Influence of Kenaf Core Fiber Incorporation on the Mechanical Performance and Dimensional Stability of Oil Palm Fiber Reinforced Poly(lactic acid) Hybrid Biocomposites

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This study demonstrated the reinforcing potential of kenaf core fiber (KCF) to complement and sustain oil palm fiber supply chain in the production of natural fiber-thermoplastic biocomposites. The lignin-rich KCF was incorporated into cellulose-rich oil palm empty fruit bunch fiber (EFBF)- and oil palm mesocarp fiber (OPMF)-poly(lactic acid) (PLA) composites, aimed at achieving synergism. The hybrid biocomposites developed by melt blending and subsequent compression molding were characterized for possible application as an alternative to medium-density fiberboards. The mechanical properties and dimensional stability of both single fiber- and hybrid fiber-PLA biocomposites were evaluated and compared. The test results showed a synergistic improvement as a consequence of fiber hybridization. Also, the findings suggested the best material performance with the incorporation of 5% KCF into 55% EFBF or OPMF and 40% PLA matrix. The OPMF-KCF-PLA hybrid biocomposites gave better results than the EFBF-KCF-PLA hybrid biocomposites.

Keywords: Hybrid biocomposite; Oil palm empty fruit bunch fiber; Oil palm mesocarp fiber; Kenaf core fiber; Poly(lactic acid)

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INTRODUCTION

Current research in the area of polymer composites has experienced increasing attention to the fabrication of green materials to curtail some of the problems associated with the production and use of traditional polymer composites, which are largely dependent on petroleum resources, for supply of both raw materials and energy (Kim *et al*. 2006; Mukherjee and Kee 2011; Dicker *et al*. 2014). In addition to the fact that some petroleum-based polymers have no biodegradability properties and exorbitant prices, petroleum resources are also non-renewable, quickly depleting, and threatening to run out in the next century (Bentley 2002; Hashim *et al*. 2012).

Conventional fiber reinforcement uses inorganic materials (*e.g.*, glass and carbon) as reinforcements with a petroleum-based or bio-polymer matrix (Zhang *et al*. 2012). However, conventional fiber reinforcement has shown quite a number of environmental, health, and economic issues (Abdul Khalil *et al*. 2012). These conventional fibers have been found to cause serious ecological and health hazards to employees working in polymer composite industries (Jawaid and Abdul Khalil 2011).

However, there is currently growing interest in natural fibers such as banana, pineapple, hemp, sisal, kenaf, oil palm, and bamboo, which have shown attractive advantages over conventional fiber reinforcements (Faruk *et al*. 2012; Then *et al*. 2014a). These benefits include natural abundance (Eichhorn *et al*. 2001), renewability (Mukherjee and Kee 2011), biodegradability (Kim *et al*. 2006), low cost (Leão *et al*. 2008), low energy requirements (Jawaid and Abdul Khalil 2011), low density (Hashim *et al*. 2012), attractive specific properties (Li 2004), good thermal and insulating properties (Khazaeian *et al*. 2015), lower wear and tear during processing (Nayak *et al*. 2009), relatively less abrasiveness, and sustainability (Abdul Khalil *et al*. 2010a, 2012).

A number of studies have been conducted to mix two or more natural and/or synthetic fibers together with biodegradable or non-degradable polymer matrices to develop hybrid fiber-polymer biocomposites (Jacob *et al*. 2004; Thiruchitrambalam *et al*. 2009; Jawaid *et al*. 2010; Pandita *et al*. 2014). The idea behind mixing two or more fibers is to achieve synergistic performance, complement material properties, or sustain the supply chain of raw material (Jawaid and Abdul Khalil 2011). The properties of weak fibers in the hybrid could be enhanced, thereby improving the overall material performance of the biocomposites. These properties include fiber orientation, length, and density. Moreover, the properties of hybrid biocomposites are related to individual fiber content, degree of fiber intermingling, and fiber-matrix interfacial bonding (Jawaid and Abdul Khalil 2011).

The combination of two or more materials often follows the rule of mixtures, which can best be used to explain the properties of natural fiber hybrid system. When the hybrid composites consist of only two different fibers, the rule of mixtures can be stated as $P_H = P_1V_1 + P_2V_2$ (Thwe and Liao 2003; Jawaid and Abdul Khalil 2011). The property of the entire mixture (P_H) relates proportionally to the corresponding properties of the first (P_1) and second (P_2) fiber components and also to their volume fractions, V_1 and V_2, respectively (Jawaid *et al*. 2012).

Some previous reports on hybrid natural fiber-synthetic fiber composites include epoxy resin-based glass-carbon fiber reinforced hybrid composites (Zhang *et al*. 2012), rooflite resin-based palmyra-glass (Velmurugan and Manikandan 2007), bamboo-glass fiber reinforced USP and VE resin (Mandal *et al*. 2010), jute-glass and kenaf-glass reinforced polyester (Akil *et al*. 2010), wood flour-glass thermoplastic composites (Valente *et al*. 2011), and oil palm empty fruit bunch-glass reinforced polypropylene hybrid composites (Rozman *et al*. 2001). There have also been reports on natural fiber-natural fiber reinforced hybrid biocomposites, including banana-kenaf reinforced polyester biocomposites (Thiruchitrambalam *et al*. 2009), oil palm empty fruit bunch fiber-jute reinforced epoxy resin hybrid biocomposites (Jawaid *et al*. 2010), and polypropylene-based kenaf fiber-wood flour hybrid biocomposites (Ghasemi *et al*. 2008).

Herein, novel natural fiber-natural fiber reinforced poly(lactic acid) (PLA) hybrid biocomposites were fabricated by a combination of EFBF-KCF and OPMF-KCF, and their

mechanical and dimensional stability properties were compared with their corresponding single fiber reinforced PLA based biocomposites. The primary goal of the present work was to enhance the mechanical performance of the single oil palm fiber-based PLA biocomposites by incorporation of KCF. To the best of our knowledge, this is the first attempt to incorporate KCF into oil palm fibers to form hybrid fiber reinforced PLA biocomposites.

The EFBF and OPMF (Fig. 1) are the biomass obtained, respectively, from fresh fruit bunch and oil palm fruits (Abdul Khalil *et al.* 2012). These two biomass types have distinct chemical properties (Table 1) from KCF (Fig. 2), which originates from the stem of the kenaf plant (Sreekala *et al*. 1997; Ververis *et al*. 2004; Deka *et al*. 2013).

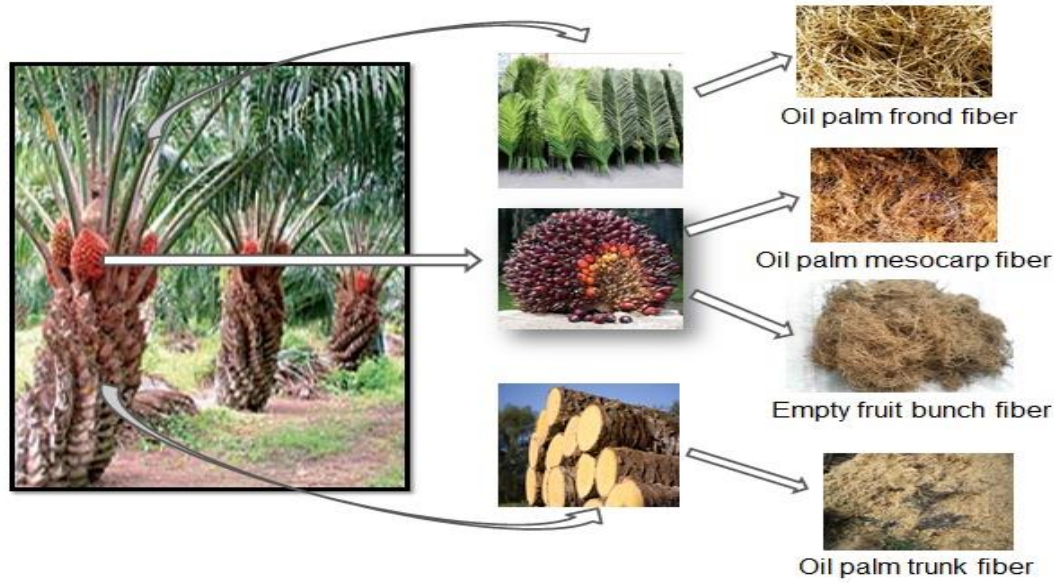

Fig. 1. Oil palm tree and biomass

The primary reason for mixing these hybrid fibers was that EFBF and OPMF are cellulose-rich, while KCF is lignin-rich (Table 1). Many studies have reported the reinforcing potential of lignin to enhance the mechanical and dimensional stability of natural fiber reinforced polymer biocomposites (Nasir *et al*. 2013; Ghaffar and Fan 2014; Thakur *et al*. 2014; Reza Barzegari *et al*. 2012); thus, incorporating KCF into the EFBF or OPMF may enable this presumed synergistic performance.

