Physico-Mechanical, Chemical Composition, Thermal Degradation and Crystallinity of Oil Palm Empty Fruit Bunch, Kenaf and Polypropylene Fibres: A Comparatives Study

(Fiziko-Mekanikal, Komposisi Kimia, Degradasi Haba dan Habluran Serabut Tandan Kosong Kelapa Sawit, Kenaf dan Polipropilena: Kajian Perbandingan)

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ABSTRACT

The physico-mechanical and chemical properties of enzyme retting kenaf and shredded empty fruit bunch of oil palm fibres (EFB) were analyzed by chemical extraction, microscopic, spectroscopic, thermal and X-ray diffraction method. Polypropylene (PP), a petroleum based fibre, was also included to compare the properties of synthetic fibre with natural fibres. Chemical extraction analysis showed that cellulose was the major component in both kenaf and EFB fibres which are 54% and 41.34%, respectively. Silica content of EFB was 5.29% higher than kenaf that was 2.21%. The result of thermogravimetric analysis showed that kenaf has higher thermal decomposition rate compared to EFB fibre. However, the residue for EFB fibre was higher than kenaf due to higher content in inorganic materials. The residual content of PP fibre was only 1.13% which was lower than the natural fibre. The diameter of EFB fibre bundle was 341.7 μm that was three times higher than kenaf. Microscopy study demonstrated that EFB surface was rough, porous and embedded with silica while kenaf showed smooth surface with small pith. Higher porosity in EFB was due to the lower fibre density that was 1.5 kg/cm³ compared to kenaf that was 1.62 kg/cm³. Kenaf has illustrated significant higher tensile strength (426.4 MPa) than EFB (150 MPa) and this result is in parallel to the pattern of the crystalline value for both fibres, 65% and 50.58%, respectively.

Keyword: EFB statistic; kenaf statistic; polypropylene; tensile strength; thermal analysis

ABSTRAK

Sifat fiziko-mekanikal dan kimia kenaf yang diretan melalui enzim dan serabut tandan kosong buah kelapa sawit (EFB) yang dipecahkan telah dianalisis melalui kaedah pengekstrakan kimia, mikroskopik, spektroskopik, haba dan pembelauan sinar-X. Kajian ini juga termasuk serabut berasaskan petroleum, polipropilena (PP) untuk membandingkan sifat serabut asli dan tiruan. Analisis pengekstrakan kimia membuktikan selulosa merupakan komponen utama dalam kedua-dua serabut kenaf dan EFB iaitu masing-masing 54% dan 41.34%. Kandungan silika EFB adalah 5.29% lebih tinggi daripada kenaf itu 2.21%. Keputusan themogravimetrik menunjukkan kenaf mempunyai kadar degradasi haba yang lebih tinggi berbanding dengan EFB. Walau bagaimanapun, sisa EFB adalah lebih tinggi daripada kenaf kerana kandungan bahan-bahan bukan organik yang lebih tinggi. Kandungan sisa serat PP hanya 1.13% iaitu lebih rendah daripada serabut semula jadi. Diameter berkas serabut EFB adalah 341.7 μm iaitu tiga kali lebih tinggi daripada serabut kenaf. Kajian mikroskopoik menunjukkan bahawa permukaan EFB adalah kasar, poros dan terkandung dengan silika manakala kenaf menunjukkan permukaan yang licin dengan saiz empulur yang kecil. Keliangan yang lebih tinggi dalam EFB adalah berkaitan dengan ketumpatan serat yang lebih rendah iaitu 1.5 kg/cm³ berbanding kenaf iaitu 1.62 kg/cm³. Kenaf menunjukkan kekuatan tegangan yang lebih tinggi yang ketara (426.4 MPa) daripada EFB (150 MPa) yang selari dengan corak nilai kristal untuk kedua-dua serat, masing-masing 65% dan 50.58%.

Kata kunci: Analisis termal; kekuatan tegangan; polipropilena; statistik EFB; statistik kenaf

INTRODUCTION

Over the last few years, the interest of academics and industrial communities in exploring the potential of natural fibre as reinforcement material in polymer composite have increased. The trend towards the application of the natural fibre in composite is due to the concern in sustainable and environmental issue on existing material, glass fibre. It is also aided by the unique features of natural fibre, namely high specific strength and modulus, large elongation at failure and low cost compared to glass fibre (Dicker et al. 2014; Han et al. 2012; John & Thomas 2007). Commonly, the contributing factor in natural fibre selection is related to

its geographical availability. For example, flax fibre in Europe whereas jute, ramie, kenaf and sisal have been of greater interest in Asia (Pickering et al. 2016). Whilst, oil palm fibre is abundantly available in Malaysia and Indonesia, accounting up to 80% of total world production of global palm oil industry (Mukherjee & Savacool 2014).

The oil palm industry is an important economy generator in Malaysia which contributed almost RM50 billion to Gross Net Income (GNI) of the country (Ahmad 2012). The oil palm industry also generates huge quantity of residues estimated at 80 million dry tonne annually (Agensi Inovasi Malaysia, 2012). Oil palm tree (*Elaeis guineensis*) is a palm type of plant belongs to the Palmea family originated from the coastal strip of Africa. In the processing plant, the fresh fruit bunches (FFB) are sterilized by steam then stripped to separate the fruit from the stalk. The fruits are then delivered to digesters and pressers for the crude oil extraction while the empty stalks (EFB) are collected for other purposes (Ibrahim et al. 2015). EFB contains cellulose (50.9%), hemicellulose (29.6%), lignin (17.84%), ash (3.4%) and extractive (3.21%) (Abdul Khalil et al. 2008). Typically the EFB moisture content is over 60% on a wet basis and the incineration of EFB without drying created 'white smoke' which unfavorable for surrounding (Yusoff 2006). As a result, there are still high amount of residues remain untapped and disposed. Roughly 4 kg of dry biomass are produced from each kg of palm oil extracted and it is estimated to produce 22 to 23 million tonnes of EFB annually (Abdullah & Sulaiman 2013).

Kenaf (*Hibiscus cannabinus*) in Malaysia was first introduced in early 1970s and was highlighted in late 1999 by the National Economic Action Council (NEAC) (Mohd et al. 2014). It is a warm-season annual fibre crop related to cotton (Abdul Khalil et al. 2010) which constitutes of 75-60% of core and 25-40% of bast. It is harvested within 3 to 6 months with the height between 2.7-3.6 m and a base diameter of 3-5 cm. The composition of kenaf is resulted by many factors, including cultivar, photosensitivity, duration of growing season, plant density and plant maturity (Akil et al. 2011). Mechanical properties of kenaf fibre are similar to jute fibre, but kenaf fibre is stronger, whiter and more lustrous (Zhang 2003). As a fibre crop, kenaf stem must be separated into bast and core by retting or mechanical extraction process (Yu & Yu 2007). Unlike the other non-wood plant, characteristics of kenaf fibres are similar to wood. It contains cellulose (44-57%), hemicellulose (22-23%), lignin (15-19%) ash (2-5%) and other elements (~6%). Presently, kenaf researches focus on production of bio-composite materials (Akil et al. 2011; Asumani et al. 2012; Bakar et al. 2006; Blezki & Gassan 1999) and bio-products (Gan et al 2014; Zakaria et al 2014).

