
- Baley, C. (2002). Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase. *Composites - Part A: Applied Science and Manufacturing*, 33(7), 939–948.
- Barnett, J. R., & Bonham, V. A. (2004). Cellulose microfibril angle in the cell wall of wood fibres. *Biological Reviews of the Cambridge Philosophical Society*, 79(2), 461–72.
- Beckwith, W. (2003). Natural Fiber Reinforcement Materials Lower Cost Technology for Composites Applications. *Composites Fabrication*, 12–16.
- Begum, K., & Islam, M. A. (2013). Natural Fiber as a substitute to synthetic fiber in polymer composites : A review. *Research Journal of Engineering Sciences*, 2(3), 46–53.
- Bernard, M., Khalina, A., Ali, A., Janius, R., Faizal, M., Hasnah, K. S., & Sanuddin, A. B. (2011). The effect of processing parameters on the mechanical properties of kenaf fibre plastic composite. *Materials and Design*, 32(2), 1039–1043.

- Biagiotti, J., Puglia, D., & Kenny, J. M. (2004). A Review on Natural Fibre- Based Composites-Part I: Structure, Processing and Properties of Vegetable Fibres. *Journal of Natural Fibers*, 1(2), 37–41.
- Bledzki, A. K., & Gassan, J. (1999). Composites reinforced with cellulose based fibres. *Progress in Polymer Science (Oxford)*, 24(2), 221–274.
- Bledzki, A. K., Reihmane, S., & Gassan, J. (1996). Properties and modification methods for vegetable fibers for natural fiber composites. *Journal of Applied Polymer Science*, 59(8), 1329–1336.
- Bledzki, A. K., Sperber, V. E., & Faruk, O. (2002). *Natural and Wood Fibre Reinforcement in Polymers*. Rapra Technology Limited.
- Bourmaud, A., Duigou, A. Le, Gourier, C., & Baley, C. (2016). Influence of processing temperature on mechanical performance of unidirectional polyamide 11–flax fibre composites. *Industrial Crops and Products*, 84, 151–165.
- Buchmeiser, M. R. (2007). Polymeric monolithic materials: Syntheses, properties, functionalization and applications. *Polymer*, 48(8), 2187–2198.
- Cai, M., Takagi, H., Nakagaito, A. N., Katoh, M., Ueki, T., Waterhouse, G. I. N., & Li, Y. (2015). Influence of alkali treatment on internal microstructure and tensile properties of abaca fibers. *Industrial Crops and Products*, 65, 27–35.
- Campbell, F. C. (2010). Chapter 1: Introduction to composite materials. In *Structural Composites Materials* (p. 3). Ohio: ASM International.
- Carfagni, M., Lenzi, E., & Pierini, M. (1994). The loss factor as a measure of mechanical damping, 580–584. Retrieved from <https://pdfs.semanticscholar.org/c33d/393514b86fea3045751429361eb4368db99d.pdf>
- Céline, A., Fréour, S., Jacquemin, F., & Casari, P. (2013). The hygroscopic behavior of plant fibers: a review. *Frontiers in Chemistry*, 1, 43.
- Charlet, K., Baley, C., Morvan, C., Jernot, J. P., Gomina, M., & Bréard, J. (2007). Characteristics of Hermès flax fibres as a function of their location in the stem and properties of the derived unidirectional composites. *Composites Part A: Applied Science and Manufacturing*, 38(8), 1912–1921.
- Crane, R. M. (1991). *Vibration damping response of composites materials*. David Taylor Research Center. Bethesda. Retrieved from <http://www.dtic.mil/dtic/tr/fulltext/u2/a235614.pdf>
- Crane, R. M., & Gillespie, J. W. (1989). *Damping loss factor determination of glass and graphite composites*. Bethesda.
- Curtis, A. J., Tinling, N. G., & Abstein, H. T. (Eds.). (1971). Vibration Equipment Requirement. In *Selection and performance of vibration tests - Allen J. Curtis, Nickolas G. Tinling, Henry T. Abstein, Shock and Vibration Information Center* (p. 103). USA: Shock and Vibration Information Center.
- Davallo, M., Pasdar, H., & Mohseni, M. (2010). Effects of laminate thickness and ply-stacking sequence on the mechanical properties and failure mechanism of unidirectional glass-polyester composites. *International Journal of ChemTech Research*, 2(4), 2118–2124.
- Dittenber, D. B., & Gangarao, H. V. S. (2012). Critical review of recent publications on use of natural composites in infrastructure. *Composites Part A: Applied Science and Manufacturing*, 43(8), 1419–1429.