Table 1. Chemical Properties of Oil Palm Fibers and Kenaf Core Fibers

Fibers	Cellulose (%)	Holocellulose (%)	Lignin (%)	References
EFBF	62.9	82.4	17.8	(Law *et al.* 2007)
OPMF	60.0	-	11.0	(Sreekala *et al.* 1997)
KCF	31.0 to 33.0	87.2	23.0 to 27.0	(Aisyah *et al*. 2013)

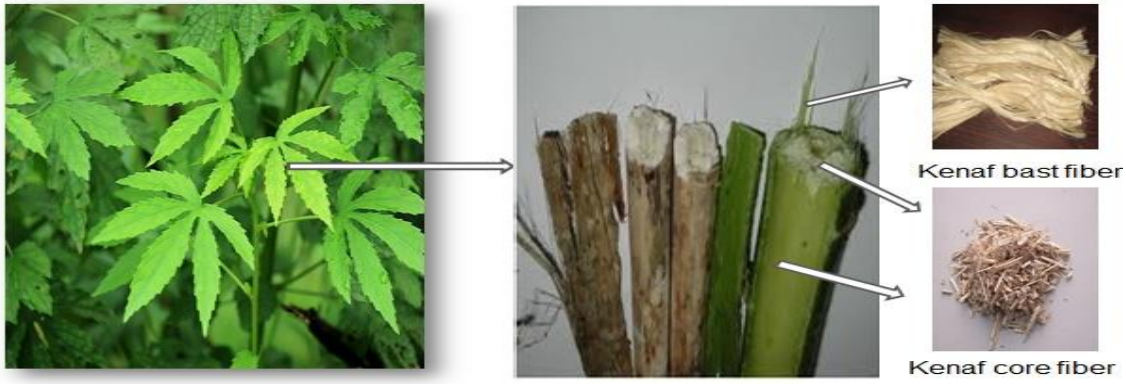

Fig. 2. Kenaf plant and fiber resources

EXPERIMENTAL

Materials

Poly(lactic acid) was purchased from Nature Work LLC (USA) under the trade name polylactide resin 4042D. It has a melting point range between 170 and 190 °C and density ranging from 1.4 to 1.5 g/cm^{-3}. The oil palm fibers, *i.e.*, EFBF and OPMF, were obtained from Sabutek (M) Sdn. Bhd., Malaysia and FELDA Serting Hilir Oil Palm Mill, Malaysia, respectively. KCF was kindly supplied by Lembaga Kenaf dan Tembakau, Malaysia.

Methods

Fiber purification

To remove impurities from the EFBF, OPMF, and KCF, they were physically treated by sorting, soaking in distilled water for 24 h at 25 °C, washing with heated water at 60 °C, cleaning with acetone, and oven-drying at 60 °C for 24 h. The oven-dried fibers were then ground into smaller particles, followed by sieving into fiber sizes ranging from 300 to 400 µm. The purified fibers were then stored in sealed plastic bags at 25 °C for further investigation.

Fabrication of single and hybrid fiber-PLA biocomposites

To prepare the single fiber-PLA biocomposites, the oven-dried EFBF, OPMF, and KCF together with the PLA were melt blended at various fiber-to-PLA loading formulations (Table 2) using a Brabender Internal Mixer (Germany) at 170 °C with a rotor speed of 50 rpm for 15 min, in accordance with the method reported by Then *et al*. (2015a). Initially, the PLA was loaded into the mixing chamber to melt for 2 min; then, the fibers were added to the molten PLA and mixing continued for 13 more minutes. The compounded single fiber-PLA biocomposites were further compression molded into sheets with dimensions of 1 mm × 150 mm × 150 mm and 3 mm × 150 mm × 150 mm (thickness × length × width). The compression molding was performed using a hydraulic hot press at 170 °C, 150 kgfm^{-2}, and 10 min molding temperature, pressure, and time, respectively. Furthermore, cooling was performed at 30 °C for 5 min.

The hybrid fiber-PLA biocomposites were prepared following a similar method. The hybrid fiber-PLA biocomposites were fabricated by randomly mixing two different fibers together manually prior to melt blending with PLA. The various hybrid fiber mixing formulations are presented in Table 3.

Table 2. Single Fiber-PLA Loading Formulations

Sample code	EFBF (%)	OPMF (%)	KCF (%)	PLA (%)
EF50	50	-	-	50
EF60	60	-	-	40
EF70	70	-	-	30
MF50	-	50	-	50
MF60	-	60	-	40
MF70	-	70	-	30
KF50	-	-	50	50
KF60	-	-	60	40
KF70	-	-	70	30

Table 3. Hybrid Fiber-PLA Loading Formulations

Sample code	EFBF (%)	OPMF (%)	Kenaf core (%)	PLA (%)
EF55KF5	55	-	5	40
EF50KF10	50	-	10	40
EF45KF15	45	-	15	40
MF55KF5	-	55	5	40
MF50KF10	-	50	10	40
MF45KF15	-	45	15	40

Mechanical properties analysis

The tensile properties of neat PLA and single and hybrid fiber-PLA biocomposites were tested using a Universal Testing Machine-Instron 3365 (USA) equipped with a 5-kN load cell at a 5-mm/min crosshead speed. The test was performed at 25 °C. Five dog-bone-shaped specimens were tested, as specified by ASTM D638-5 (2000). Average values of tensile strength, tensile modulus, and elongation at break are reported.

The flexural properties (*i.e.*, three-point testing) of the composites were determined using a Universal Testing Machine-Instron 3365 (USA) equipped with a 5-kN load cell at a 1.3-mm/min crosshead speed and 48-mm span length. The test was performed at 25 °C on five specimens with dimensions of 127.0 × 12.7 × 3.0 mm^3 (length × width × thickness) in accordance with ASTM D790 (2000). Average values of flexural strength and flexural modulus are reported.

The impact strength of the composites was examined following the un-notched IZOD impact test, as specified by ASTM D256 (2000). The impact tester (India) was equipped with a 7.5-J pendulum. The test was performed at 25 °C on five specimens having dimensions of 63.5 x 12.7 x 3.0 mm^3, and the average value is reported.