Polypropylene (PP) is a thermoplastic polymer made by polymerizing of propylene molecules. PP fibres are widely used in nonwovens industry and occupy a share of about 63% of all fibres used for nonwoven production in the world (Das et al. 2012). The filaments are produced from polymer by melt-spinning and used with all nonwoven technologies for applications such as geo-textiles, hygiene products, air purification systems and automotive parts. The crystallinity of PP ranges between 40 and 60% with isotactic PP has a melting point ranging from 160°C to 166°C (Shubhra et al. 2011). PP possesses several useful properties namely high heat distortion temperature, transparency, flame resistance and dimensional stability which widen its application. As a matrix material, PP is widely used because it has some excellent characters for composite fabrication. PP is also very suitable for filling, reinforcing and blending. PP with natural fibrous polymers is one of the most promising routes to create natural-synthetic polymer composites (Shubhra et al. 2011).

Sufficient supply of fibre is one of important factor for industry to consider using it in industrial application. Also, understanding fibre's physical-mechanical and chemical properties are significant to determine its suitability for targeted product application; aiding in industrial processing and subsequently increasing the potential use of fibres in value-added products. In order to explore the vast economic potential of EFB and kenaf biomass, properties and statistical data of its availability is required. The objective of this study was to generate data set on kenaf and EFB fibre morphology, chemical composition, thermal, mechanical properties and to establish latest 5 years availability of kenaf and EFB in Malaysia; a key for commercial consideration. Also, PP fibre properties are being compared to explore potential application of natural fibre's combination with PP in polymer composite.

MATERIALS AND METHODS

Kenaf fibre from 4 months old plant were obtained from Dynamic Agrofarm Sdn Bhd in a ribbon form with length ranges between 2 and 3 meters. The fibre were cleaned with water to remove dust and contamination before cut into 60 mm length. EFB fibre were purchased from Ecofibre Sdn Bhd. The length of EFB fibre were in the range between 30 and 180 mm. PP fibre were bought from Hua long Chemical Fibre Co. Ltd, De Zhou, Shan Dong, China. The length of PP fibre were 65 mm with 10 denier diameter. All chemicals such as Sodium Chlorite, Ethanol and acetic acid were purchased from Merck Chemical Malaysia and were used without further purification.

STATISTICAL DATA

Statistical data of oil palm biomass were calculated based on the estimated generation rate of the residues against the amount of FFB processed by palm oil mills. The generation rates were established by the average rate of several references. The data on the plantation area and the amount of FFB harvested yearly between 2011-2015 were obtained from Malaysia Palm Oil Board (MPOB) website. The latest statistical data for kenaf were obtained from Ministry of Plantation Industries and Commodities (MPIC) website.

CHEMICAL CHARACTERIZATION

Kenaf bast and EFB fibre were air dried under the shade for 3 days with the relative humidity of the area between 60-70%. The dried samples were grinded in IKA pulveriser machine and sieved shake. Only fibre that passed the BS40 mesh sieve and retained on BS60 mesh sieve were collected for further applications. Moisture content was determined following Malaysian Standard MS544:2001 Method for Determination of Moisture Content of Timber using oven dry method where fibre (EFB and kenaf) were placed in oven at 105°C for 24 h.

Chemicals composition of the natural fibres was analysed. Extractive-free particle was derived by 6 h Soxhlet extraction with ethanol-toluene based on T 204 cm-97 Solvent extractives of wood and pulp. Holocellulose content was determined by treating the extractive free particle with sodium chlorite ($NaClO_2$) and acetic acid solution (Wise & Murphy 1946). The α-cellulose content of fibre was determined by further treating of holocellulose with 17.5% of NaOH and acetic acid to remove hemicelluloses according to TAPPI Standard T203 om-93. The content of hemicellulose was obtained by calculating the difference between holocelluloses and α-cellulose. Lignin content was determined by treating the extractive free particle with 72% sulphuric acid in accordance to TAPPI standard T222 om-06 Acid-insoluble lignin in wood and pulp. The samples were analysed in three replicate and the results were reported as the mean value for each set of measurements. Extractive derived from the Soxhlet extraction then dried by removing ethanol-toluene using rotary evaporator. Cold and hot water solubility were run based on TAPPI Standards T 207 os-25 while the ash content was run accordance to TAPPI Standard T 15 os-58.

FOURIER TRANSFORM INFRARED (FT-IR) SPECTROSCOPY

Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR) (Perkin Elmer Spectrum 400 FT-IR) was employed to obtain the spectra of each sample kenaf, EFB and PP. FT-IR spectra of each sample were recorded in the range of 4000-400 cm^{-1}, with a resolution of 4 cm^{-1} and averaged over 32 scans.

FIBRE MORPHOLOGY

All fibres were observed in bright field on Olympus Stereomicroscope Model SZX10 equipped with a DFC350 FX fluorescence camera (Leica Microsystems Inc., Wetzlar, Germany). Olympus software was used to measure the diameter of the fibres. Microscopic appearance of fibre surface and cross sections were obtained with a scanning electron microscopy Hitachi T1000. Prior to the SEM evaluation, the sample were coated with gold with plasma sputtering apparatus to increase fibre conduction.

The density of all fibre were determined by Pycnometer method with Archimedes theory DIN EN ISO 18753:2006-01 where 2 g of fibre were mixed with 20 mL of water for 2 h. Vacuum machine ZK-1 S Tianjin Zhonghuan was used to remove the air bubbles during the experiment.

THERMAL PROPERTIES

Thermogravimetry (TG) technique was employed to analyse the thermal properties of fibres. Scans were carried out at a heating rate of 10°C/min in nitrogen atmosphere with a gas flow of 20 mL/min from 30°C to 600°C. The measurements were performed using a SHIMADZU TGA-50 thermo analyser (SHIMAZDU Inc., Tokyo, Japan). All samples are pre-conditioned at 22±1°C and a relative humidity of 49±3% for 24 h.

Differential scanning calorimetric (DSC) measurements were performed on a SHIMADZU DSC-60 (SHIMAZDU Inc., Tokyo, Japan) at a heating rate of 5°C/min in nitrogen atmosphere with a gas flow of 40 mL/min. Each thermogram was recorded from 30 to 600°C.

