Effects of Fiber Content and Processing Parameters on Tensile Properties of Unidirectional Long Kenaf Fiber Reinforced Polylactic-Acid Composite

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ABSTRACT

Recent developments within the field of natural fiber-reinforced polymer composite have led to a renewed interest in fully biodegradable composite or green composite. Interest in using green composites is due to environmental awareness concerns as well as stringent government regulations. The study is aimed to determine the optimum percentage of fiber content for unidirectional long kenaf fiber (LKF) reinforced polylactic-acid composites. In addition, the influence of processing parameters has also been investigated but only for the optimum fiber content. The percentage of kenaf used are 10 wt% to 50 wt% prepared by film stacking method with a hot-

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press machine. A series of tensile tests was performed to obtain the tensile strength, including the Young's modulus of the composite. The 40 wt% fiber composites showed tensile strength, and the Young's modulus increased linearly to 230% and 650%, respectively. Furthermore, for 50 wt% of fiber content, tensile strength and young modulus increased linearly to 244% and 625% compare to pure polymer, respectively. It was found that better tensile properties were achieved with 50%wt fiber composites produced at a temperature of 200°C, 5MPa compression pressure and 5 minutes holding time. Applications for fabricated compoistes are in non-structural to structural with medium loads, especially in the field of aerospace, automotive, and building construction industries.

Introduction

Over the years, the application of natural fibers as reinforcements in polymer composites have increased due to its good specific mechanical properties, lightweight, non-toxic, non-abrasive, low cost, and also environmental friendly properties. There are three types of origin for natural fibers namely animals, plants and minerals. However, plants fiber is the most popular and widely used due to its availability, abundance, and cost. In Malaysia, kenaf cultivation has started early in the year 2000 to replace tobacco plantation. Kenaf has been identified as the new potential crop as it is fast growing, and prone to low pest attacks compared to other plants [1].

The use of kenaf as a reinforcement in synthethic polymers makes the composite semi-biodegradable or partially biodegraded. Therefore, to achieve fully biodegradable composite or green composite, a biodegradable polymer matrix was introduced to be reinforced with kenaf or natural fiber. There are many types of biodegradable polymer from renewable resources and one of them is PLA. PLA or polylactic-acid or sometimes called polylactide is an aliphatic polyester and biocompatible thermoplastic, which can be semicrystalline or totally amorphous in nature. PLA is currently the most promising and popular material with the brightest development prospect and considered as a 'green' eco friendly material. PLA offers excellent properties such as high strength and stiffness and it is being used in several applications, such as consumers packaging, degradable plastic bags, as well as in automotive applications [2].

Mechanical properties of fiber composites can be affected by factors such as length and composition of fiber as well as orientation of the fiber itself. It is known that properties of fiber aligned unidirectionally are much higher than randomly oriented depending on the load applied to the composites. Furthermore, a number of researchers have already reported the properties of fully biodegradable composite products with a variety of fiber types, composition, and orientation as well as the processing methods. Romhany et al. found that unidirectional flax fiber reinforced starch composites strongly improves both stiffness and strength compared to the pure matrix at a fiber content of 40% [3]. Nassiopoulos and Njuguna [4] indicated that flax/PLA can be a promising material to replace traditional choices in load bearing applications with the strength of 72 MPa and stiffness of 13 GPa, respectively. Huda et al. [5] demonstrated that a good mechanical properties laminated composite could be succesfully developed with the surface treated with 40 wt% kenaf fibers reinforced with PLA by using the film-stacking method. On the other hand, Ochi [6] reported that the unidirectional kenaf fiber reinforced PLA composites at 60% fiber content have the highest tensile and flexural strength respectively. According to Shah [7], the manufacturing technique can also have a noticeable effect on the natural fiber composites' mechanical properties, particularly in the case of composites in unidirectional fiber orientation. Ku et al. [8] revealed that the processing conditions are one of the factors that significantly influence the properties and characteristics of the polymer matrix composites products. Kumar and Balachandar [9] studied the effects of hot press forming mould pressure, temperature, and holding time on flexural strength of Glass/PP composites through Box Behnken analysis and found that the optimum temperature is 220°C with low holding time. Kandar and Akil [10] studied the effects of moulding temperature, pressure, and time on the impact strength of woven flax fiber reinforced PLA composites. They found an increase in impact strength with increasing moulding temperature but at minimum moulding pressure and time. In the work of Kiran et al [11], processing temperature has the most significant effect on mechanical properties that can be measured in his work. Hence, an attempt is made to analyze the effects of hot press process parameters on mechanical properties of unidirectional long kenaf fiber/PLA composites. Although there are many previous literatures reported concerning fully biodegradable composites, most of them were focused mainly on fiber treatments as well as optimum fiber composition. Yet, very few studies have been carried out on the effects of processing parameters on tensile properties of long natural fiber in a unidirectional orientation.