Density measurement

The density of the biocomposites was measured in accordance with BS EN 323 (1993) (European Committee for Standardization 1993; Abdul Khalil *et al.* 2010a). The masses of the test samples were obtained and recorded by weighing using an analytical

balance. Thereafter, the volume of the test samples was also obtained and noted by measuring their dimensions, *i.e.*, multiplying their length, width, and thickness, respectively. The density was subsequently calculated using the following formula:

$$\text{Density} = \text{mass/volume} \quad (1)$$

Dimensional stability measurement

To test for water absorption and thickness swelling of the biocomposites, test samples with dimensions of 10.0 × 10.0 × 1.0 mm^3 were cut according to ASTM D570 (2005) and European standard EN 317 (1993), respectively. The initial weight (W_1) and thickness (T_1) of the oven-dried test samples were measured and noted. Test samples were then immersed in distilled water for 24 h at 25 °C. Thereafter, they were removed, excess water was wiped off with a tissue, and their weight (W_2) and thickness (T_2) were measured again. Duplicate tests were conducted to determine the average, mean, and standard deviation. The water uptake and thickness swelling were calculated using the following equations:

$$\textit{Water Absorption}\ (\%) = \frac{W_2 - W_1}{W_1} \times 100 \quad (2)$$

$$\textit{Thickness Swelling}\ (\%) = \frac{T_2 - T_1}{T_1} \times 100 \quad (3)$$

Scanning electron microscopy (SEM)

The surface morphologies of the pure fibers (EFBF, OPMF, and KCF) and the fractured surfaces of neat PLA and the various fiber-PLA biocomposites were analyzed using scanning electron microscopy. The instrument used to conduct the test was a LEO 1455 VP scanning electron microscope (Japan) operated at 10-kV accelerating voltage. The metal holder of the instrument was used to hold the oven-dried samples in place. Thereafter, the samples were coated with gold for 3 min using a Bio-rad coating system (USA) to enhance conductivity before the commencement of analysis.

Fourier transform infrared spectroscopy (FTIR)

The chemical properties, *i.e.*, functional groups, bond types, and chemical components, of the pure oil palm fibers (EFB, OPMF, and KCF) were analyzed using a Perkin Elmer Spectrum 100 series spectrophotometer (USA). The instrument was equipped with attenuated total reflectance (ATR) capacity. The wavenumber range from 400 to 4000 cm^{-3} was employed to record the FTIR spectra.

RESULTS AND DISCUSSION

Single Fiber-PLA Biocomposites

Initially, water-treated EFBF, OPMF, and KCF at 90%, 80%, 70%, 60%, and 50% were melt-blended with 10%, 20%, 30%, 40%, and 50% poly(lactic acid) (PLA). This was done to develop single fiber-PLA biocomposites and investigate the effect of fiber loading

on the mechanical properties and dimensional stability of PLA to optimize the single fiber-PLA biocomposites as control samples. It was observed that the fibers at 90% and 80% loadings could not be wetted with 10% and 20% PLA, respectively, resulting in poor fiber-polymer bonding. This was in accordance with observations made by Then *et al*. (2013). Therefore, the 90% and 80% fiber loadings were removed from the research. The PLA can sufficiently wet the fibers at 30% PLA loading.

However, the TS dropped broadly at 70:30 fiber-to-PLA loading and presented comparable results for the EFBF and OPMF composites, while the KCF composite showed the least TS. The TS at 50:50 fiber-to-PLA loading showed reduced values relative to those at 60:40 fiber-to-PLA loading. This is a result of the high composition of PLA, which is relatively brittle and makes the material easily breakable (Brostow and Hagg 2010). Generally, The TS shows optimum results at a 60% fiber loading, with 40% PLA. The KCF-filled biocomposites had the lowest TS, which is expected considering that KCF fibers possess low quality for this sort of application because of the high contents of holocellulose and lignin and the low content of α-cellulose (Abdul Khalil and Suraya 2011). The fiber also has a low aspect ratio (*i.e.*, fiber length to fiber diameter).

Table 4. Mechanical Properties and Densities of Single Fiber-PLA Composites

Sample code	Tensile Strength (MPa)	Tensile Modulus (MPa)	Elongation (%)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Impact Strength (J/m)	Density (kgm^{-3})
PLA	64.27±4.99	464.19±20.55	18.33±1.35	38.67±1.89	2.98±0.32	29.16±1.13	1250
EF50	9.16±2.73	87.92±0.47	11.59±0.59	33.72±2.03	4.27±0.22	10.12±1.21	1030
EF60	10.19±1.97	98.92±1.05	9.26±0.39	33.92±2.41	4.88±0.33	9.98±1.03	1055
EF70	9.32±3.81	110.95±2.86	8.40±1.03	29.53±2.33	2.60±0.18	6.48±0.66	1108
MF50	10.99±1.28	107.85±1.42	10.19±0.90	32.53±1.93	2.68±0.20	7.97±0.70	1040
MF60	12.32±1.79	121.6±0.57	8.98±1.08	27.36±2.36	4.27±0.16	6.58±1.22	1062
MF70	10.92±1.53	145.97±1.59	8.44±0.52	23.32±1.43	3.12±0.22	6.25±1.19	1114
KF50	7.77±1.79	102.99±1.22	10.05±0.99	29.93±2.47	3.53±0.19	5.31±1.14	1045
KF60	10.34±0.99	117.01±0.95	6.64±1.05	17.35±1.74	1.96±0.13	4.66±1.05	1064
KF70	4.85±2.89	102.89±1.20	6.37±1.09	12.44±2.17	1.69±0.30	4.63±0.62	1130

OPMF had a better TS than did EFBF. This could be because the OPMF possesses a more soft and flexible nature and thus has a greater tendency to be more uniformly distributed and oriented in the PLA matrix, thereby bringing about higher TS and reinforcing ability than EFBF. This conforms to the findings observed by Then *et al*. (2013).

The elongation at break (EB) of the biocomposites decreased with low PLA loading and high fiber content, as shown in Table 4. This may be due to the poor interfacial bonding between the fiber and the PLA, producing micro cracks which can easily propagate in the composites, causing them to break (Okubo *et al*. 2005; Then *et al*. 2013). The decrease in EB with increasing lignocellulosic fiber loading in biocomposites has also been observed and reported elsewhere (Habibi *et al*. 2008).

The results for the tensile modulus (TM), which measures the stiffness of PLA biocomposites, are given in Table 4. The TM of neat PLA had the highest value, but upon

reinforcement with EFBF and OPMF, it declined and tended to rise with increased fiber loading which could be because of stiffness variation between the fibers and the PLA matrix. This finding was also observed by (El-Shekeil *et al*. 2012). Though, the TM for the KCF-PLA composites appeared to be low at 70% fiber loading, which can be ascribed to the poor interfacial bonding resulting from poor quality of KCF. The extent of fiber-polymer bonding capacity have been shown to influence the TM (Nor Azowa *et al*. 2010). Thus, in terms of TS, EB, and TM, the optimum fiber and PLA loadings were 60% and 40%, respectively.

The flexural strength (FS) and flexural modulus (FM) of single fiber-PLA biocomposites are presented in Table 4. The FS of neat PLA declined upon reinforcement, and the decrease tended to be lower with higher fiber loading and appeared to be smallest for EFBF, moderate for OPMF, and greatest for the KCF. This disparity could be due to the strength of the three fibers and poor fiber-PLA bonding as a result of the presence of hemicellulose and wax, which makes the fiber surface unclean and impermeable and does not allow strong adhesion to occur (Then *et al*. 2014a). The FM of the neat PLA was 2.98 GPa, which increased upon introduction of fibers. This increase could be clearly seen for EFBF (50% and 60%), OPMF (60%), and KCF (50%), respectively. A similar trend was also observed by previous researchers, who attributed the FM increase upon fiber incorporation to the relative fiber stiffness compared with the polymer matrix (Then *et al*. 2015a).

The results obtained in the un-notched IZOD impact strength test for neat PLA and EFBF-, OPMF-, and KCF-PLA biocomposites (Table 4) showed that the neat PLA had the highest impact strength (IS). The EFBF-PLA biocomposites recorded the highest IS amongst the fiber-PLA biocomposites, while the KCF-biocomposites had the least IS. This is expected, as the EFB fiber has the toughest fiber surface and higher cellulose content, enabling it to properly mix with the PLA matrix to form a stronger interfacial bond. The KCF, being weak because of its low cellulose content and high content of hydrophilic groups, such as holocellulose, could not be properly be wetted by the PLA matrix, consequently forming poor interfacial bonding, which results in low IS because of low resistance to crack propagation at the point of impact. The OPMF-PLA biocomposites showed moderate performance, which could be due to the softness of the fiber as compared with the EFBF and better fiber quality than the KCF-PLA biocomposites. Generally, the IS of all the fiber-PLA biocomposites decreased with increasing fiber loading. This could be because the PLA could not properly wet the fibers as the fiber loading increased, resulting in stress concentration because the PLA matrix was unable to effectively transfer stress to the fibers, thereby hampering the ability of the biocomposites to absorb impact energy (Tawakkal *et al*. 2012).

