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The Effect of Kenaf Filler Reinforcement on the Mechanical and Physical Properties of Injection Moulded Polypropylene Composites

(Kesan Penguatan Pengisi Kenaf ke atas Sifat Mekanik dan Fizikal Pengacuan Suntikan Komposit Polipropilena)

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ABSTRACT

Natural fibres potentially offer better reinforcement to improve the mechanical and physical properties of polymer composites. However, these natural materials at this stage are not fully explored yet due to the fibres themselves have limited heat resistance and are quite sensitive to moisture. This limitation will weaken the adhesion when interacting with thermoplastic matrices during the processing of composites. Therefore, the main purpose of this study is to investigate inherent strength characteristics among kenaf (core and bast) fillers as a reinforcement in polypropylene composites at various geometries and loadings via the injection moulding process. The composite materials consisted of kenaf with the geometric core filler of the 20 mesh (992 µm), 40 mesh (460 µm) and bast filler (166.9 µm) were mixed with polypropylene based on the filler loadings of 10 up to 40 wt. %. The results showed that bast filled composites had the highest tensile strength of 19.52 MPa at 30 wt. %, compared to core filled composites. Instead, 20 mesh core filled composites were obtained had the highest flexural strength which values were 25 MPa and 29 MPa at 20 wt. % and 30 wt. %, respectively. While 40 mesh core filled composites had the highest values of 25.35 MPa at 40 wt. % of filler loading compared to bast filled composites. SEM micrograph images showed the good interfacial bonding of core filler which surrounded by PP leading to diffusion and permeation of bonding. In conclusion, the use of kenaf (core and bast) fillers as a reinforcement in composite materials is reasonable to maximise the use of fibre from natural sources.

Keywords: Injection moulding; kenaf filler; mechanical properties; polypropylene; SEM micrograph images

ABSTRAK

Serabut semula jadi menawarkan kekuatan penguat yang lebih baik bagi meningkatkan sifat mekanik dan fizikal komposit polimer. Walau bagaimanapun, bahan semula jadi ini masih belum diterokai sepenuhnya kerana sifat serabut itu sendiri yang mempunyai rintangan haba yang terhad dan sensitif terhadap lembapan. Keterbatasan ini akan melemahkan rekatan apabila digandingkan bersama matrik termoplastik semasa pemprosesan komposit. Maka kajian ini bertujuan untuk mengkaji kekuatan yang wujud antara pengisi (teras dan bast) kenaf sebagai penguat dalam komposit polipropilena dengan pelbagai geometri dan pembebanan melalui proses pengacuanan suntikan. Bahan komposit yang terdiri daripada kenaf dengan geometri pengisi teras 20 mesh (992 µm), teras 40 mesh (460 µm) dan pengisi bast (166.9 µm) yang dicampur dengan polipropilena berdasarkan pengisi sebanyak 10 hingga 40 % bt. Keputusan menunjukkan komposit berpengisi bast mempunyai nilai kekuatan tegangan yang tertinggi sebanyak 19.52 MPa pada 30 % bt., berbanding komposit berpengisi teras. Sebaliknya, komposit berpengisi teras 20 mesh pula didapati mempunyai kekuatan lenturan yang tertinggi sebanyak 25 MPa dan 29 MPa masing-masing pada 20 dan 30 % bt. Manakala komposit berpengisi 40 mesh mempunyai kekuatan lenturan bernilai 25.35 MPa pada beban 40 % bt., berbanding komposit berpengisi bast. Keputusan mikrograf SEM menunjukkan ikatan antara muka yang terbaik terbentuk oleh pengisi teras kenaf yang dikelilingi sepenuhnya dengan PP, dengan ikatan terbentuk melalui penyebaran dan penyerapan. Kesimpulannya, penggunaan pengisi (teras dan bast) kenaf sebagai bahan penguat dalam komposit adalah munasabah dalam usaha untuk memaksimumkan sepenuhnya penggunaan gentian daripada sumber semula jadi.

Kata kunci: Kekuatan mekanik; mikrografi SEM; pengacuanan suntikan; pengisi kenaf; polipropilena

Introduction

Natural plants have the potential to replace traditional and mineral fillers as reinforcements in polymer matrix composites (Ismail et al. 2013). Over the past few decades, biodegradable materials have attracted considerable interest due to their impact on the environment caused by petroleum-based resources. Since the inception of glass fibres being used in the interior and exterior of car components, several shortcomings were evident. These included; high energy consumption, high relative fibre density (approximately 40% greater than natural fibres), difficulty in processing, distressed recycling properties and non-degradability. All shortcomings were recognised as requiring improvement and potentially (Koronis et al. 2013; Ku et al. 2011). In fact, many of the deficiencies contributed to environmental pollution and potential health hazards caused by glass fibre particles.