- Dong, C., & Davies, I. J. (2014a). Flexural and tensile moduli of unidirectional hybrid epoxy composites reinforced by S-2 glass and T700S carbon fibre. *Materials & Design*, *54*, 893–899.
- Dong, C., & Davies, I. J. (2014b). Flexural and tensile strengths of unidirectional hybrid epoxy composites reinforced by S-2 glass and T700S carbon fibres. *Materials and Design*, *54*, 955–966.
- Dong, Y., Ghataura, A., Takagi, H., Haroosh, H. J., Nakagaito, A. N., & Lau, K. T. (2014). Polylactic acid (PLA) biocomposites reinforced with coir fibres: Evaluation of mechanical performance and multifunctional properties. *Composites Part A: Applied Science and Manufacturing*, *63*, 76–84.
- Duhovic, M., Horbach, S., & Bhattacharyya, D. (2009). Improving the interface strength in flax fibre poly(lactic) acid composites. *Journal of Biobased Materials and Bioenergy*, *3*(2), 188–198.
- Edeerozey, A. M. M., Akil, H. M., Azhar, A. B., & Ariffin, M. I. Z. (2007). Chemical modification of kenaf fibers. *Materials Letters*, *61*(10), 2023–2025.
- Elsaid, A., Dawood, M., Seracino, R., & Bobko, C. (2011). Mechanical properties of kenaf fiber reinforced concrete. *Construction and Building Materials*, *25*(4), 1991–2001.
- Fei, P., Fei, B., Yu, Y., Xiong, H., & Tan, J. (2014). Thermal properties and crystallization behavior of bamboo fiber/high-density polyethylene composites: Nano-Nano-TiO<sub>2</sub> effects. *Journal of Applied Polymer Science*, *131*(3), 1–10.
- Frihart, C. R. (2006). Wood structure and adhesive bond strength. In D. D. Stokke & L. H. Groom (Eds.), *Characterization of the Cellulosic Cell Wall* (pp. 241–254). Ames: Blackwell Publishing.
- Fujimoto, J. (1994). Damping Properties under Flexural Vibration for CFRP / Damping Materials Laminates. *日本複合材料学会誌*, *4*(1994), 144–153.
- Gelfuso, M. V, Thomazini, D., de Souza, J. C. S., & de Lima, J. J. (2014). Vibrational Analysis of Coconut Fiber-PP Composites. *Materials Research-Ibero-American Journal of Materials*, *17*(2), 367–372.
- Gibson, L. J. (2012). The hierarchical structure and mechanics of plant materials. *Journal of The Royal Society Interface*, *9*(76), 2749–2766.
- Gnanapragasam, A. A., Chitra, G., & Ravi, S. R. (2016). Study on strengthening of RC beam column joint using hybrid FRP composites. *Circuits and Systems*, *7*, 2846–2856.
- Goda, K., Takagi, H., & Netravali, A. N. (2008). Fully biodegradable green composites reinforced with natural fibers. In S. Thomas & L. A. Pothan (Eds.), *Natural Fibres Reinforced Polymer Composites from Macro to Nanoscale* (pp. 329–355). Philadelphia: Old City Publishing, Inc.
- Gordon, J. E. (1978). *Structures, or Why Things Don't Fall Down*. London: Penguin Group.
- Gosline, J. M., Guerette, P. A., Ortlepp, C. S., & Savage, K. N. (1999). The mechanical design of spider silks: from fibroin sequence to mechanical function. *The Journal of Experimental Biology*, *202*, 3295–3303.
- Graupner, N., & Müssig, J. (2011). A comparison of the mechanical characteristics of kenaf and lyocell fibre reinforced poly(lactic acid) (PLA) and poly(3-hydroxybutyrate) (PHB) composites. *Composites Part A: Applied Science and Manufacturing*, *42*(12), 2010–2019.

- H'ng, P. S., Khor, B. N., Tadashi, N., Aini, a. S. N., & Paridah, M. T. (2009). Anatomical structures and fiber morphology of new kenaf varieties. *Asian Journal of Scientific Research*.
- Han, S. O., Karevan, M., Bhuiyan, M. A., Park, J. H., & Kalaitzidou, K. (2012). Effect of exfoliated graphite nanoplatelets on the mechanical and viscoelastic properties of poly(lactic acid) biocomposites reinforced with kenaf fibers. *Journal of Materials Science*, 47(8), 3535–3543.