CRYSTALLINITY

The degree of crystallinity of the fibres was determined using the X-ray diffractometry (XRD) technique with machine Bruker AXS, United States Model D8 Discover. The analysis was performed on the sample using radiation of Cu Kα = 1.5458Å in a diffraction angle (2θ) range of 5 to 60° with a step size of 0.0250°. The crystallinity index was calculated based on reflected intensity data according to (1) Segal formula (Segal et al. 1959).

$$\text{Cristallinity index (CrI)} = \frac{((\text{Intensity}_{002}-\text{Intensity}_{AM})}{(\text{Intensity}_{002}))} \times 100 \quad (1)$$

MECHANICAL PROPERTIES

Tensile tests of the fibres were performed in ambient condition using Electronic Single Fibre Strength Tester Shimadzu GX1000 20N Load Cell (Japan) according to ASTM (D 3822-01) test methods. The test was conducted with 20 replicates. The gauge length and the cross-head speed were set at 20 mm and 2 mm/min, respectively.

RESULTS AND DISCUSSION

STATISTIC OF EFB AND KENAF IN MALAYSIA

Table 1 shows the statistical data of oil palm plantation and biomass produced in 2011 until 2015. The total plantation area of oil palm in Malaysia increased from 5 million hectares in 2011 to 5.64 million hectares in 2015 (MPOB 2016). The production of fresh fruits bunch (FFB) has increased from 98 million tonne in 2011 to 104 million tonne in 2015 which is linear with the increment of planting area. The process of crude palm oil from FFB generates huge quantity of biomass residue such as EFB, fibre, kernel

TABLE 1. Statistics of Malaysian oil palm plantation and products 2011-2015

	2011	2012	2013	2014	2015	Generation rate (%)	Source
Total plantation (Million hectare)	5.00	5.08	5.23	5.39	5.64		MPOB
Export revenue (RM Billion)	80.4	71.40	61.36	63.62	60.17		MPOB
Total FFB (Million tonne)	98	96	99.5	100	104		MPOB
Palm oil (Million tonne)	20.58	20.16	20.89	21	21.84	21 (Husain et al. 2007)	
Mesocarp Fibre (Million tonne)	12.74	12.48	12.94	13	13.52	13 (Ahmad 2012)	
Kernel Shell (Million tonne)	5.29	5.18	5.37	5.4	5.62	5.4 (Ahmad 2012)	
EFB (Million tonne)	17.64	17.28	17.91	18	18.72	18 (Ahmad 2012)	

shell and palm oil mill effluent (POME) as shown in Table 1. The generation rate of oil palm biomass is estimated based on method by Ahmad (2012) and Husain et al. (2002). The statistics shows that the generation of the biomass increases every year in line with the increase of FFB production. Among all of oil palm biomass, EFB fibre is one of the most utilised residues in the market. It can be utilised to produce biocomposite, bioethanol, pulp and paper and many other value-added products (Mohd et al. 2014; Zakaria et al. 2014). Over the last five years, the amount of EFB generated every year increased from at 17.64 million to 18.72 million tonne. This is also attributed by the increased of amount of FFB processed. Mesocarp fibre is by-product of oil palm fruits after extraction process. It can be used as fuel for steam boiler due to its porous nature (Ahmad et al. 2012). The mesocarp fibre average generation rate estimated in this study is 13% from production of FFB. The results showed the generation amount of the fibre ranged between 12.74 and 13.52 tonne in 2011 to 2015. Oil palm kernel shell (PKS) is the smallest generation rate of biomass from oil palm at 5.4%. It has a complex pore structure and fibre matrix, which could be used as solid biofuel to replace petroleum based fuels (Ahmad et al. 2012). This is also the lowest oil palm biomass generated from FFB which at 5.62 million tonne recorded in 2015.

Table 2 shows the statistical data of kenaf plantation in Malaysia from 2011 to 2015. The plantation area for kenaf within 5 years has gradually increased by double from 1,140 hectares in 2011 to 2,271 hectares in 2015. As a fibre crop, kenaf bast and core are separated by retting or mechanical decorticator machine. The production of kenaf bast fibre was reported to reach around 11,602 tonne in 2015. The kenaf fibre is raw material for several types of products such as panels, woven and nonwoven textile, reinforced composites, high quality paper and furniture (Mohd et al. 2014). This variety of application of kenaf has contributed to RM309,385 and RM784,231 for domestic trade and export trade respectively for Malaysia economy in 2015.

FIBRE MORPHOLOGY

Kenaf and EFB present similar morphology, but they differ from each other by various factors such as diameter, length, number of fibre bundles, surface porosity, cell wall thickness and size of lumen. PP fibre, a man-made fibre shows different morphology from natural fibre. Kenaf fibres bundle have a diameter and length of 89.2 μm and 60 mm, respectively. Morphological images and SEM images of kenaf fibre are shown in Figures 1(a) and 2(a). EFB fibre bundle length was in the range of 30 to 180 mm with average diameter of 341.7 μm as shown in Figure 1(b) and 2(b). PP fibre has diameter and length of 36.3 μm and 6.64 mm, respectively. Diameter of the palm

TABLE 2. Statistics of Malaysian kenaf plantation 2011-2015

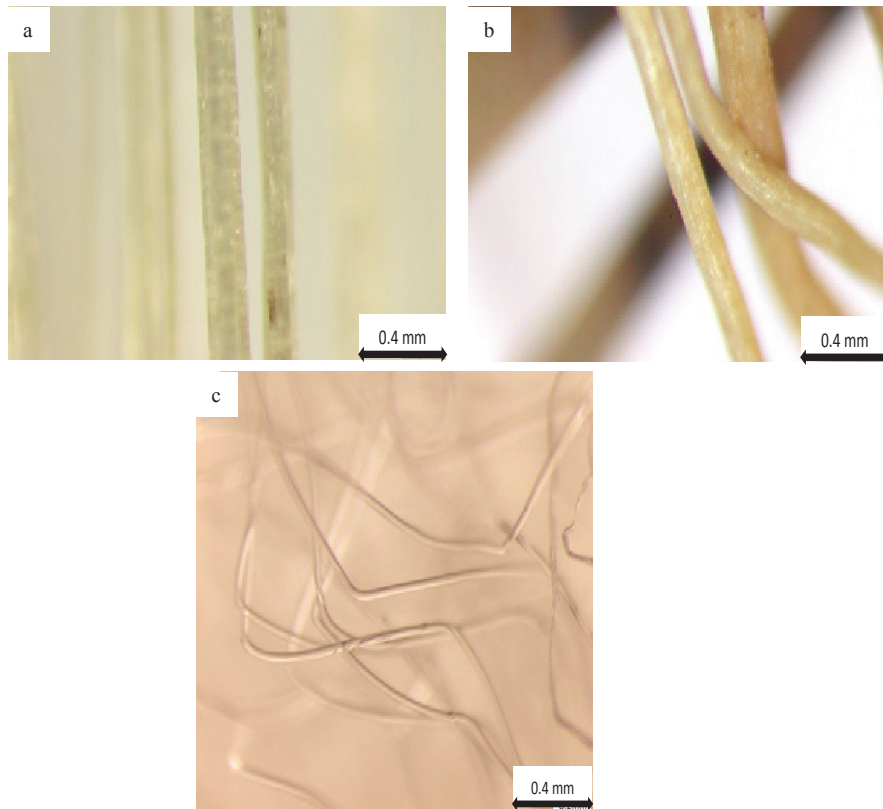|	2011	2012	2013	2014	2015
Total plantation (Hectare)	1,140	1,331	1,824	2,000	2,274
Production of kenaf bast fibre (Tonne)	2,266	4,090	7,176	7,573	11,602
Domestic trade value (RM)	-	-	63,764	208,906	309,385
Export trade value (RM)	-	-	-	11,557	784,231