Density is one of the important parameters used for the assessment of natural fiber thermoplastic composites fabricated for use as alternative medium-density fiberboards (MDF). It has been reported that the density of MDF is between 496 to 801 kgm^{-3}; therefore, natural fiber- or wood-thermoplastic biocomposites with densities within this reported range can be used as alternatives to MDF (Rivela *et al*. 2007). The density of the composites is the mass per unit volume. Table 4 presents the results of the densities of neat PLA and EFBF-PLA, OPMF-PLA, and KCF-PLA biocomposites at various fiber loadings.

The results demonstrate that the density of the neat PLA composite declines upon incorporation of natural fiber reinforcements. The densities of the natural fiber-PLA

biocomposites increase with increasing fiber content, with the EFBF-PLA biocomposites having the lowest densities and the KCF-PLA biocomposites the highest densities. The decline in the density of the neat PLA biocomposite after reinforcement with the natural fibers could be due to the density variation between the PLA and the fibers. The PLA is denser than each of the individual fibers. It has been reported that the density of commercial PLA is between 1.25 and 1.28 gcm^{-3} (Drumright *et al*. 2000). The natural fibers have densities of 0.70 to 1.55 gcm^{-3} for oil palm fibers (Jawaid *et al*. 2015) and 0.21 gcm^{-3} for KCF (Abdul Khalil 2010b; Jawaid and Abdul Khalil 2011). These density variations could have a bearing on the overall densities of the natural fiber-PLA biocomposites.

Moreover, the increase in the density of the fiber-PLA biocomposites with increasing fiber loading could be due to the hydrophilic groups on the fiber surfaces, which are likely to attract moisture from the atmosphere and consequently add to the densities of the composites.

The neat PLA had low water uptake and thickness swelling, while the water uptake and thickness swelling for the single fiber-PLA biocomposites increased with increasing fiber loading (Fig. 3). This is in agreement with findings observed elsewhere (Tawakkal *et al*. 2012). The neat PLA absorbed less than 2% water, while the biocomposites with 70% fiber loading absorbed up to approximately 13% moisture. This is expected, as the fibers have only been treated with distilled water, which could not efficiently remove impurities and hemicellulose. Thus, an increase in the amount of the fiber will increase the amount of alcohol hydroxyl functional groups on the fiber surface. These hydroxyl groups are capable of attracting water from the atmosphere, thereby making the fibers and their corresponding composites more hydrophilic (Shinoj *et al*. 2011; Then *et al*. 2014b).

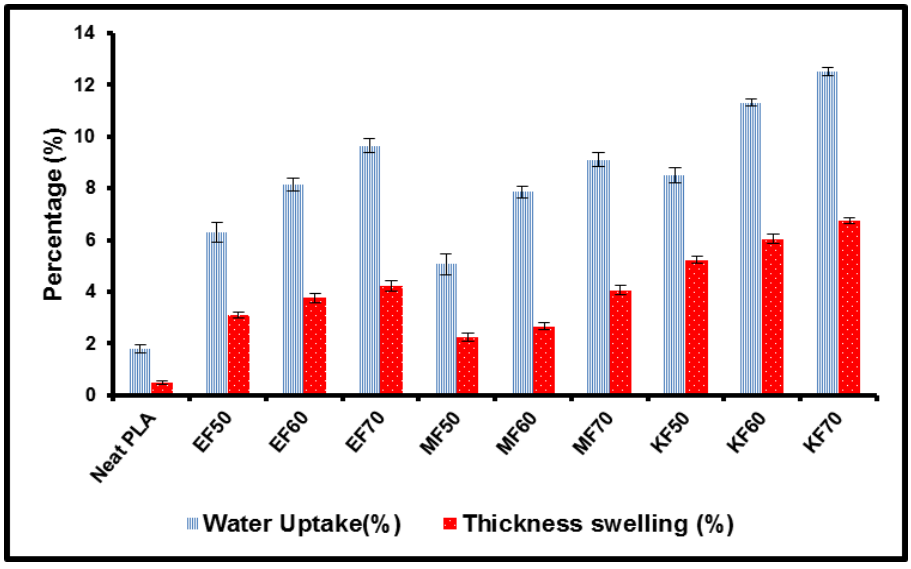

Fig. 3. Dimensional stability of single fiber-PLA composites

Because the natural fibers (EFBF, OPMF, and KCF) are hydrophilic, their internal bonding with the hydrophobic polymer will be very difficult, which consequently renders the biocomposites poor, weak, and brittle (Kalia *et al*. 2013).

However, KCF is known to contain more hydrophilic groups on its surface because of its higher content of holocellulose, which shows a greater tendency to absorb water

during immersion and swell (Fig. 3). The oil palm fibers showed lower water uptake and thickness swelling because they have better fiber surface quality, which, even though hydrophilic, could not attract moisture to the extent of the KCF. Generally, the water uptake appeared to be directly proportional to the thickness swelling for all the PLA composites.

Hybrid Fiber-PLA Biocomposites

The optimum sample formulation for the single fiber-PLA biocomposites based on the mechanical properties and density was a fiber to PLA weight ratio of 60:40. This formulation was used as a control to fabricate the hybrid fiber-PLA biocomposites. The sample formulations are presented in Table 3. The KCF was incorporated as a secondary fiber into the EFBF and OPMF composites.

The TS and TM (Figs. 4 and 5) showed positive hybrid effects, which could be a result of the incorporation of KCF into the oil palm fibers.