More recently, the utilisation of lightweight, renewable resources and the recycling of natural fibre composites, attributed to lower CO_2 emissions and less dependence on fuel sources. This provided the opportunity to replace the variety of glass fibres employed within the automotive and like industries, (i.e. building & construction) (Othman & Ismail 2007). Bast and core are found in agricultural plant crops such as hemp, flax, jute, sisal, ramie and kenaf and are suitable to use as natural reinforcement materials in polymer composites (Dehbari et al. 2014). Kenaf (*Hibiscus cannabinus* L.) fibres have a low density (1.2 - 1.6 g/cm^3), have excellent properties, are non-abrasive and offer high stiffness properties and significantly affect both the physical and mechanical properties of polymer composites (Rowell et al. 1997). Kenaf as one of Malaysia's many industrial crops is expected to make a major contribution to the processing industry. This is owing to the significant increase in research being undertaken relative to the technical potential of the commercialised fibre in comparison with other fibre plants available globally. Kenaf is a cheap renewable material source offering a sustainable alternative to commercialise composite materials for the development of high-performance engineering products.

Kenaf is a fibrous (woody) plant with the stem consisting of two distinct fibre sources namely core (65%) and bast (35%) (Akil et al. 2011). Habibi et al. (2008) observed that cellulose is the main component of bast and core as well as, hemicellulose and lignin which influences the mechanical properties of the composite. High cellulose content provides excellent strength and stiffness to reinforce composites owing to the strong hydrogen bonds and other linkages. Bast has a higher cellulose content (52% - 59%) than core, of about (44% - 46%). In fact, hemicellulose, lignin, ash and other chemical composition materials are responsible for biodegradation, moisture absorption, thermal degradation and UV degradation of kenaf fibres (Dzuhri et al. 2015; Hashim et al. 2016). Moreover, based on previous research, natural fibres with a larger diameter of lumen, with smaller cells and thicker walls, have the potential to produce composites given the strong reinforcement characteristics that result in higher mechanical and physical strength (Abdul Khalil et al. 2010).

Previous studies highlighted that most natural fibres reinforced composites are fabricated using compression moulding (Ishak et al. 2010; Sarifuddin et al. 2013), hot pressing (Aisyah et al. 2013; Saad & Kamal 2012) and hand layout process (Rahman & Halim 2012). Earlier research endeavoured to blend several types of natural fibres with polypropylene (PP) using an injection moulding process (Rowell et al. 1997). The improvements of tensile and flexural properties of kenaf/PP composites at 50 wt. % were obtained. This achieved a strength comparable with injection-moulded glass fibre/PP composites at 40 wt. %. Researchers have investigated bast (i.e. short and long) fibres obtained from natural plants as a reinforcement, applying various methods and processes. Unfortunately, limited studies identified the potential mechanical strength of the core filler as a reinforcement in polymer composites by applying injection moulding (Clemons & Sanadi 2007; Islam et al. 2012; Jeyanthi et al. 2011). Notwithstanding that core filler material has not been extensively researched, further studies are required specifically on how to optimise the usage of fibre from natural sources. Therefore, the purpose of this study was to compare and observe the potential of core and bast in the variable geometry of fillers as a reinforcement in kenaf/PP composites using the injection moulding process. This study concentrates on the effects of the numerous sizes and loadings of kenaf core and bast filler on the tensile and flexural properties of polypropylene composites.

Materials and Methods

The kenaf core (20 and 40 mesh) in the form of filler and bast fibre was purchased from the National Kenaf and Tobacco Board, Malaysia (LKTN). Table 1 lists the properties. The length of the kenaf bast fibre was shortened manually using a Pallman Knife Ring Flaker. The fibre was then sieved to obtain a size less than 500 μm using a vibrating sieve machine. The material was measured using a Malvern Particle Analyser to observe the distribution of kenaf 20 mesh, 40 mesh and bast filler having D_{50} of (992.3 μm), (460.0 μm) and (166.9 μm), respectively. The matrix used was polypropylene graded SM850 (Lotte Chemical Titan (M) Sdn. Bhd.) having a high melt flow rate of 45 g/10 min (ASTM D1238) and being suitable for the injection moulding process. Figure 1 shows the image and SEM micrograph of kenaf filler materials.