(Source: NKTB)

FIGURE 1. Microscope images of (a) Kenaf, (b) EFB and (c) PP fibres

fibres was roughly 4 times higher than kenaf fibres and the distribution of diameter is uneven between fibre and along fibre length. PP fibre is finer and homogenous than natural fibre in diameter and length as shown in Figures 1(c) and 2(c). It is known that homogeneity and strength of fibre are influencing the strength of composite where the fibre is important to receiving the loads (Farkhan et al. 2016). Therefore, the selection fibre raw material especially in composite must have uniformity as possible.

Table 3 shows the physical properties of kenaf, EFB and PP fibres. Kenaf has highest density that was 1.62 g/cm³ as compared to EFB and PP fibre. Density measured in this experiment was slightly higher than those reported by other researchers (Asumani et al. 2012). Density of PP was 0.92 g/cm³. Distribution of kenaf fibre length was homogenous than EFB fibre since it has been reduced into 6 mm length. The distribution of EFB length was in the range of 3 to 18 mm and the distribution of the length was depending on the method of fibre extraction used by the company. The length of PP fibre was depending on the

TABLE 3. Physical properties of kenaf, EFB and PP fibre

	Diameter (µm)	Density g/cm3	Length (cm)
Kenaf fibres	89.2(37.7)	1.62(0.75)	6
EFB fibres	341.7(20.1)	1.52(0.44)	3-18
PP	36.35(11.55)	0.92(0.66)	6.64

( ): Coefficient of Variance in percentage

length prepared by manufacturer which determined by user. The critical length of fibre has an important effect to the composite tensile property where a composite with fibre at below than critical length damaged easily because of fibre pullouts. Ideally, fibre length would be much greater than the critical fibre length to allow for efficient reinforcement of a composite (Pickering et al. 2016). The critical fibre length for kenaf fibre reinforced composite has suggested around 6 mm (Abdul Khalil et al. 2010; Mohd et al. 2014).

Kenaf fibre possessed smooth surface, with few fibre bundle and distribution of pith and boarder pits could be observed. Kenaf cell wall was thicker compared to EFB cell wall (Figure 3). Each fibre bundles comprised of more than 25 fibres which were attached to one another by thin layered of middle lamella, composed of lignin which served as glue. A fibre bundle of kenaf is varying from 15 to 40 (Cheng et al. 2000). The cell wall fibres were multicellular type of cell, round to polygonal in shape with slightly rounded corner and medium lumen size (Tahir et al. 2011).

Unlike the wood of dicotyledons and gymnosperms, oil palm consisted of primary tissue without cambium. EFB consist of primary vascular bundle imbedded in parencymatous ground tissues (Figure 3(b)) and this is not comparable with developmental terms to the wood of dicotyledons and gymnosperm which is mostly consist of secondary xylem (Abdul Khalil et al. 2008). EFB fibre was a hard and tough multicellular fibre. It has porous and rough surface with many voids as clearly shown in Figure

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2(b). Inorganic materials perhaps silica was observed and embedded on the fibre surface. It is reported that the fibre may contained oil and other impurities (Ibrahim et al. 2015). The cross section of the EFB fibres is rounded to polygonal in shape as shown in Figure 3(b). Since the EFB fibre bundles have higher diameter compared to kenaf, the individual fibre which built up the EFB fibre bundle were more than 50 fibres. A vascular bundle of EFB fibres is a simple bundle surrounded by a sheath of a thickened cell layer. The vascular bundle surrounded by a few thick cell layers of fibre elements in a monocot plant is function as buckling resistance against lateral compression forces (Abdul Khalil et al. 2008).

PP fibres often have a smooth surface and circular cross-sectional shape as shown in Figures 2(c) and 3(c), but different cross-sectional shapes can be produced by different spinneret shapes to achieve enhanced performance. The production of nonwovens largely depends on PP fibres to achieve the lightweight, chemical and bacteria resistant qualities desirable for making disposable as well as durable nonwovens (Das et al. 2012).

Alves Fidelis et al. (2013) have found that the tensile strength of individual fibre have correlation with their different morphologies such as the internal area of the lumens, the number of lumens, the number and size of fibre-cells, the thickness of the secondary cell-walls and the real cross-section. Among the fibres analyzed, kenaf has the thickest cell wall, less number of lumen and small area of lumen which led to kenaf has better mechanical property than EFB.