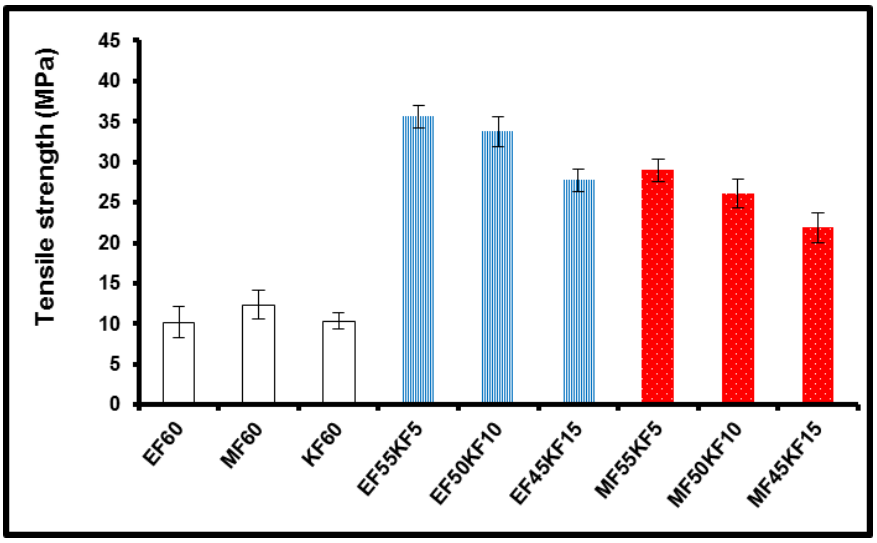

Fig. 4. Tensile strength of hybrid fiber-PLA composites

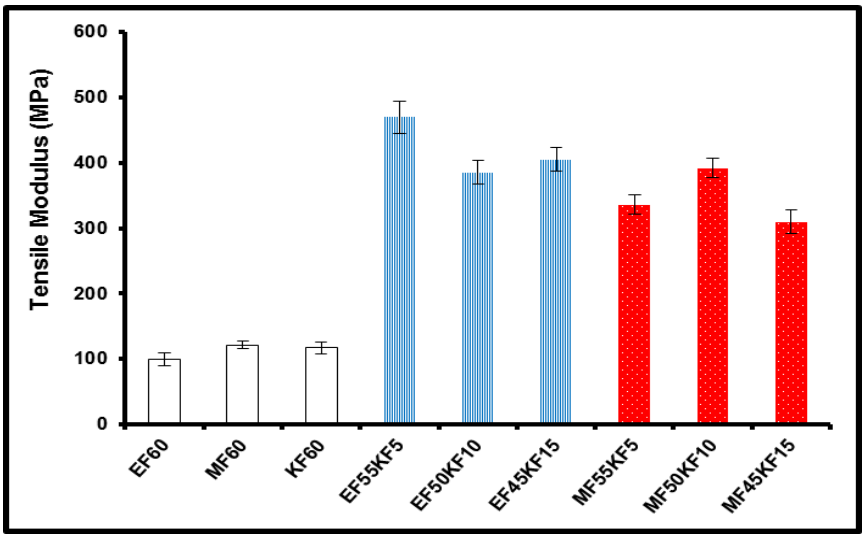

Fig. 5. Tensile modulus of hybrid fiber-PLA composites

The main reason for the improved TS and TM with KCF incorporation could be the rich lignin content of the KCF, which facilitated interfacial bonding between the oil palm fibers and the PLA matrix.

Lignin is made up of polar hydroxyl groups and non-polar hydrocarbons and hence could act as a compatibilizer between the hydrophilic fiber and hydrophobic polymer, thereby improving the mechanical properties of the biocomposites (Rozman *et al*. 2000). The decrease in TS and TM with increasing amount of KCF could be attributed to the formation of agglomerates and non-uniform dispersion of the KCF in the hybrid, thereby causing stress concentration in the glassy PLA. The stress concentration could generate crack formation and propagation along the interface, leading to mechanical failure. This decrease in the tensile properties of polymer composites as a result of the presence of lignin was also observed by Reza Barzegari *et al*. (2012).

The percentage EB (Fig. 6) of the EFBF- and OPMF-PLA composites did not show much disparity before and after the incorporation of KCF. The EB tends to be lower at high KCF, which may be due to micro crack generation as a result of KCF agglomerate formation. Similarly, at high KCF loadings, the amount of hydrophilic groups would be high and the propensity of the biocomposites to attract water molecules from the atmosphere would also increase. Thus, with the incorporation of more hydrophilic constituents in the composites, the fiber-PLA interfacial bonding would become poorer and weaker and the tendency to create micro defect that could easily cause failure during stress would increase, consequently lowering the EB.

With the addition of 5% KCF, the flexural strength (FS) dropped by approximately 34% and 2% for the EFBF- and OPMF-PLA, respectively (Fig. 7). This diminution in FS could be because of residual fiber surface impurities, and the increased fiber-fiber interaction due to KCF addition which disallowed sufficient wetting of the fiber by the PLA, hence preventing the even distribution of stress. Generally, the incorporation of KCF in the OPMF-PLA gave better flexural strength than the addition of KCF in the EFBF-PLA. OPMF has been previously reported to have better flexural strength than EFBF (Then *et al*. 2013).

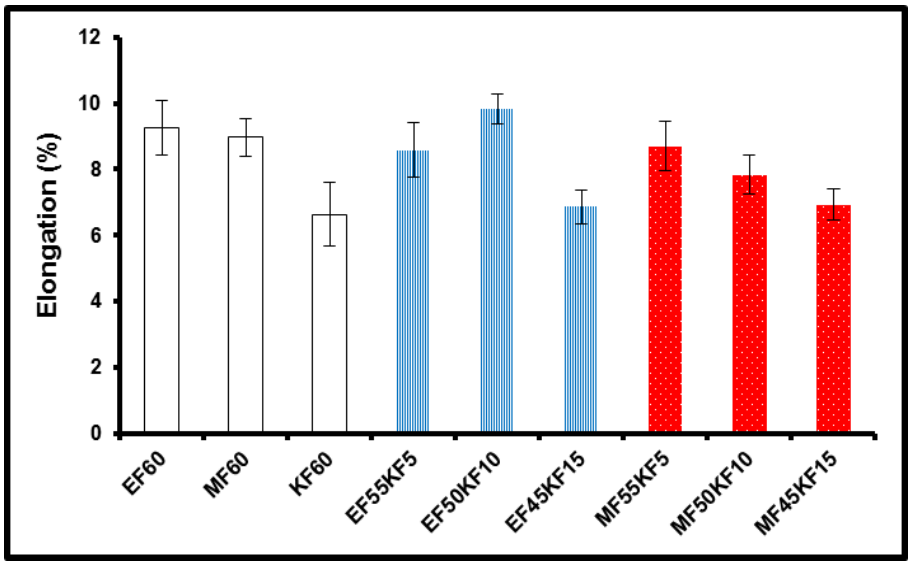

Fig. 6. Elongation at break of hybrid fiber-PLA composites

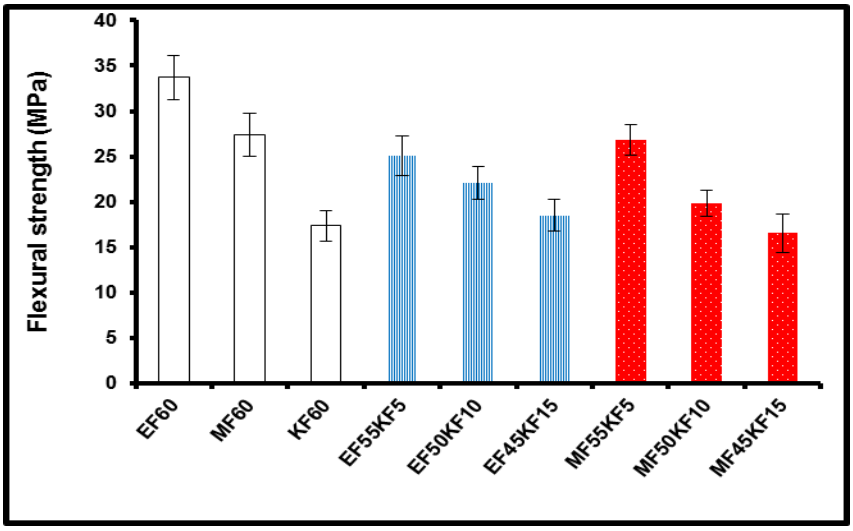

Fig. 7. Flexural strength of hybrid fiber-PLA composites

Similarly, increasing the KCF content in both EFBF-PLA and OPMF-PLA decreased the FS. Perhaps, this could be associated with the high content of hydrophilic groups in the KCF, which weaken the fiber-PLA interface, and also the possibility of agglomerate formation of the KCF as its content increased. Fiber agglomeration has been observed to weaken the interfacial region (Thwe and Liao 2003). These agglomerates formed because of uneven fiber distribution and are difficult to manually separate, leading to poor stress transfer between the oil palm fibers and the PLA. Previous researchers have also pointed out that kenaf fiber agglomerates are very difficult to separate manually (Zampaloni *et al*. 2007). In addition, the presence of these KCF agglomerates at the point of flexure during testing could easily cause the composites to break because of the poor quality of the KCF.