Therefore, this study aims to determine the optimum percentage of long kenaf fiber content in unidirectional orientation for fabrication of fully biodegradable composites. The effects of fiber content on strength and Young's modulus of the composites were discussed. In addition, the influences of hot press processing parameters on tensile properties of composites for optimum fiber content were also investigated.

Experimental

Materials

Continuous long fiber of kenaf from bast undergone water-retting process were supplied locally by Innovative Pultrusion Sdn. Bhd. Long kenaf fiber (LKF) bundles were combed manually and aligned in a single direction as in Figure 1(a) before being cut to the length ~175 mm for fit with the size of platen of a hot press mould. This is to produce long and clean fibers; and untangle the strong bonding of individual fibers [12]. PLA pellets and microfine powders in the size of 20 μm from Shenzun Esun China Ltd as in Figure 1 (b) and 1 (c) were also supplied by Innovative Pultrusion Sdn. Bhd. The properties of the pellets can be found in Table 1. LKF and PLA were dried in an oven at 80 °C 24 hours prior to use. PLA pellets, powders, and fibers in five different kenaf contents (10, 20, 30, 40 and 50 wt.%) were measured to determine the weight ratio. At 60% fibers percentage, the fabrication could not be processed due to the insufficient matrix to cover the composite plates. For matrix calculation, the ratio of pellets and powders used are 70:30. A solid pair of PLA films were prepared by hot-pressing the pellets at 190 °C for 7 minutes under 5 MPa pressure and stored in the laboratory under ambient conditions prior to composite fabrication.

Table 1: Typical properties of PLA pellets [MSDS]

Properties	Details
Melt Index at 190 ^{0}C/2.16kg	10-12 g/10 min
Density	1.24 g/cm^3
Yield Strength	56 MPa
Elongation at Break (%)	2
Flexural Strength	78 MPa
Flexural Modulus	3110 MPa
Impact Strength	4.4 kJ/m^2

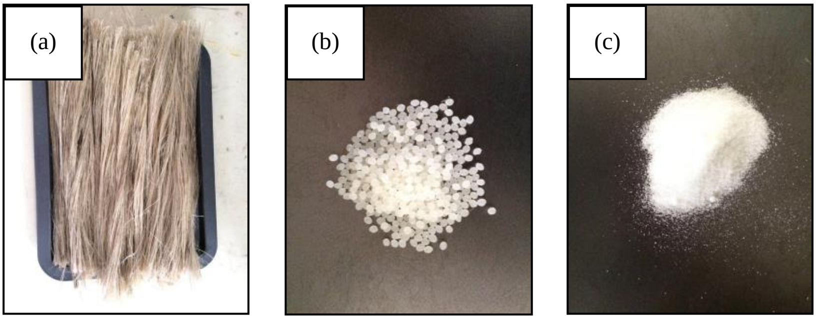


Figure 1: (a) Combed and aligned LKF (b) PLA pellets (c) PLA powder

Composite Fabrication

At first, the fabrication of the kenaf reinforced PLA composites was successfully achieved. However, prior to tensile tests, the fibers delamination occured due to interfacial adhesion problems as shown in Figure 2 below. Therefore, microfine PLA powder was sprinkled between the fibers and films. This is to improve the interface between fibers as well as the matrix. In addition, the powders also act as a binder to ensure that the fibers are relatively parallel and avoiding from uneven fiber distributions.

Figure 2: Fibers delamination from the composite sample

LKF were sprinkled with PLA powder and shaken manually in the container. Afterwards, LKF were distributed evenly between two PLA solid films. Stacked materials were hot pressed with LKF at 190^{o}c for 5 min while keeping a constant pressure of 5 MPa using a 50 tonne hot press machine. A similar cooling time approach was employed to ensure the cooling and solidification of the prepared composites. A schematic diagram for the layout of composite fabrication is illustrated in Figure 3, in which the teflon sheets are non-stick material layers to facilitate the composite release. Composite plates were cut to standard rectangular shapes using a shearing machine for tensile test.