CHEMICAL COMPOSITION

Cellulose, hemicellulose and lignin form major constituents of the natural fibres. The composition is differed depending on species, soil conditions, weather effect, plants age, growth condition and testing method used (Abdul-Hamid et al. 2009). Table 4 shows the differences in chemical composition between kenaf and EFB fibres. Chemical analysis of kenaf and EFB showed that the principle component of both fibres was cellulose. The α-cellulose in kenaf and EFB were 54.1% and 41.34%, respectively. Cellulose content has positive influence on the mechanical strength of fibre. This study showed that cellulose content and tensile strength of fibre have similar pattern where kenaf with higher cellulose content also have found to have higher mechanical strength than EFB. This is in agreement with Alves Fidelis et al. (2013) which have found that sisal with 73% cellulose showed higher tensile strength than jute with 65% cellulose content. The study by Salmén (2009) showed that the spectral molecular deformation during the loading of cellulosic material was assigned to the cellulose and no molecular deformation could be detected for lignin or hemicellulose which it can be concluded that cellulose is the main component for load transfer in fibre.
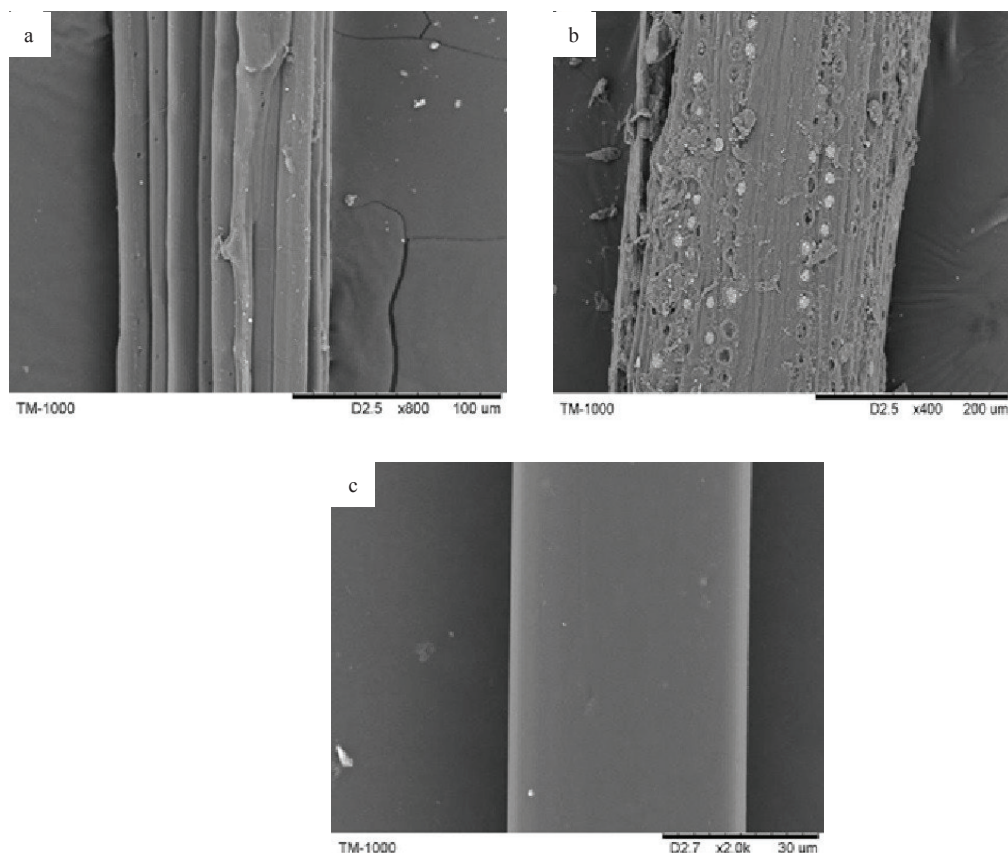

FIGURE 2. SEM images of (a) longitudinal section of kenaf fibre, (b) longitudinal section of EFB fibre and (c) longitudinal section of PP fibre

TABLE 4. Chemical component of kenaf and EFB

Component	EFB (%)	Kenaf (%)
Holocellulose	62.6	86.3
α-cellulose	41.34	54.1
Lignin	27.71	20.35
Extractive	2.81	1.38
Cold Water Solubility	9.20	1.15
Hot Water Solubility	9.40	2.16
Ash	5.29	2.21

Lignin content in EFB was 27.71% which was higher than lignin in kenaf at 20.35%. Kenaf lignin was composed of three lignin units, p-hydroxyphenyl, guaiacyl and syringyl in varying ratios and the distribution of lignin in fibre has related to the maturity of tissues (Abdul Khalil et al. 2010). Kuroda et al. (2005) found that variety of kenaf have influent to p-coumaric and ferulic acid moieties, a large abundance of syringylglycerolaryl ether subunits with the CH2OAc group, coniferyl and sinapyl alcohol acetates and S and G lignin content. EFBs had already been shown to be particularly rich in p-hydroxybenzoates and presence of natural acetates in lignin (Lu et al. 2015). The desirability content of lignin is subjected to the end products. Lignin in undesirable in pulp paper because its removal during pulping and bleaching requires high amount of energy and chemical. Paper with high purity of cellulose (chemical

pulp) is considered as strong paper compare with paper made from pulp which has high proportion of lignin (mechanical pulp). Beg and Pickering (2007) have also found that high amount of residual lignin content in composite led to lower tensile and impact strength and increased weathering degradability of composites.

Ash content in EFB fibre, at 5.29%, was higher than in kenaf fibre, at 2.21%. Ash consists of inorganic element such as calcium, magnesium and silica (Ibrahim et al. 2015). This silica content can be clearly seen embedded on the fibre surface when observed under SEM. High content of silica in plant can cause the mechanical cutter become blunt during the cutting process. This inorganic element will also create problem especially chemical recovery during pulping process. The alkali treatment with NaOH made the fibre surface rougher with fewer amounts of silica bodies and studies claimed that increasing surface roughness results in better mechanical interlocking that improved adhesion ability of fibre with the matrix (Ibrahim et al. 2015).

The cold-water procedure removes a part of extraneous components, such as inorganic compounds, tannins, gums, sugars and coloring matter present in fibre while the hot water procedure removes, in addition, starches (TAPPI T207). The cold and hot water extraction of EFB was 9.2% and 9.4%, respectively, which higher than kenaf were 1.38% and 1.15%, respectively. The study by Abdullah and Sulaiman (2013) showed that the water washing pretreatment of EFB was sufficient to remove ash. High amount of cold and hot water extraction
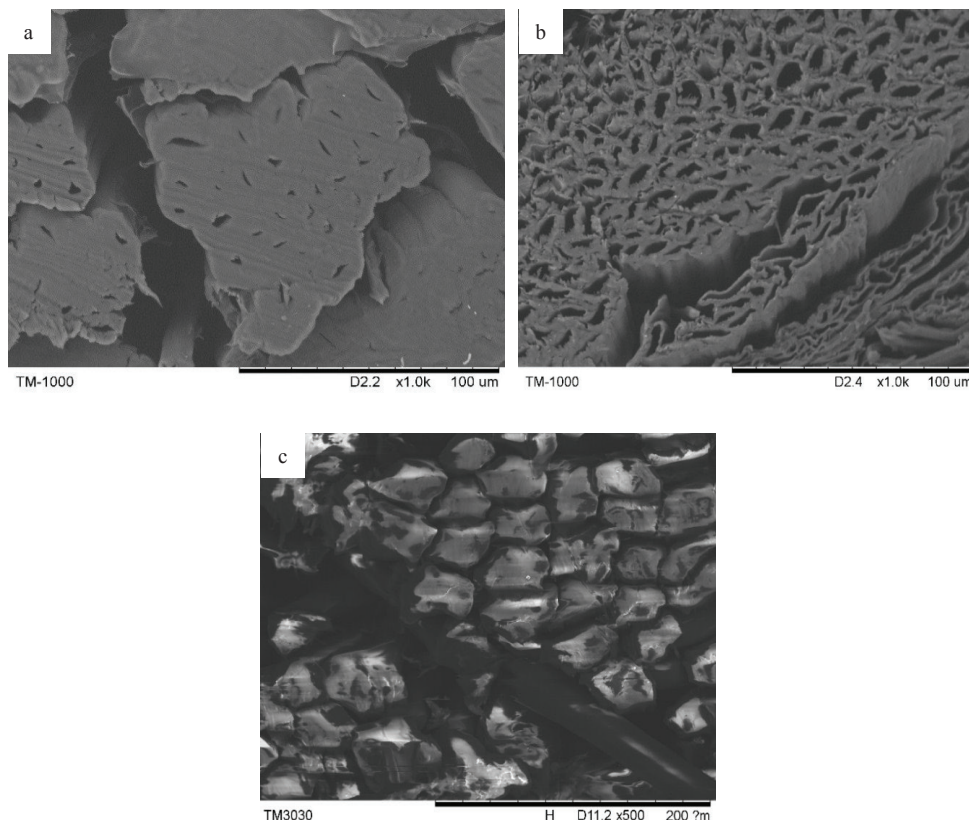