Conversely, the flexural modulus (FM) of the hybrid fiber-PLA biocomposites, presented in Fig. 8, indicates better improvement for the KCF-EFBF-PLA than the KCF-OPMF-PLA, particularly at 10% KCF content, at which an approximately 36% improvement compared with the EFBF-PLA biocomposites was seen. This could be associated with the fiber surface toughness of the EFBF. The fluctuation in the FM of the KCF-incorporated oil palm fiber-PLA composites could be a result of the uneven dispersion of the KCF, the heterogeneity of the fiber mixture, the weak fiber-PLA interface, and increased fiber-fiber contact of the two fibers in the biocomposites. The mechanical properties of hybrid fiber biocomposites has been reported to be related to the fiber orientation and the manner of the hybrid combination (Çöpür *et al*. 2008).

The impact strength (IS) of the biocomposites, presented in Fig. 9, showed improvements with KCF addition to the oil palm fibers. The optimum IS was observed with 5% KCF incorporated into both EFBF-PLA and OPMF-PLA biocomposites, which could be due to enhanced fiber-PLA bonding. Perhaps the KCF as a secondary fiber at 5% loading was able to be uniformly distributed in the oil palm fiber-PLA biocomposites, and the relatively higher lignin content in the KCF (hydrophilic as well as hydrophobic features) served as a bridge between the hydrophilic oil palm fibers and the hydrophobic PLA, enhancing their interface and consequently improving their IS.

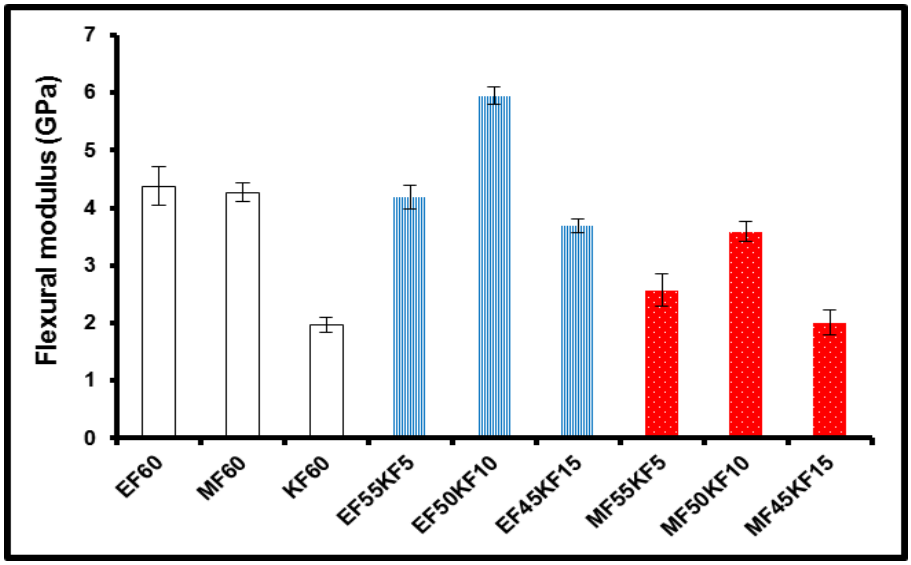

Fig. 8. Flexural modulus of hybrid fiber-PLA composites

Previous researchers also observed improvement in IS because of lignin, particularly when modification is done on the lignin to harness its optimum reinforcing abilities (Thakur *et al*. 2014). It can also be seen that the IS tended to decline with more KCF as a secondary fiber (Fig. 9). This may have occurred because of increased inter fiber interaction resulting in fiber breakage thereby restricting effective stress transfer between the fibers and PLA (Sreekala *et al.* 2002). KCF can also easily form fiber clumps at high concentrations, thereby making the biocomposites easily breakable under impact.

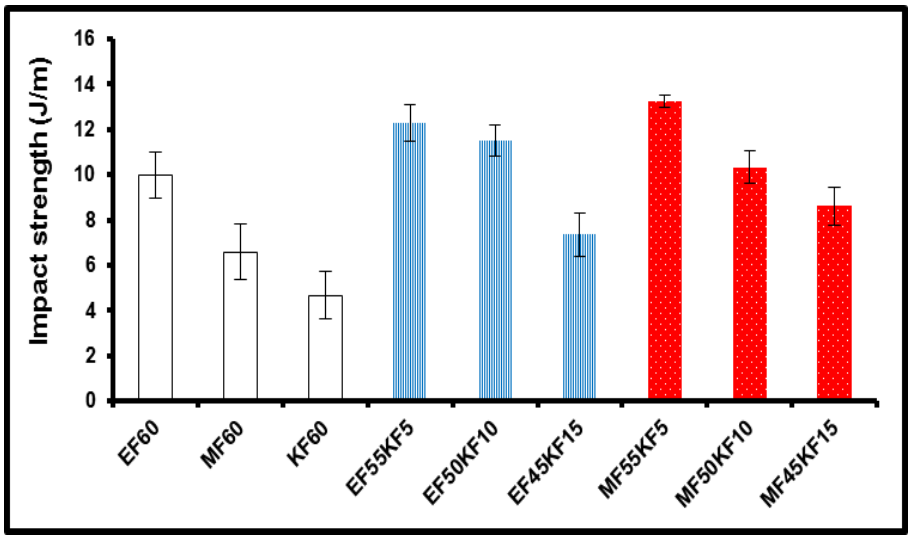

Fig. 9. Impact strength of the hybrid fiber-PLA composites

Interestingly, the densities of the oil palm fiber-PLA biocomposites decreased upon addition of 5% KCF as a secondary fiber, by approximately 5% and 3% for EFBF-PLA and OPMF-PLA, respectively. This decrease could be associated with the low density

profile of the KCF compared with the oil palm fibers: kenaf core fiber (0.21 gcm^{-3}) and oil palm fibers (0.70 to 1.55 gcm^{-3}), as reported in the literature (Jawaid and Abdul Khalil 2011; Abdul Khalil 2010b). Moreover, the density appeared to increase with increasing KCF loading. This could be due to the increased hydrophilicity of the biocomposites because of the high content of holocellulose in the KCF, which is capable of attracting water molecules from the atmosphere, hence increasing the density of the biocomposites (Khazaeian *et al*. 2015).

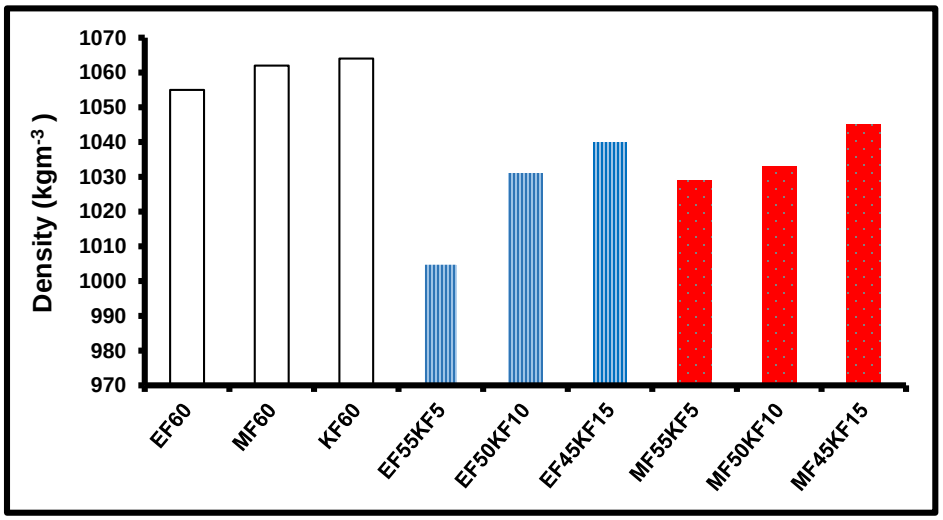

Fig. 10. Density of the hybrid fiber-PLA composites

The results for dimensional stability of the hybrid fiber-PLA biocomposites presented in Fig. 11 indicate that the water uptake and thickness swelling of the oil palm fiber-PLA biocomposites slightly increased with the introduction of 5% KCF.