Kenaf fillers and PP were dried in an oven for 24 h at 80°C to remove any absorbed moisture and to avoid,

TABLE 1. Properties of kenaf fillers

Kenaf filler	Average particle size (μm)			*Measured moisture content (%)
	D_{10}	D_{50}	D_{90}	
20 mesh core	558.8	992.3	1576.2	< 12
40 mesh core	147.4	460.0	1140.1	< 12
Bast	34.9	166.9	883.5	< 18

*Provided by LKTN

FIGURE 1. Image and SEM micrograph of kenaf filler: (a-i & a-ii) 20 mesh core, (b-i & b-ii) 40 mesh core, and (c-i & c-ii) bast

the occurrence of porosity, the possible deterioration in mechanical properties and any loss of dimensional stability (Sarifuddin et al. 2013). The materials were then melt-blended for 25 min at 190°C using a Sigma Blade mixer at 45 rpm. The mixing temperature was set higher than the melting point of the PP matrix (166.82°C) to ensure that the polymer mixed with the fillers homogenously. Initially, the PP pellet was placed in the mixer for 15 min, until the mixing›s torque was stable (El-Shekeil et al. 2011). The fillers were then added carefully, allowing to operate at the time and speed as mentioned above, to obtain a homogeneous mixture. After mixing, the melt-blended materials, called feedstock were cooled to ambient room temperature and crushed using a hard crusher machine. Figure 2 shows the images of the crushed pellets.

Feedstock pellets were again dried in the oven for 12 h to maintain their dryness before being injected using a Battenfeld BA 250 CDC injection machine. The injection temperature, injection pressure, holding pressure and injection rate were set to 190°C, 120 MPa, 180 MPa and 18 cm^3/s, respectively. The parameter values were selected based on trial-and-error tests, to identify suitable values for the parameters of the injection-moulded kenaf/PP composite. The process was applied without producing any moulding defects incurred on the samples, such as short shot, sink marks and voids (Anuar et al. 2012; Khalina et al. 2011). The prepared composites were then moulded into standard shapes for tensile and flexural tests. Table 2 lists the formulation of the composition in the kenaf/PP composites. Filler loadings of 10 to 40 wt. % were applied for both types of kenaf fillers.

TABLE 2. Composition of the composites

Filler	Compositions mixture (wt. %)	
	PP	filler
20 mesh core	90	10
	80	20
	70	30
	60	40
40 mesh core	90	10
	80	20
	70	30
	60	40
Bast	90	10
	80	20
	70	30
	60	40

FIGURE 2. Crushed pellets of the kenaf/PP composites

The tensile (ASTM D638) and flexural (ASTM D790) properties of the composites were evaluated using a Universal Testing Machine (Instron 5567). Based on the standard, the fixed of tensile crosshead speeds at 5 mm/min was used as designated follows the type of samples. While the flexural crosshead speeds of 1.37 mm/min was calculated through the equation below:

$$R = ZL^2/6d, \quad (1)$$

where R is the crosshead speeds (mm/min) $Z = 0.01$; L is the support span dimension (mm); and d is the beam depth. The support span dimension was mentioned as 16 ± 1 times by the depth of specimen, which is 51.2 mm was used in this test. Figure 3 illustrates the dimensions of the samples with a thickness of 3.2 mm, used for both mechanical testing. At least five specimens were tested to obtain a meaningful average and standard deviation. All tests were performed at ambient room temperature.

The morphology of the tensile fracture specimens was observed using a SEM (Table top Microscopic, model-TM 1000) with 10.00 kV of voltage to visualise the bonding and interaction between kenaf fillers and PP on the properties of the kenaf/PP composites.

RESULTS AND DISCUSSION

PROCESSING EVALUATION

Figure 4 illustrates the mixing condition in the compounder. During the mixing process, the mixing, friction and shear forces were carefully considered due to the significant increase in the temperature that could burn the fillers. For a similar outcome, the compounds were mixed applying a set temperature of 190°C to avoid any effect on the composite properties caused by the degradation of kenaf fillers. The mixing temperature was set to 30°C above the melting temperature of the polypropylene matrix (El-Shekeil et al. 2012). A further issue arose when the highest filler loading percentage was applied up to 50 wt. %. Mixing did not work beyond this limit because of the insufficient polymer matrix needed to completely wet the fillers. This led to poor interfacial bonding between the fibre and the matrix. During the moulded injection process, several problems were observed where the injection of kenaf/PP composites was dependent upon the feeding step. Yang et al. (2012) reported that pellet feeding should be carried out manually instead of automatically, to prevent jamming and occasionally stopping the filling process.