FIGURE 3. SEM images of (a) Cross section of kenaf fibre, (b) Cross section of EFB fibre and (c) Cross section of PP fibre

of EFB may due to high content of ash washed during the extraction.

PP is one of the most versatile thermoplastic polymers available commercially, produced by polymerization of Propene. It is generally available as chips or granules which are white and semi crystalline. The density of PP is 0.905 g/cm^{-3}, one of the lightest thermoplastics materials with melting point of 167°C. The propene molecule is asymmetrical and when polymerized, can form three basic chain structures dependent on the position of the methyl groups: two are stereoregular (isotactic and syndiotactic) and the third does not have a regular structure and is termed atactic. PP used in this study is isotactic PP polymer which is mainly used in nonwoven industry in the world. This is resulted from property, cost and ease procedure of processing of PP (Kalebek & Babaarslan 2016).

FTIR ANALYSIS

FT-IR spectroscopy is a tool for studying the physico-chemical and conformational properties of Polysaccharides. Figure 4 shows the FT-IR spectra for kenaf, EFB and PP. There were major differences observed in FT-IR spectra of PP and natural fibre EFB and kenaf. Kenaf and EFB showed a broad peak at 3,100 to 3700 cm^{-1} were corresponded to the stretching vibration of the hydroxyl group (Jonoobi et al. 2011). Hydroxyl group was contributed from moisture and abundant of hydroxyl group available in cellulose gluco-pyranose ring and hemicellulose. The absorption band between 3200 and 1800 cm^{-1} is attributed by the hydrocarbon stretching region (Jonoobi et al. 2011), which appeared at 2905, 2880 and 2905 cm^{-1} for EFB, kenaf and PP, respectively. The carbon double bond region will appear around 1800 - 1500 cm^{-1} and this stretching of carbonyl group is important peaks for the present of any carbonyl bond in key feature of spectrum (Zakaria et al. 2014). This

peak can be seen at 1720 cm^{-1} in kenaf and 1736 cm^{-1} in EFB attributed to the C=O stretching in the acetyl and uronic ester groups of hemicellulose or the ester carbonyl groups in the p-coumaric units of the lignin. The peak observed at 1578 cm^{-1} is attributed to the C=C stretching of the lignin carbonyl group while the peak at 1250 cm^{-1} is associated with the C-O-C stretching of the aryl–alkyl ether linkage in lignin (Yang et al. 2007). The broad peaks at 1050 cm^{-1} are assigned to ether linkage (C=O=C) from lignin or hemicellulose (Bakar et al. 2006).

MECHANICAL PROPERTIES

A tensile strength property indicated how a material will react to the force applied in tension direction and this an important characteristic for fibre properties. Table 5 shows the tensile strength, tensile modulus and tensile elongation of kenaf, EFB and PP. Kenaf fibre has the highest tensile strength that was 426.4 MPa followed by PP at 161.44 Mpa and EFB at 150.9 Mpa. Kenaf also has highest tensile modulus among others that was 30 Mpa. The tensile modulus of EFB was higher than PP that were 2.9 Gpa and 1.6 Gpa, respectively. PP fibre has signicantly higher tensile elongation than natural fibre EFB and kenaf that was 210.9%. EFB considered as one of the natural fibre that has high tensile elongation at 30% similar to coconut coir (Bledzki & Gassan 1999).

The fibre strength will influence the composite strength if the fibres are used as the reinforcement material. The properties of the fibres such as tensile strength, flexural strengths, and rigidity depend on the alignment of cellulose fibrils, which are generally arranged along the fibre length (John & Thomas 2007). The tensile strength of the oil palm fibres was only about 1/3 of the kenaf fibres and the tensile modulus of the palm fibres was only about 1/10 of the kenaf fibres. Meanwhile the tensile elongation of the palm fibres was about 10 times of the kenaf fibres. In this case, kenaf is
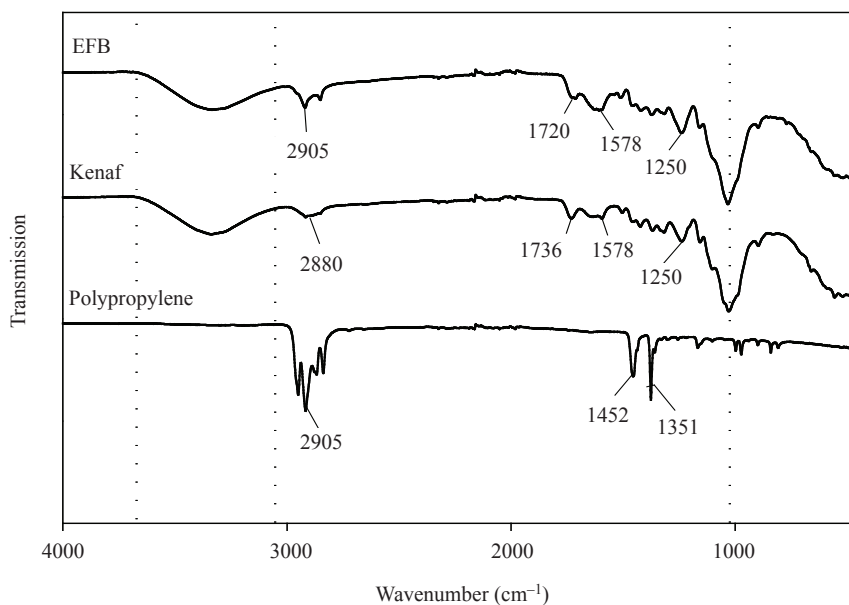

FIGURE 4. FTIR patterns of kenaf, EFB and PP fibre

TABLE 5. Mechanical properties of the kenaf, EFB and PP fibres

	Tensile strength (MPa)	Tensile modulus (GPa)	Tensile elongation (%)
Kenaf fibres	426.4(37.2)	30.0(34.2)	2.9(24.4)
EFB	150.9(81.9)	2.9(68.1)	30(32.7)
PP	161.44(3.17)	1.6(23.3)	210.93(9.3)

( ): Coefficient of Variance in percentage

a better choice between this natural fibre since it has better tensile strength and modulus thus will contribute to good mechanical properties for composite panel.