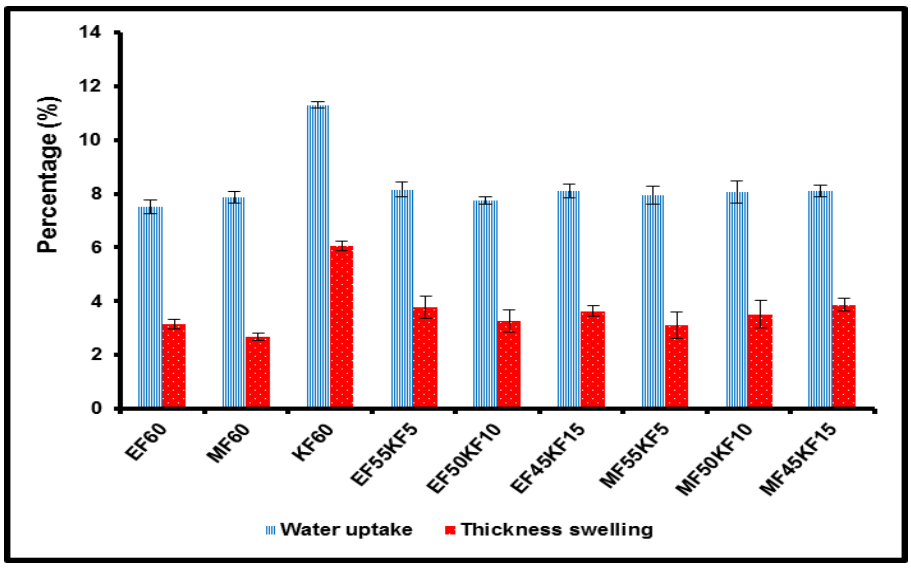

Fig. 11. Dimensional stability of hybrid fiber-PLA composites

The increase in the water uptake was 8.37% and 1.02%, while thickness swelling increased by 19.36% and 16%, for EFBF-PLA and OPMF-PLA biocomposites, respectively. However, both water uptake and thickness swelling of the hybrid fiber-PLA biocomposites decreased compared with those of the KCF-PLA biocomposites. The increase in water uptake and thickness swelling with the incorporation of 5% KCF as a secondary fiber into the oil palm fiber-filled PLA composites could be attributed to the high holocellulose content of the KCF. It can also be seen that increasing the amount of the KCF, particularly in the OPMF-PLA biocomposites, tended to increase the water uptake and thickness swelling of the hybrid fiber-PLA biocomposites.

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR was employed in order to study the various functional groups, chemical composition, and types of bonds. The spectra, showing different absorption bands for the three natural fibers, are presented in Fig. 12.

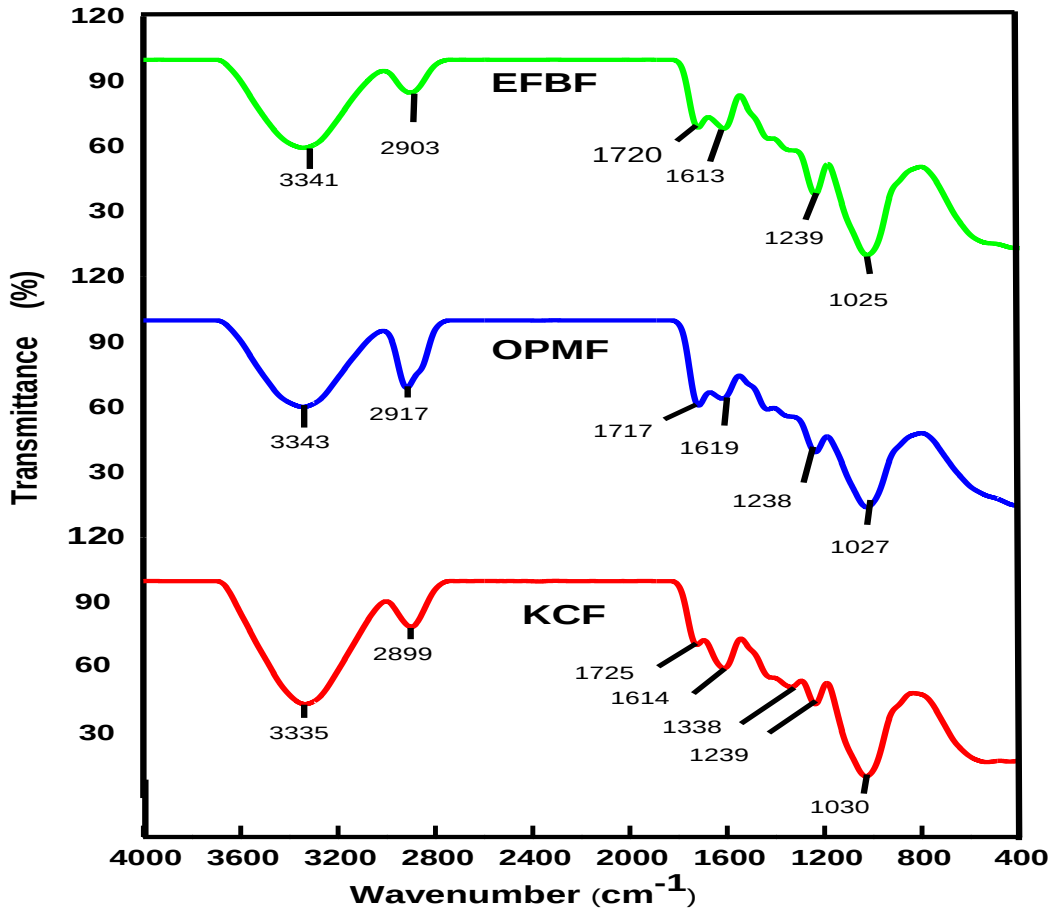

Fig. 12. Fourier transform infrared spectroscopy of pure fibers

The FTIR spectra showed broad peaks at 3335, 3343, and 3341 cm^{-1} corresponding to the O-H (hydroxyl) stretching of cellulose and hemicellulose in the KCF, OPMF, and EFBF, respectively. The peak intensity was slightly broader for OPMF and EFBF compared to KCF, which implies variation in cellulose content among the fibers. The absorption bands that appeared at 2899, 2917, and 2903 cm^{-1} represent the C-H stretching of cellulose and hemicellulose in KCF, OPMF, and EFBF, respectively. This concurred with previous findings by Sgriccia *et al*. (2008) and Then *et al*. (2015a).

The peak corresponding to C=O stretching of carbonyl groups in hemicellulose or lignin compounds of the three fibers appeared at 1725, 1717, and 1720 cm^{-1} for the KCF, OPMF, and EFBF, respectively. Clearly, the absorption band for the C=O stretching was somewhat broader for KCF, which can be ascribed to its high lignin and hemicellulose content relative to OPMF and EFBF. The absorption bands at 1614, 1619, and 1613 cm^{-1} represent the C=C stretching of aromatic rings in lignin (Then *et al*. 2015a). This band range can also be attributed to vibrational stretching of unconjugated C=O groups in hemicellulose, as well as conjugated carbonyl groups existing in lignin (Merlini *et al*. 2011).

The absorption peaks seen at 1239, 1238, and 1239 cm^{-1} in KCF, OPMF, and EFBF, respectively, represent C-O stretching of alcohols, ethers, and phenols in waxy residues as well as the acetyl groups in lignin. This conforms with observations by Liu *et al*. (2004), Sgriccia *et al*. (2008), and Merlini *et al*. (2011).

However, a band at 1338 cm^{-1} was noted only in the spectra of KCF. This band can be ascribed to the C=C bond stretching of benzene present in lignin. A similar finding was obtained by Liu and Wang (2009). Also, its appearance only in the spectra of KCF implies a high lignin content compared with the OPMF and EFBF.