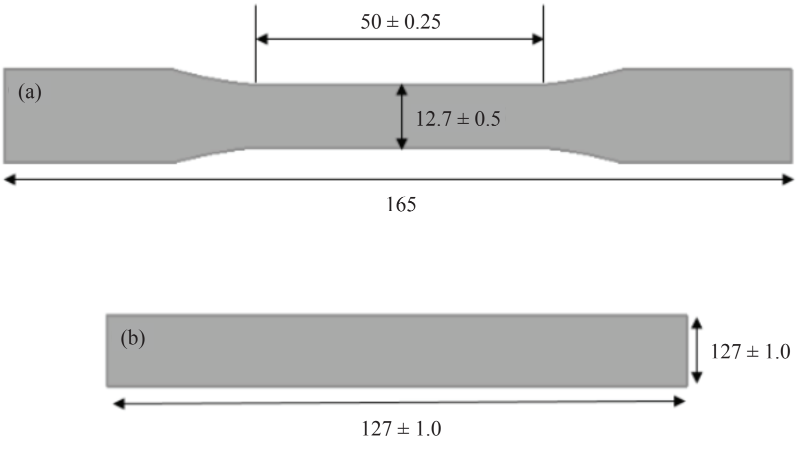


FIGURE 3. Samples dimension: (a) tensile and (b) flexural test

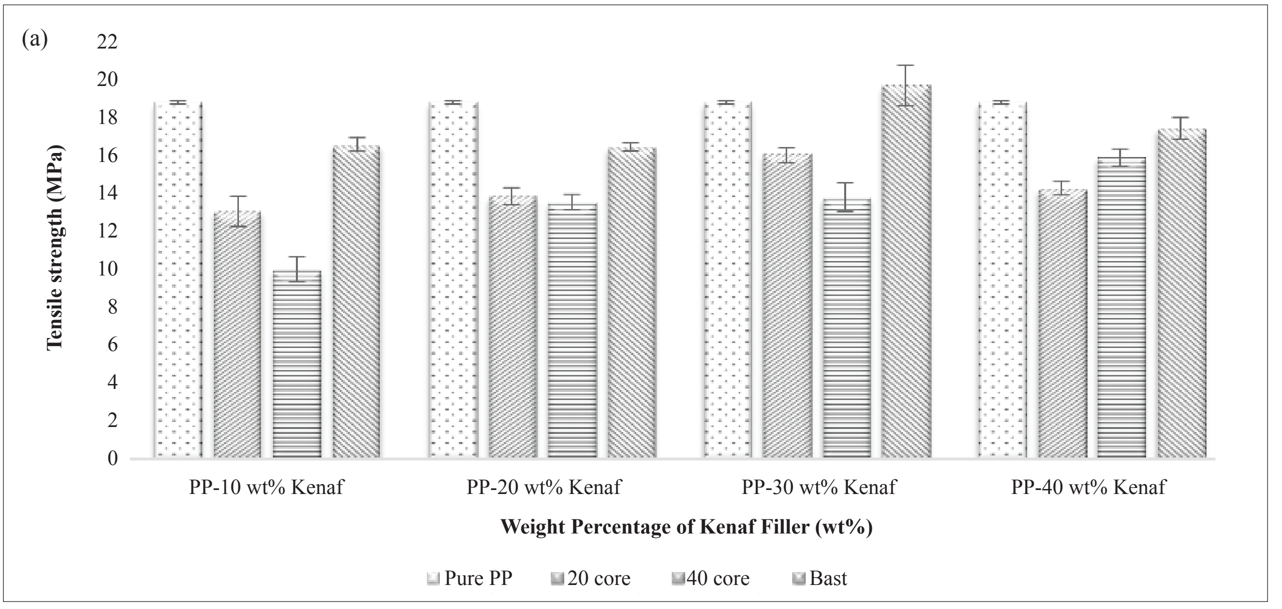
FIGURE 4. The mixture condition in the compounder at (a-i & a-ii) 40 wt. % and failure mixture at (b-i & b-ii) 50 wt. %

TENSILE PROPERTIES

Figure 5 illustrates the histogram pattern of the tensile strength and Young's modulus of both kenaf core and bast filled composites at different filler loadings. Comparisons were determined between the core and bast filled composites. Based on preliminary observations, core filler with varied sizes was observed contributing to the strength of the composites with increases in the filler loading. For the core filled composites, the highest tensile strength for 20 and 40 mesh core filled composites was achieved at 30 wt. % (15.91 MPa) and 40 wt. % (15.76 MPa) of filler loadings, respectively. Furthermore, the 20 mesh core filled composite exhibited higher tensile strength than the 40 mesh core filled composite at certain filler loadings. This occurred due to the stronger filler and matrix interfacial adhesion which significantly affects the strength of the reinforced composites (Fu et al. 2008). Moreover, the low strength of the 40 mesh core filled composite is a result of the smaller size of the filler which yields a larger surface area thereby leaving more nonreactive surfaces to the matrix. Also, the reduction of the tensile strength occurred due to a greater number of stress points created from the matrix to fibre (Rozman et al. 2011). In fact, the variation of the fibre sizes significantly affected the interfacial shear, normal stresses and the fracture characteristics (Bismarck et al. 2002).