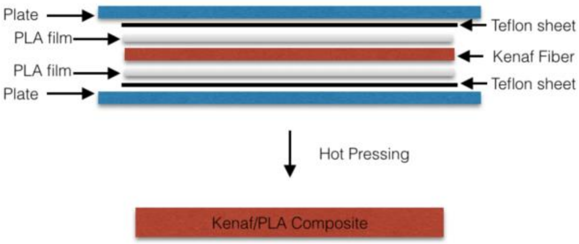


Figure 3: Schematic diagram of the composite fabrication

The optimum fiber content obtained in this study is 50%wt. Therefore, 50%wt of LKF with 50%wt matrix is used throughout the experiment for further experimental investigation on processing parameters. The parameters setting for the hot press are temperatures 190°C and 200°C; pressures 3 and 7 MPa; and heating times of 5 and 10 minutes.

Tensile Test

The tensile tests were performed according to the ASTM D3039 standard on Zwick Roell universal tensile test machine with 100kN load. Five samples were tested per material at a 5 mm/min cross-head velocity with 65 mm gauge length. The length and width of the samples were 115 mm and 20 mm respectively with a 2 mm thickness.

Results and Discussion

Effects of Fiber Content on Tensile Properties

The average tensile strength and Young's modulus of LKF/PLA composite specimens with different fiber contents and pure PLA specimens are presented below in Figure 4 and 5, respectively.

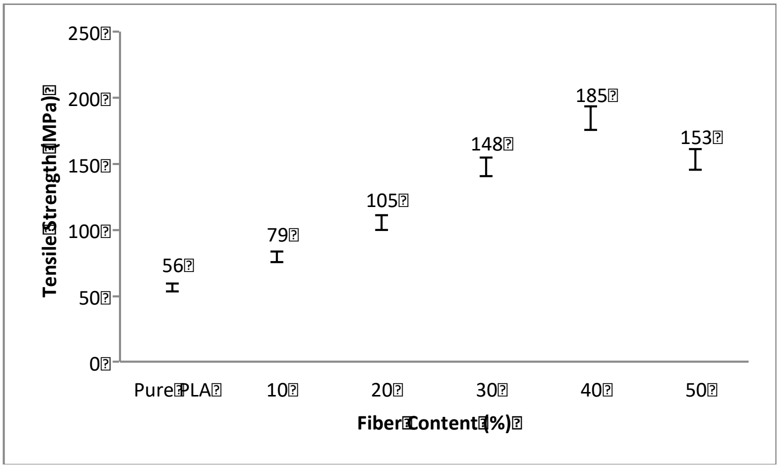


Figure 4: Tensile strength of LKF/PLA composite

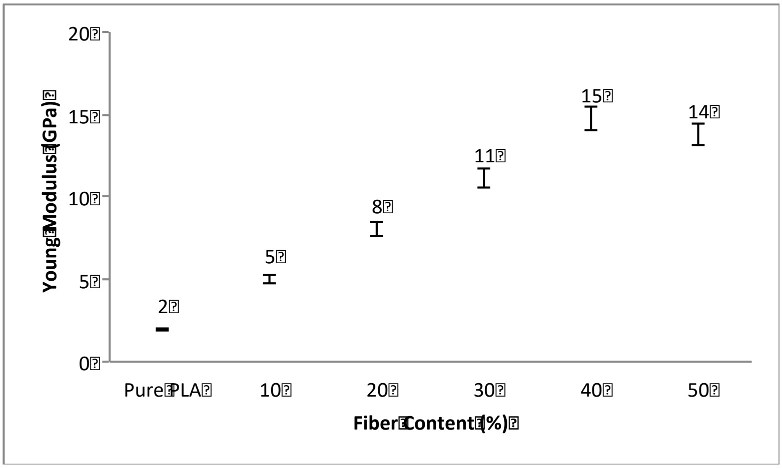
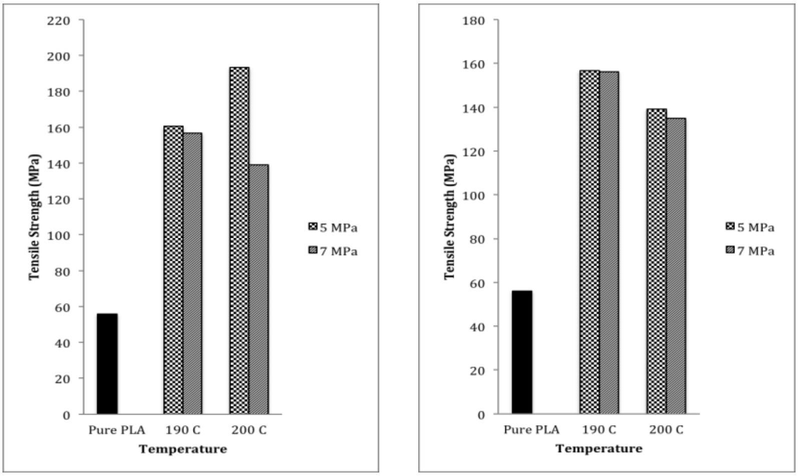