- Han, S. O., Karevan, M., Sim, I. N., Bhuiyan, M. A., Jang, Y. H., Ghaffar, J., & Kalaitzidou, K. (2012). Understanding the reinforcing mechanisms in kenaf fiber/PLA and kenaf fiber/PP composites: A comparative study. *International Journal of Polymer Science*, 2012, 1–8.
- Harish, S., Michael, D. P., Bensely, a., Lal, D. M., & Rajadurai, a. (2009). Mechanical property evaluation of natural fiber coir composite. *Materials Characterization*, 60(1), 44–49.
- Hoa, S. V., & Ouellette, P. (1984). Damping of composite materials. *Polymer Composites*, 5(4), 334–338.
- Huda, M. S., Drzal, L. T., Misra, M., & Mohanty, A. K. (2006). Wood-fiber-reinforced poly(lactic acid) composites: Evaluation of the physicochemical and morphological properties. *Journal of Applied Polymer Science*, 102(5), 4856–4869.
- Huda, M. S., Drzal, L. T., Mohanty, A. K., & Misra, M. (2008). Effect of fiber surface-treatments on the properties of laminated biocomposites from poly(lactic acid) (PLA) and kenaf fibers. *Composites Science and Technology*, 68(2), 424–432.
- Hull, D. (1981). *An Introduction to Composite Materials*. Cambridge University Press.
- Ichhaporia, P. K. (2008). Composites from Natural Fibers. *Thesis*.
- Iwaniec, M. (2003). Damping loss factor estimation in plates. *Molecular and Quantum Acoustics*, 24, 61–68.
- Jain, S., Kumar, R., & Jindal, U. C. (1992). Mechanical behaviour of bamboo and bamboo composite. *Journal of Materials Science*, 27(17), 4598–4604.
- Jawaid, M., & Abdul Khalil, H. P. S. (2011). Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. *Carbohydrate Polymers*, 86(1), 1–18.
- Jawaid, M., Abdul Khalil, H. P. S., Bhat, A. H., & Abu Baker, A. (2011). Impact Properties of Natural Fiber Hybrid Reinforced Epoxy Composites. *Advanced Materials Research*, 264–265(March 2016), 688–693.
- John, M. J., & Thomas, S. (2008). Biofibres and biocomposites. *Carbohydrate Polymers*, 71(3), 343–364.
- Jonoobi, M., Harun, J., Shakeri, A., Misra, M., & Oksmand, K. (2009). Chemical composition, crystallinity, and thermal degradation of bleached and unbleached kenaf bast (*Hibiscus cannabinus*) pulp and nanofibers. *BioResources*, 4(2), 626–639.
- Kalia, S., Dufresne, A., Cherian, B. M., Kaith, B. S., Avérous, L., Njuguna, J., & Nassiopoulos, E. (2011). Cellulose-based bio- and nanocomposites: A review. *International Journal of Polymer Science*, 2011.
- Kang, J. T., Park, S. H., & Kim, S. H. (2014). Improvement in the adhesion of bamboo fiber reinforced polylactide composites. *Journal of Composite Materials*, 48(21), 2567–2577.
- Khalil, H. P. S. A., Alwani, M. S., & Omar, A. K. M. (2006). Chemical composition, anatomy, lignin distribution, and cell wall structure of Malaysian plant waste fibers. *BioResources*, 1(2), 220–232.

- Kistaiah, N., Kiran, C. U., Reddy, G. R., & Rao, M. S. (2014). Mechanical characterization of hybrid composites: A review. *Journal of Reinforced Plastics and Composites*, 33(14), 1364–1372.
- Kulkarni, A. G., Satyanarayana, K. G., Sukumaran, K., & Rohatgi, P. K. (1981). Mechanical behaviour of coir fibres under tensile load. *Journal of Materials Science*, 16(4), 905–914.
- La Mantia, F. P., & Morreale, M. (2011). Green composites: A brief review. *Composites Part A: Applied Science and Manufacturing*, 42(6), 579–588.
- Landro, L. Di, & Lorenzi, W. (2009). Mechanical Properties and Dynamic Mechanical Analysis of Thermoplastic-Natural Fiber / Glass Reinforced Composites, 145–155.
- Lertwattanaruk, P., & Suntijitto, A. (2015). Properties of natural fiber cement materials containing coconut coir and oil palm fibers for residential building applications. *Construction and Building Materials*, 94, 664–669.