The absorption peaks at 1030, 1027, and 1025 cm^{-1} in the spectra of KCF, OPMF, and EFBF, respectively, can be ascribed to C-H and C-O stretching vibrations (Merlini *et al*. 2011). The C-O vibration within this region of the absorption band was previously described by Maizatul *et al*. (2013) to be associated with aliphatic and aromatic primary alcohols present in cellulose, hemicellulose, and lignin.

Scanning Electron Microscopy (SEM)

The morphologies of the natural fibers, *i.e.*, pure EFBF, pure OPMF, and pure KCF, as well as the fractured surfaces of neat PLA, single fiber-PLA biocomposites, and hybrid fiber-PLA biocomposites, are presented in Fig. 13.

The pure EFBF (Fig. 13a) appeared smoothly intact, with few residual impurities, as also observed by Then *et al*. (2013). Residual impurities and few pores were noted on the OPMF (Fig. 13b); perhaps the pores appeared because of the removal of impurities, as previously observed by Nordin *et al*. (2013) and Then *et al*. (2014 a,b). The KCF (Fig. 13c) looked roughly disfigured, which could be ascribed to its high hydrophilicity. Previous works have reported similar features and described them as pit structures, considered the weak point that decreases the fiber strength (Abdul Khalil *et al*. 2010b).

The brittle behavior of the neat PLA (Fig. 13d) made it present a rough surface, although it was smoother than those of its biocomposites. The roughness of neat PLA has been reported elsewhere (Alam *et al*. 2014). The single oil palm fiber-PLA biocomposites (Fig. 13e through 13f) revealed fiber pullout, gaps, voids, and micro cracks, suggesting poor fiber-PLA bonding.

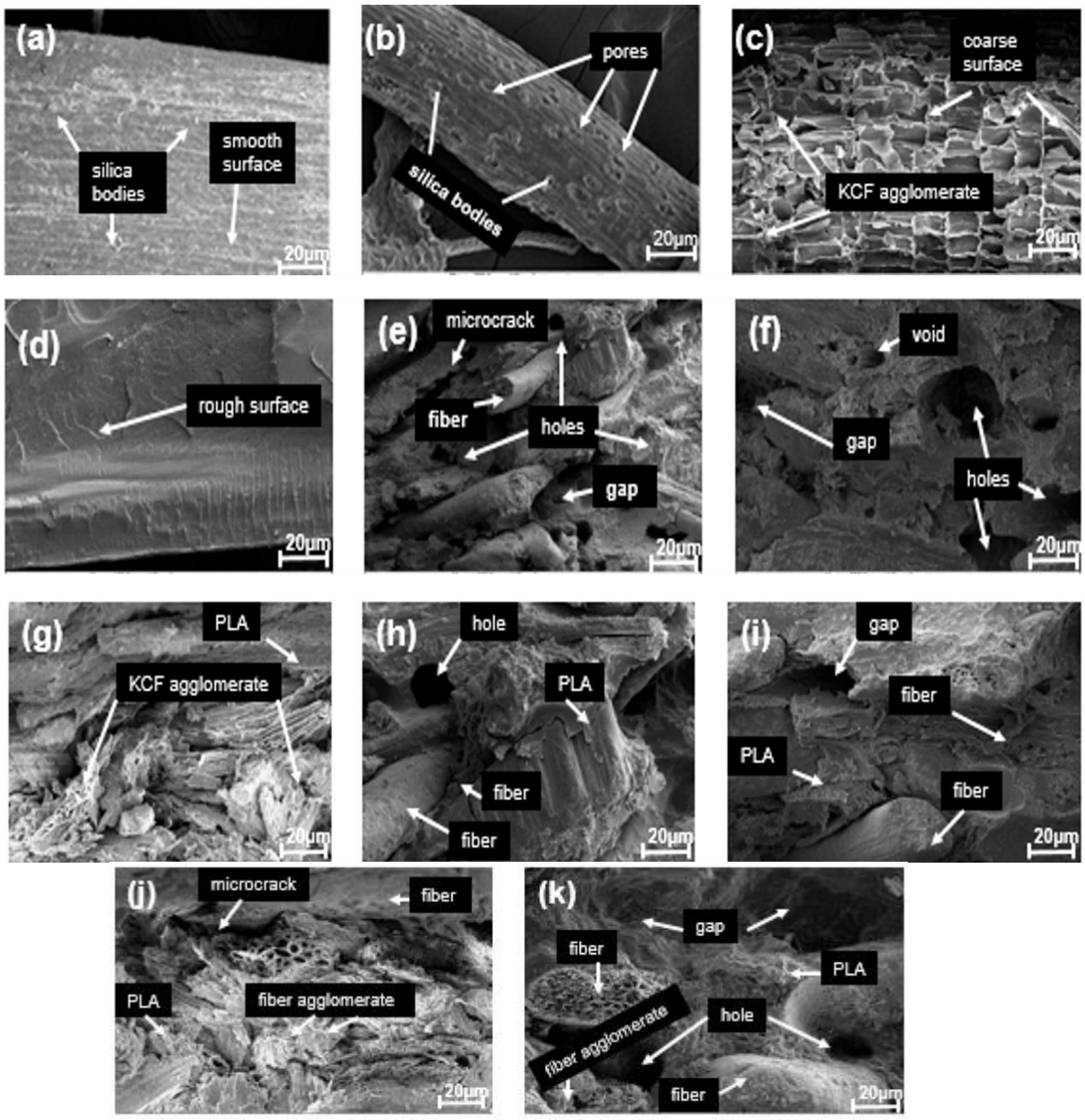

Fig. 13. SEM micrographs of (a) EFBF, (b) OPMF, (c) KCF, (d) neat PLA, (e) EFBF-PLA, (f) OPMF-PLA, (g) KCF-PLA, (h) EFBF/KCF(55%:5%)-PLA, (i) OPMF/KCF(55%:5%)-PLA, (j) EFBF/KCF(50%:10%)-PLA, and (k) OPMF/KCF(45%:15%)-PLA

The KCF-PLA biocomposite (Fig.13g) showed gaps as well as fiber clumps, which perhaps contributed to its poor mechanical behavior and dimensional stability. Moreover, the micrographs of the hybrid fiber-PLA biocomposites (Fig. 13h through 13i) showed that the fiber pullout, fibers protruding from the surface, and holes were relatively reduced with 5% KCF, implying an improved fiber-PLA interface because of fiber hybridization. A similar observation was reported by Jawaid *et al*. (2013). On the other hand, KCF clumps, voids, gaps, and micro cracks can be seen (Fig. 13j through 13k) at higher KCF loadings (*i.e.*, 10 and 15%), which translates to poor mechanical behavior and dimensional stability for these hybrid fiber-PLA biocomposites.

Generally, the micrographs of both the single fiber-PLA biocomposites and the hybrid fiber-PLA biocomposites (Fig. 13) showed weak features (*e.g.* gaps and voids), which is reasonable considering the fact that the fibers were treated with distilled water only, which may not effectively eliminate impurities from the fiber surfaces in order to enable stronger interface bonding to occur. Further fiber treatment methods such as alkalization as reported by (Manikandan *et al.* 2012; Fiore *et al.* 2014; Then *et al.* 2015b) can be employed to enhance the fiber-polymer interfacial adhesion.

CONCLUSIONS

1. KCF was successfully incorporated into EFBF-PLA and OPMF-PLA biocomposites, yielding improved mechanical properties.
2. A synergistic performance was observed because of the hybridization of lignin-rich KCF and cellulose-rich EFBF and OPMF.
3. Improved mechanical properties (*i.e.*, tensile and flexural strengths), impact strength, and density of the hybridized fiber-PLA biocomposites were obtained with the addition of 5% KCF to EFBF-PLA and OPMF-PLA biocomposites.
4. The dimensional stability was considerably better at 5% KCF than at higher loadings because of its high hydrophilicity.
5. This study revealed that KCF incorporation into oil palm-based fibers could offer synergism, complement the material performance of KCF, and sustain the supply chain of oil palm fibers.

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