For the bast filled composites, the highest tensile strength of 19.52 MPa was achieved at 30 wt. % filler loading, representing good strength of the filled composites. The strength properties of bast filled composites may influenced by chemical compositions (i.e. cellulose, hemicellulose and lignin) of plant fibres (Reddy & Yang 2005). Where cellulose as a primary structural component, is one of the strongest and stiffest organic mechanisms existing in natural fibres providing strength and stability to plant cell walls. According to Bismarck et al. (2002), and Kwon et al. (2014), tensile strength properties of reinforced composites increase with the increase of fibre loadings due to the addition of cellulose composition in the fibres. Ishak et al. (2010) also applied the optimal fibre loadings in achieving the highest tensile strengths of 19.0 MPa and 16.0 MPa for their kenaf core and bast filled composites, respectively.

However, the enhancement of the tensile strength for 20 mesh core and bast filled composites were ceased at 40 wt. % filler loading due to excessive filler content (≥ 40 wt. %). This excessive filler content causing the polymer matrix to insufficiently wet the filler in its entirety and leading to poor interfacial bonding between the filler and the matrix. This poor wetting condition was shown in Figure 4. But the trend observed in the results for tensile strength of 40 mesh core filled composite increasing with increase filler loading and were closely comparable to the strength of the bast filled composite. Even having nonreactive surfaces due to larger surface area, the smaller size of 40 mesh core fillers also have



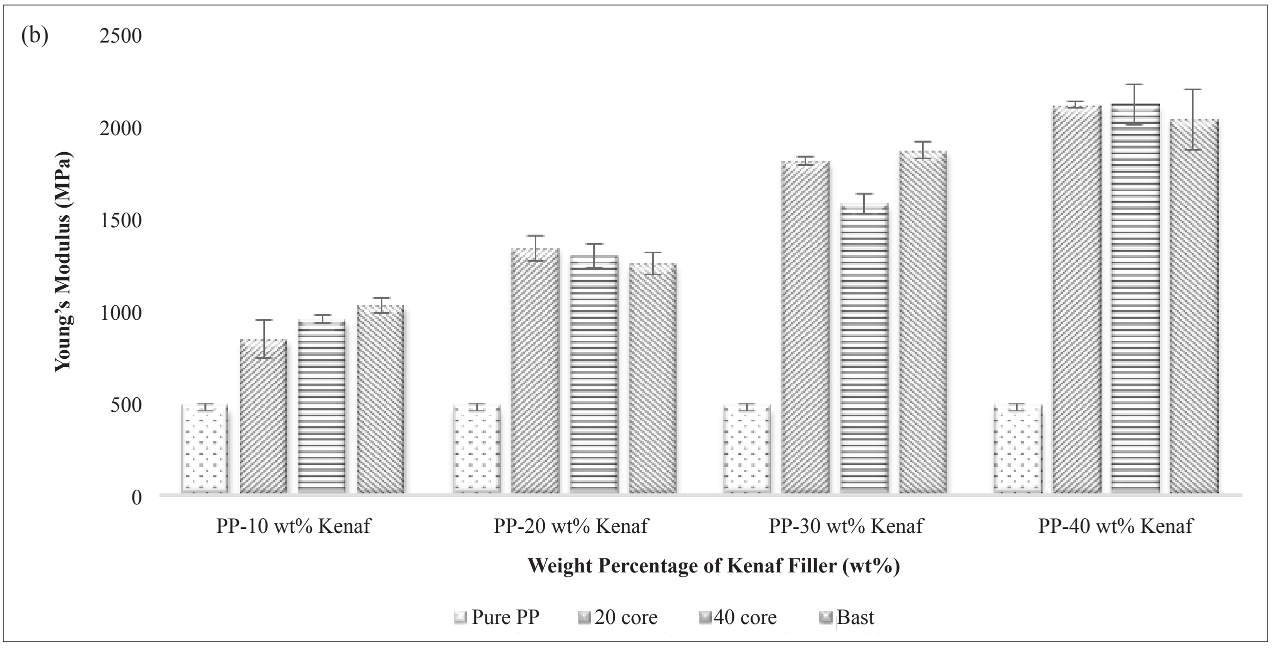


FIGURE 5. (a) Tensile strength and (b) Young's modulus of kenaf/PP composite at different filler loadings

a lot of hollow lumens on their surfaces when involved with highest filler loading percentage. This characteristic would improve the interfacial bonding, where the matrix can penetrate through some of the hollow lumens in core filler fibres (Balasuriya et al. 2001). Therefore, the tensile strength of 40 mesh core filled composites could increase when achieved the 40 wt. % of filler contents compared to 20 mesh core filled composites.