- Lewin, M., & Pearce, E. M. (Eds.). (1998). *Handbook of Fiber Chemistry* (2nd ed.). New York: Marcel Dekker, Inc.
- Li, L.-J., Wang, Y.-P., Wang, G., Cheng, H.-T., & Han, X.-J. (2010). Evaluation of properties of natural bamboo fiber for application in summer textiles. *Journal of Fiber Bioengineering and Informatics*, 3(2), 94–99.
- Mansor, M., Sapuan, S., Zainudin, E., Nuraini, A., & Hambali, A. (2013). Stiffness prediction of hybrid kenaf/Glass Fiber reinforced polypropylene composites using Rule of Mixtures ( ROM ) and Rule of hybrid mixtures ( RoHM ). *J. Polym. Mater.*, 30(November), 321–334.
- McGregor, O., Duhovic, M., Lin, R. J. T., & Bhattacharyya, D. (2010). High performance biodegradable composite materials. In *7th Asian-Australasian Conference on Composite Materials 2010*, 1, 47–50.
- Mitra, B. C. (2014). Environment friendly composite materials: Biocomposites and green composites. *Defence Science Journal*, 64(3), 244–261.
- Mohammed, L., Ansari, M. N. M., Pua, G., Jawaid, M., & Islam, M. S. (2015). A review on natural fiber reinforced polymer composite and its applications. *International Journal of Polymer Science*, 2015, 1–15.
- Monteiro, S., N., Satyanarayana, K., G., Ferreira, A., S., Nascimento, D., C., O., Silva, I., L., A., Bevitori, A., B., Portela, T., G. (2011). Selection of high strength natural fibers. *Revista Matéria*, 15(4), 488–505.
- Monteiro, S. N., Lopes, F. P. D., Barbosa, A. P., Bevitori, A. B., Silva, I. L. A. Da, & Costa, L. L. Da. (2011). Natural Lignocellulosic Fibers as Engineering Materials—An Overview. *Metallurgical and Materials Transactions A*, 42(10), 2963–2974.
- Müssig, J. (2010). *Industrial Applications of Natural Fibres: Structure, Properties and Technical Applications*. John Wiley & Sons.
- Mwaikambo, L. Y. (2006). Review of the history, properties and application of plant fibres. *African Journal of Science and Technology*, 7(2), 120–133.
- Nam, T. H., Ogihara, S., Tung, N. H., & Kobayashi, S. (2011). Effect of alkali treatment on interfacial and mechanical properties of coir fiber reinforced poly(butylene succinate) biodegradable composites. *Composites Part B: Engineering*, 42(6), 1648–1656.
- Nema, T., Chan, E. C. Y., & Ho, P. C. (2014). Applications of monolithic materials for sample preparation. *Journal of Pharmaceutical and Biomedical Analysis*, 87, 130–141.

- Netravali, A., & Chaba, S. (2003). Composites get greener. *Materials Today*, 9(7–8), 8. Retrieved from [http://doi.org/10.1016/S1369-7021\(06\)71564-4](http://doi.org/10.1016/S1369-7021(06)71564-4)
- Nishino, T., Hirao, K., Kotera, M., Nakamae, K., & Inagaki, H. (2003). Kenaf reinforced biodegradable composite. *Composites Science and Technology*, 63(9), 1281–1286.
- Ochi, S. (2008). Mechanical properties of kenaf fibers and kenaf/PLA composites. *Mechanics of Materials*, 40(4–5), 446–452.
- Oksman, K., Skrifvars, M., & Selin, J.-F. (2003). Natural fibres as reinforcement in polylactic acid (PLA) composites. *Composites Science and Technology*, 63(9), 1317–1324.
- Okubo, K., Fujii, T., & Yamamoto, Y. (2004). Development of bamboo-based polymer composites and their mechanical properties. *Composites Part A: Applied Science and Manufacturing*, 35(3), 377–383.
- Okubo, K., Fujii, T., & Yamashita, N. (2005). Improvement of interfacial adhesion in bamboo polymer composite enhanced with micro-fibrillated cellulose. *JSME International Journal Series A*, 48(4), 199–204.
- Paci, R., Flores, D., & Ayson, F. A. (2014). Monolithic composite structures for vehicles. *Patent US 2014/0042271 A1*. USA.
- Pandey, J. K., Ahn, S. H., Lee, C. S., Mohanty, A. K., & Misra, M. (2010). Recent Advances in the Application of Natural Fiber Based Composites. *Macromolecular Materials and Engineering*, 295(11), 975–989.