CRYSTALLINITY

X-ray diffractometer was used to measure the crystalline structure of lignocellulosic materials, where it is possible to capture the image of cellulose crystal directly. Cellulose is partly crystalline and partly amorphous in molecular structure (Jonoobi et al. 2011). This implied that the cellulose chains will be closely held by mutual H-bonding in the crystalline (ordered) regions whereas no H-bonding occurs in the amorphous (disordered) regions of the cellulose chains (Parikh et al. 2007). The X-ray diffraction patterns of kenaf, EFB and PP are shown in Figure 5. The crystallinity values of all fibres varied widely and kenaf samples in general had higher degree of crystallinity than EFB and PP. The crystallinity values of the kenaf, EFB and PP were 65%, 50.58% and 50.81%, respectively. PP is semi crystalline polymer (Kalebek & Babaarslan 2016) and the crystalline forms of isotactic PP are α-monoclinic which is the most commonly observed, β-hexagonal, which occurs under specific conditions that is, temperature gradients, shearing forces or β-nucleating agents and γ-triclinic, which is the least common observed in low molecular weight PP (Han et al. 2012).

Kenaf has higher cellulose content than EFB as well as the crystallinity value and this result in agreement with the theory of the cellulose crystallinity where cellulose content has linear correlation with crystallinity. Joonobi et al. (2011) studied the effect of alkaline treatment on crystallinity of cellulose and result showed that optimum alkaline treatment changed the amorphous and crystalline region in cellulose where peeling reactions in the amorphous areas increased the crystallinity of cellulose. However severe alkaline treatments on fibre change the chemical structure in cellulose molecule chains and caused the reduction of the crystalline structure (Farkhan et al. 2016).

THERMOGRAVIMETRIC ANALYSIS

Figure 6 shows the TGA analysis of all fibres. Thermal decomposition of lignocellulosic materials begins with an early weight loss of moisture content, decomposition of hemicelluloses, followed by depolymerization of cellulose, pyrolysis of lignin, active flaming combustion and char oxidation (Lee et al. 2014). It showed clearly that the thermal decomposition of kenaf fibre and palm fibre were similar, which can be divided into three stages. The first stage 80-120° were the initial thermal decomposition of moisture, the second stage 120-400° were the main thermal decomposition of hemicellulose and cellulose
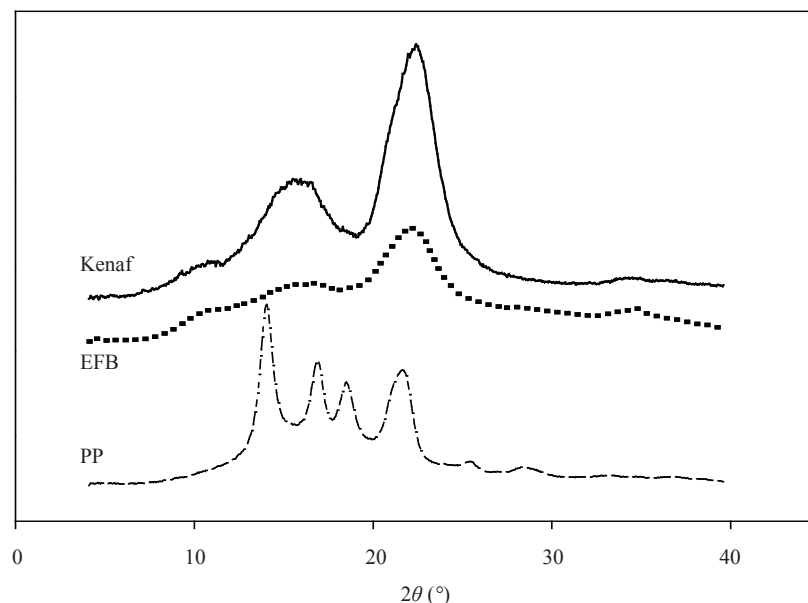

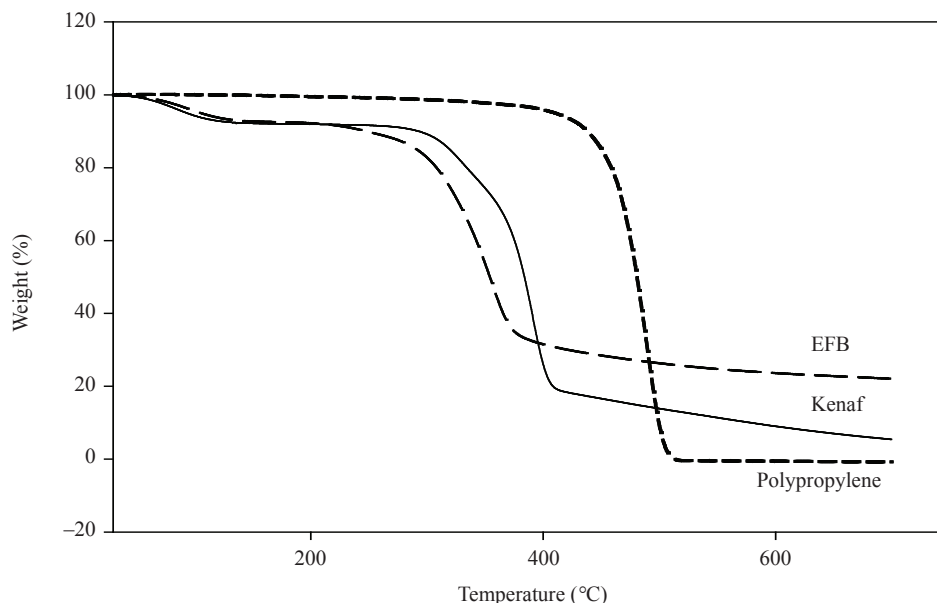FIGURE 5. The XRD patterns of kenaf, EFB and PP fibre

FIGURE 6. TG curves of kenaf, EFB and PP fibre

and followed by 400-600° were the pyrolysis of lignin and char oxidization. Comparing kenaf fibre and palm fibre with PP, the thermal decomposition process of PP was relatively simple with one stage decomposition started at 396°.