Figure 5(b) shows Young's modulus of the kenaf filled composites reflecting the influence of the differences in filler sizes and loadings of the core and bast filled composites. The addition of both kenaf fillers resulted in an increase in Young's modulus of kenaf filled composites. Also, the modulus of kenaf filled composites increased significantly with the increase in filler loadings, where the highest value for 20 and 40 mesh core filled composites was 2102.21 MPa and 2110.18 MPa, respectively, at 40 wt. %. By comparison, a slightly lower modulus of 2026.51 MPa was observed for kenaf bast filled composite at 40 wt. %. Since the Young's modulus are represented the stiffness of a composite. Hence based on the modulus strength results, kenaf 20 and 40 mesh core filled composites are obtained stiffer than bast filled composites at certain loading percentage. This decision is referred to Ismail et al. (2010), where they state that the incorporation of cellulose fibres may enhance the stiffness of composite materials.

FLEXURAL PROPERTIES

Figure 6 illustrates the flexural strength and flexural modulus of the kenaf filled composites. At 30 wt. % filler loading, the flexural strength of the 20 mesh core filled composite is slightly higher (29.42 MPa) than the 40 mesh core (23.58 MPa) and bast (25.98 MPa) filled composites.

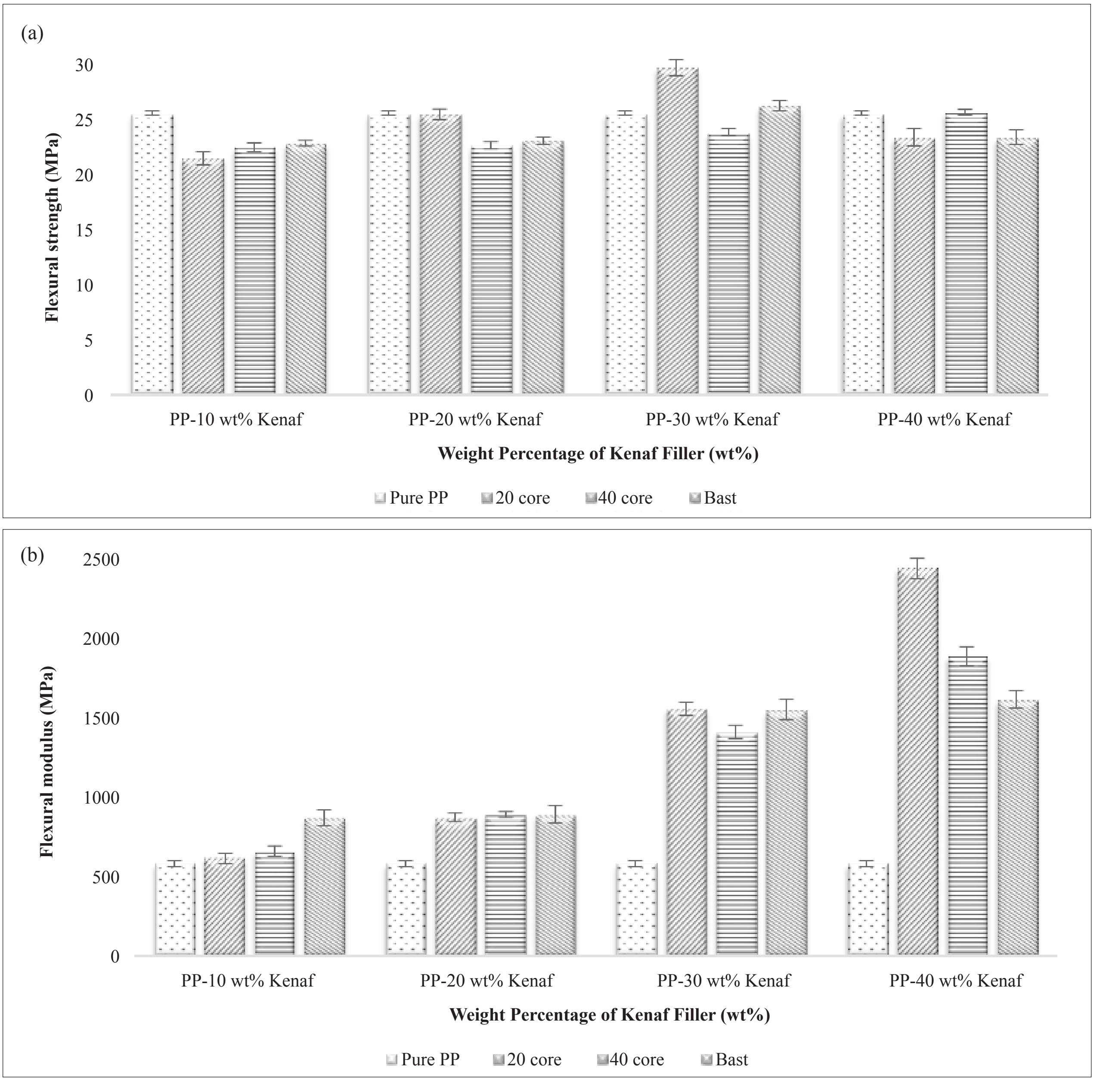


FIGURE 6. (a) Flexural strength and (b) Flexural modulus of kenaf/PP composites at different filler loadings

While the 40 mesh core filled composite showed the highest strength (25.35 MPa) at 40 wt. % filler contents compared with the 20 mesh core and bast filled composites. The results indicate that the cellulose composition in the 20 and 40 mesh core filled composite may increase with an increase in the filler percentage. Where increasing the loading percentage will increase the chemical (cellulose) composition which providing greater strength to the reinforced composite (Khalil et al. 2013).

Furthermore, according to Abdul Khalil et al. (2010) the effect of cell wall lumen of kenaf core fillers with variability in size, shape and structure (i.e. polygonal in shape), will provide much better performance as a reinforcement in the composites structure. Thus, in this study, most core filled composites which have larger lumen diameter and a narrower thickness of the cell wall exhibited much better flexural strength than bast filled composites.

Figure 6(b) shows the 20 mesh core filled composite with a massive increase in modulus strength. The highest value (2423.77 MPa) was achieved at 40 wt. % filler loading. The lowest value of 1600.79 MPa was acquired from the bast filled composite at 40 wt. % filler loading. The results identify that 20 mesh core filled composite is stiffer than the 40 mesh core and bast filled composites.

MORPHOLOGICAL

Figure 7 shows the SEM micrograph of the fractured cross-section of kenaf core and bast filled composites. The SEM micrograph images were selected from the optimal tensile strength of the fractured cross section (area) of both composites at 30 wt. % filler loading for the 20 mesh core and bast filled composite and 40 wt. % for the 40 mesh core filled composite. The 20 mesh core