- Peltola, H., Pääkkönen, E., Jetsu, P., & Heinemann, S. (2014). Wood based PLA and PP composites: Effect of fibre type and matrix polymer on fibre morphology, dispersion and composite properties. *Composites Part A: Applied Science and Manufacturing*, 61, 13–22.
- Pérez-Fonseca, A. A., Robledo-Ortíz, J. R., Ramirez-Arreola, D. E., Ortega-Gudiño, P., Rodrigue, D., & González-Núñez, R. (2014). Effect of hybridization on the physical and mechanical properties of high density polyethylene–(pine/agave) composites. *Materials & Design*, 64, 35–43.
- Pickering, K. L., & Aruan Efendy, M. G. (2016). Preparation and mechanical properties of novel bio-composite made of dynamically sheet formed discontinuous harakeke and hemp fibre mat reinforced PLA composites for structural applications. *Industrial Crops and Products*, 84, 139–150.
- Pickering, K. L., Aruan Efendy, M. G., & Le, T. M. (2016). A review of recent developments in natural fibre composites and their mechanical performance. *Composites Part A: Applied Science and Manufacturing*, 83, 98–112.
- Piggott, M. R. (1987). The effect of the interface/interphase on fiber composite properties. *Polymer Composites*, 8(5), 291–297.
- R'Mili, M., Moevus, M., & Godin, N. (2008). Statistical fracture of E-glass fibres using a bundle tensile test and acoustic emission monitoring. *Composites Science and Technology*, 68(7–8), 1800–1808.
- Ramesh, M., Palanikumar, K., & Reddy, K. H. (2013). Comparative evaluation on properties of hybrid Glass fiber- sisal/jute reinforced epoxy composites. *Procedia Engineering*, 51, 745–750.
- Rowell, R. M., Young, R. A., & Rowell, J. K. (1996). *Paper and Composites from Agro-Based Resources*. (R. M. Rowell, R. A. Young, & J. K. Rowell, Eds.). Boca Raton: CRC Press.

- Saba, N., Paridah, M. T., & Jawaid, M. (2015). Mechanical properties of kenaf fibre reinforced polymer composite: A review. *Construction and Building Materials*, 76, 87–96.
- Saba, N., Tahir, P., & Jawaid, M. (2014). A review on potentiality of nano filler/natural fiber filled polymer hybrid composites. *Polymers*, 6(8), 2247–2273.
- Salleh, Z., Berhan, M., Hyei, K., & Isaac, D. (2012). Cold-pressed kenaf and fibreglass hybrid composites laminates: effect of fibre types. *World Academy of Science, Engineering and Technology*, 6(11), 740–744.
- Sathishkumar, T., Naveen, J., & Satheeshkumar, S. (2014). Hybrid fiber reinforced polymer composites - a review. *Journal of Reinforced Plastics and Composites*, 33(5), 454–471.
- Satyanarayana, K. G., Arizaga, G. G. C., & Wypych, F. (2009). Biodegradable composites based on lignocellulosic fibers-An overview. *Progress in Polymer Science (Oxford)*, 34(9), 982–1021.
- Saw, S. K., Akhtar, K., Yadav, N., & Singh, A. K. (2014). Hybrid composites made from jute/coir Fibers: Water absorption, thickness swelling, density, morphology, and mechanical properties. *Journal of Natural Fibers*, 11(1), 39–53.
- Sen, T., & Reddy, H. N. J. (2011). Application of Sisal , Bamboo , Coir and Jute Natural Composites in Structural Upgradation. *International Journal of Innovation, Management and Technology*, 2(3), 186–191.
- Shah, D. U. (2013). Developing plant fibre composites for structural applications by optimising composite parameters: A critical review. *Journal of Materials Science*, 48(18), 6083–6107.
- Shringi, D., Solanki, G., & Sharma, P. (2014). Mechanical Properties Characterization of Natural ( Coir Based ) Fiber Polymer Composite by Numerical Methods, 3(4), 465–470.
- Sreekala, M. S., George, J., Kumaran, M. G., & Thomas, S. (2002). The mechanical performance of hybrid phenol-formaldehyde-based composites reinforced with glass and oil palm fibres. *Composites Science and Technology*, 62(3), 339–353.
- Sreekala, M. S., Kumaran, M. G., & Thomas, S. (1997). Oil palm fibers: Morphology, chemical composition, surface modification, and mechanical properties. *Journal of Applied Polymer Science*, 66(5), 821–835.