Kenaf showed higher thermal degradation temperature than EFB at 342° and 311°, respectively at the second stage of decomposition. Lee et al. (2014) also reported that hemicellulose established an earlier decomposition than cellulose in terms of thermal breakdown. The random and branched structure of hemicellulose is predominantly amorphous with lower molecular weight in their chemical structures. Thus, hemicellulose is more readily oxidised compared to cellulose especially during oxidation. In addition, lack of crystallinity may be also leading to the low thermal stability (Joonobi et al. 2011). This suggested that cellulose content and crystallinity has impact to the higher thermal degradation at the second stage of lignocellulose decomposition where kenaf with higher cellulose content and crystallinity than EFB proved to have higher thermal degradation temperature. The residual mass of kenaf fibre was 17.69% and 25.05% for EFB. The higher residual mass in EFB may due to the higher content of inorganic material such as silica compared to kenaf. This was verified in the study conducted by Lee et al. (2014) where the residues left at the end of combustion showed similar trend with the content of impurities between control and treated EFB.

PP fibre possessed the highest thermal stability with lowest residue compared to both natural fibre. It started from 396 to 460°, and fibre was almost exclusively thermal decomposed with the residual mass of 1.13%. The selection of processing method, matrix system and range of temperature application of natural fibre as composite material has to consider the thermal degradation temperature of the natural fibre (Dicker et al. 2014).

DIFFERENTIAL SCANNING CALORIMETRY (DSC)

Differential Scanning Calorimetric (DSC) is a thermo analytical technique to determine the thermal transitions of polymer and Figure 7 shows the DSC of all fibres. The DSC curve of PP showed strong endothermic peak in the vicinity of 165°C, which was produced by the melting of PP and another one generated in the vicinity of 469°C, which was the decomposition temperature of PP. Hence, the processing temperature of the PP fibre should exceeded 165°C above the melting point. It showed clearly a strongly endothermic peak appears at 87°C for kenaf and 80°C for oil palm fibre which was caused by the evaporation of free water in the fibres. A strongly endothermic peak also appeared at 375°C for kenaf and exothermic peak at 365°C for palm fibre, was due to the thermal decomposition of cellulose, lignin, pectin, hemicellulose and other components. Kenaf showed better thermal stability than EFB fibre may due to higher cellulose and crystalline area (Joonobi et al. 2011).

Low glass transition temperature and melting point temperature reduce the shelf-life of PP due to changes in the configuration of polymer chains at high temperature which make them poor resistance to ultraviolet radiation, susceptibility to degradation under oxidative and radiation-induced conditions and poor adhesive properties (Das et al. 2012). Hao (2013) indicated that the activation energy value of kenaf/PP composite was higher than pure PP and this value proved that natural fibre with PP mixture were more thermally stable than pure PP plastics.

CONCLUSION

Comparison study of physico-mechanical, chemical composition, thermal degradation and crystallinity among natural fibre kenaf and EFB and between synthetic fibre PP were achieved by various method in this work. Our morphological results showed that diameter of the EFB
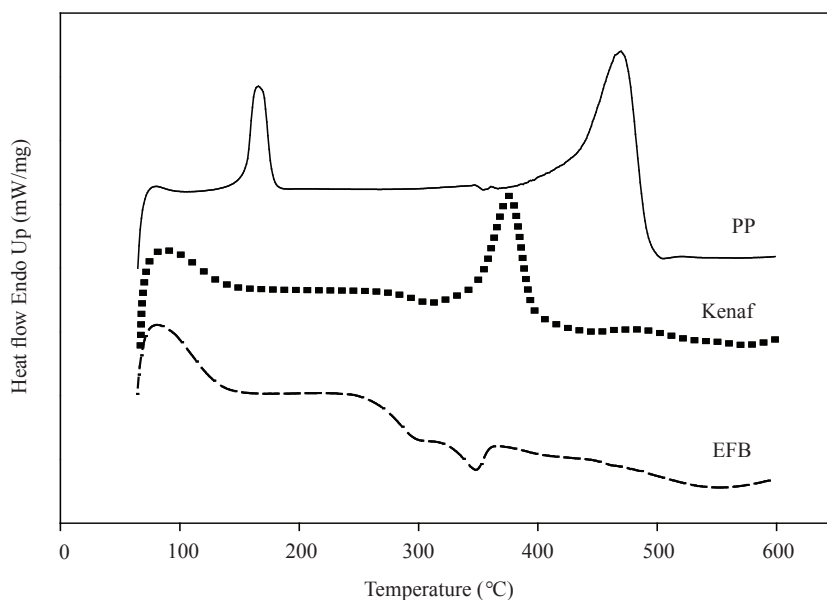
FIGURE 7. DSC curves of kenaf, EFB and PP fibre

fibres was roughly 4 times higher than kenaf fibres and the distribution of diameter was uneven between fibre and along fibre length. Characterization of the fibres by SEM provided evidence that kenaf has the thickest cell wall, less number of lumen and small area of lumen. Kenaf fibre possessed smooth surface, with few fibre bundle and distribution of pith and boarder pits while EFB has porous and rough surface with inorganic materials such silica embedded in many voids on the fibre surface.

The chemical structures of the kenaf, EFB and PP fibres were compared by FTIR spectroscopy. The FTIR spectra indicated nearly similar peak between kenaf and EFB due to the same composition of cellulose, hemicellulose and lignin in the natural fibre. The quantity of the chemical composition in kenaf and EFB were compared by extraction method and kenaf indicated higher amount of cellulose than EFB. In the other hand, EFB showed high amount of extractive and ash.

Results of the thermogravimetric analysis demonstrated that kenaf has better thermal degradation property relative to the EFB fibres. PP showed melting temperature at 165°C while no melting temperature observed from natural fibre. The XRD analysis, which was employed for estimation of the crystallinity of the produced fibres, unfolded that kenaf has highest crystallinity at 65% among the fibres.

The results obtained from X-ray diffraction analysis, chemical composition and thermogravimetric analyses indicated that there were significant different between the studied fibre sources in the cellulose crystallinity, thermal stability and chemical composition. Based on our results, it is suggested that kenaf has better mechanical strength, thermal degradation and crystallinity than EFB which influenced by the morphological and chemical composition of the fibre. It subsequently suggested that a stronger mechanical strength and higher thermal degradation can be obtained from kenaf/PP composite than EFB/PP composite.

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