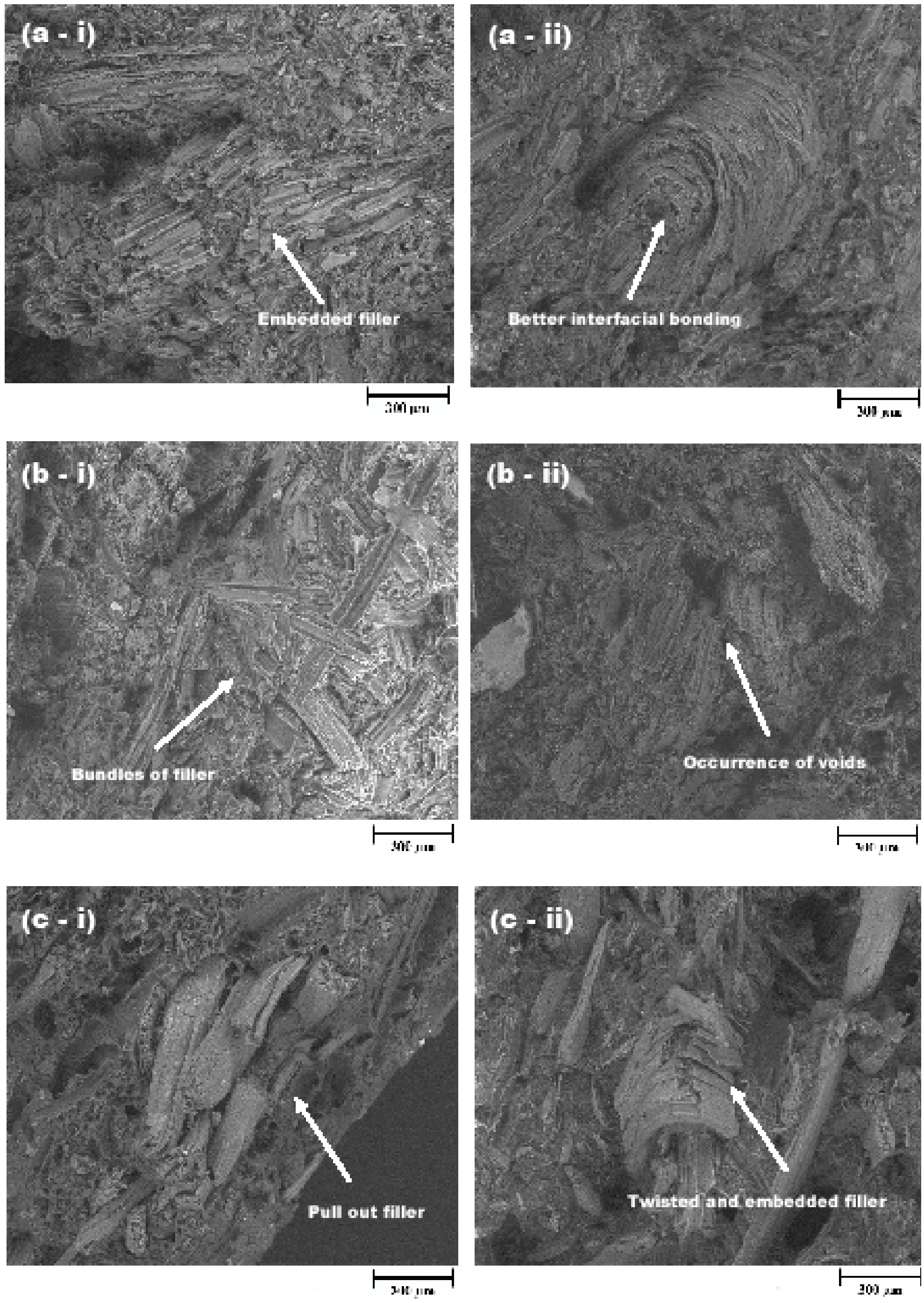


FIGURE 7. SEM micrograph of; (a) 20 mesh core, (b) 40 mesh core and (c) bast filled composite at (i) spot 1 and (ii) spot 2

filled composite revealed much better interfacial bonding than the other filled composites. The excellent interfacial bonding indicates that the core was surrounded by the PP matrix, leading to diffusion and permeation of the PP within the core filler. Furthermore, the core had high absorption properties due to the high porosity of the filler (Paridah et al. 2009; Shibata et al. 2006). This may have attributed to the penetration of the PP matrix and the successful coating on the core surface.

Slight variations were observed for the 40 mesh kenaf core filled composite, in which there was less physical contact and poor matrix wetting between the filler and matrix. The weak adhesion resulted in lower mechanical strength. Furthermore, several of the 40 mesh core filler formed bundles due to the creation of hydrogen bonds in the core material, thereby decreasing the strength of the reinforced composites (Saad & Kamal 2012).

For the kenaf bast filled composites, the SEM micrograph showed that the bast filler was twisted and embedded within the PP matrix. This relatively stronger interfacial bonding resulted in good stress transfer between the matrix and the bast filler. However, some of the bast filler was extracted (pull out) from the surface of the PP matrix as filler loadings increased (Yusoff et al. 2010).

These defects will have adverse effects and hinder the increase of mechanical strength of filled composites. Other researchers have observed that proper processing conditions and the absence of moisture absorption during the preparation of kenaf/PP composite could yield excellent traction of interfacial adhesion between the fibre and matrix, as well as inhibit the formation of any voids (Anuar et al. 2012). Blended with kenaf filler and PP, consistently achieved homogeneity of the composition under proper mixing conditions and sufficient pressure and temperature during the injection process.