- Sudhir, A., Madhukiran, J., Rao, S. S., & Madhusudan, S. (2014). Tensile and flexural properties of sisal / jute hybrid natural fiber composites. *International Journal Of Modern Engineering Research (IJMER)*, 4(7), 29–35.
- Sukmawan, R., Takagi, H., & Nakagaito, A. N. (2016). Strength evaluation of cross-ply green composite laminates reinforced by bamboo fiber. *Composites Part B: Engineering*, 84, 9–16.
- Suryanegara, L., Nakagaito, A. N., & Yano, H. (2009). The effect of crystallization of PLA on the thermal and mechanical properties of microfibrillated cellulose-reinforced PLA composites. *Composites Science and Technology*, 69(7–8), 1187–1192.
- Takagi, H., & Ichihara, Y. (2004). Effect of Fiber Length on Mechanical Properties of “Green” Composites Using a Starch-Based Resin and Short Bamboo Fibers. *JSME International Journal Series A*, 47(4), 551–555.

- Thakur, V. K., & Thakur, M. K. (2014). Processing and characterization of natural cellulose fibers/thermoset polymer composites. *Carbohydrate Polymers*, *109*(2014), 102–117.
- Ticoalu, A., Aravinthan, T., & Cardona, F. (2010). A review of current development in natural fiber composites for structural and infrastructure applications. In *Southern Region Engineering Conference* (pp. 1–5). University of Southern Queensland, Toowoomba, 11-12.
- Tokoro, R., Vu, D. M., Okubo, K., Tanaka, T., Fujii, T., & Fujiura, T. (2008). How to improve mechanical properties of polylactic acid with bamboo fibers. *Journal of Materials Science*, *43*(2), 775–787.
- Tran, L. Q. N., Minh, T. N., Fuentes, C. A., Chi, T. T., Van Vuure, A. W., & Verpoest, I. (2014). Investigation of microstructure and tensile properties of porous natural coir fibre for use in composite materials. *Industrial Crops and Products*, *65*, 437–445.
- Truong, M., Zhong, W., Boyko, S., & Alcock, M. (2009). A comparative study on natural fibre density measurement. *The Journal of The Textile Institute*, *100*(6), 525–529.
- Venkateshwaran, N., Elayaperumal, A., & Sathiya, G. K. (2012). Prediction of tensile properties of hybrid-natural fiber composites. *Composites Part B: Engineering*, *43*(2), 793–796.
- Verma, D., Gope, P. C., Shandilya, A., Gupta, A., & Maheshwari, M. K. (2013). Coir fibre reinforcement and application in polymer composites: A review. *Journal of Materials and Environmental Science*, *4*(2), 263–276.
- Wallenbergeger, F. T., & Weston, N. E. (2004). Science and technology. In F. T. Wallenbergeger & N. E. Weston (Eds.), *Natural Fibers, Plastics and Composites* (pp. 3–7). New York: Springer Science & Business Media.
- Yan, L., Chouw, N., Huang, L., & Kasal, B. (2016). Effect of alkali treatment on microstructure and mechanical properties of coir fibres, coir fibre reinforced-polymer composites and reinforced-cementitious composites. *Construction and Building Materials*, *112*, 168–182.
- Yan, L., Chouw, N., & Jayaraman, K. (2014). Flax fibre and its composites - A review. *Composites Part B: Engineering*, *56*, 296–317.
- Yousif, B. F., Shalwan, A., Chin, C. W., & Ming, K. C. (2012). Flexural properties of treated and untreated kenaf/epoxy composites. *Materials and Design*, *40*, 378–385.
- Yusoff, R. B., Takagi, H., & Nakagaito, A. N. (2016). Tensile and flexural properties of polylactic acid-based hybrid green composites reinforced by kenaf, bamboo and coir fibers. *Industrial Crops and Products*, *94*, 562–573.
- Zhang, J., Chaisombat, K., He, S., & Wang, C. H. (2012). Hybrid composite laminates reinforced with glass/carbon woven fabrics for lightweight load bearing structures. *Materials & Design*, *36*, 75–80.
- Zhang, Y., Li, Y., Ma, H., & Yu, T. (2013). Tensile and interfacial properties of unidirectional flax/glass fiber reinforced hybrid composites. *Composites Science and Technology*, *88*, 172–177.
- Zhu, X., Kim, B. J., Wang, Q. W., & Wu, Q. (2014). Recent advances in the sound insulation properties of bio-based materials. *BioResources*, *9*(1), 1–23.