Conclusion

In this study, the effect of kenaf filler reinforcement on mechanical and physical properties of injection moulded polypropylene composites was investigated. The filler loadings varied from 10%, 20%, 30% and 40% in weight was applied. Based on the results, the highest tensile strength for kenaf 20 and 40 mesh core filled composites were achieved at 30 wt. % (15.91 MPa) and 40 wt. % (15.76 MPa) of filler loadings, respectively. While for the kenaf bast filled composites, the highest tensile strength of 19.52 MPa was obtained at 30 wt. % filler loading. Thus, the tensile strength results showed that kenaf bast filled composites had the highest values at each loading percentage, compared to kenaf core filled composites.

Instead, kenaf 20 mesh core filled composites was obtained had the highest flexural strength which values were 25 MPa and 29 MPa at 20 wt. % and 30 wt. %, respectively. While kenaf 40 mesh core filled composites had the highest values of 25.35 MPa at 40 wt. % of filler loading compared to kenaf bast filled composites. However, the modulus strength properties for both types of kenaf/PP composites increased with an increase in filler loadings.

In conclusion, the results identified that core filled composites possess mechanical properties comparable to bast filled composites. Therefore, to maximise the use of fibre from natural sources, kenaf core fillers is reasonable to be used to increase the mechanical strength of injection moulded polypropylene composites.

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