Figure 5: Young’s modulus of LKF/PLA composite

The results show that the increase in tensile properties depends strongly on the addition of LKF even at a fiber content as low as 10 wt%. It was almost triple the tensile strength and more than triple the Young’s modulus of the PLA by incorporating LKF. Both tensile strength and Young modulus’s of LKF/PLA composites increased linearly with fiber content up to 40 wt% with 185 MPa and 15 GPa, respectively. The increase in the Young’s modulus with fiber content was in line with the tensile strength. This indicated that a good fibers/matrix surface compatibility and good stress transfer between the fibers and the matrix. In addition, the same polarity (hydrophilic) of both fiber and matrix may also have contributed to this compatibility. It is known that PLA is hydrophilic, as are natural fibers [13]. The results obtained were similar and in agreement with the reported tensile properties for the long natural fiber reinforced PLA composites by Ochi [6]. However, as fiber content increased to 50 wt%, the tensile properties of LKF/PLA decreased to 153 MPa. In addition, several authors have observed a decrease in composite strength with higher fiber content. Ibrahim et. al [14] observed a decrease in tensile strength from 32 to 16 MPa when the fiber composition is more than 30%wt. Hu and Lim [15] also described a reduction in tensile strength with 54%wt fibres instead of 44%wt fibres for hemp/PLA composites. This could be due to the worse fiber wetting that reduces the composite tensile strength caused by the weak fiber/matrix bonding [16]. Therefore, it is suggested that the processing conditions could be emphasized with regards to the higher fiber content. According to Mukherjee and Kao [13], the mechanical properties of the fiber reinforced PLA composites depend on the fibre matrix adhesion, which may also be directly related to the processing conditions. The processing conditions affect the degree of fiber wetting, crystallinity, and composite porosity. In the author's point of view, a higher processing time or more heating time is required for the PLA to melt in order to fully disperse the entire fiber in the case of higher fiber content. This is to make sure of the fiber's wettability as well as improved impregnation.

Effects of Processing Parameters on Tensile Properties

The results of the unidirectional LKF/PLA with 50%wt mechanical properties in terms of tensile strength and Young's modulus at 5 minutes heating time and 10 minutes heating time were presented in Figure 6 and 7 below. It can be seen that the tensile results are strongly influenced by the processing parameters of the hot-press. In addition, the composites' mechanical properties also depend on the unidirectional LKF which gave higher results compared to pure PLA matrix. An increment of up to 244% in tensile strength and 625% in Young's modulus can be achieved with kenaf as the reinforcement compare to pure polymer. Hence, this indicates the good adhesion and stress transfer efficiency between kenaf fiber and PLA matrix [17].



(a) 5 mins heating time (b) 10 mins heating time

Figure 6: Results of tensile strength for unidirectional LKF/PLA

Figure 6 (a) shows the tensile strength at 5 minutes heating time and Figure 6 (b) shows the tensile strength at 10 minutes heating time. At lower heating time and pressure, the tensile strength is significantly increased when the temperature increased from 190ºC to 200ºC. It can be suggested that the impregnation of PLA matrix into kenaf fibers increased and is favorable at 200ºC processing temperature. However, the decrease in tensile strength is obvious at a higher pressure with the increase in temperature. This is due to the higher the pressure, the matrix tends to flow out from the mold, which results in a lack of matrix as well as the decreasing of fiber strength. On the other hand, at prolonged heating time; there is a slight decrease in tensile strength for the same temperature even at different pressures. It is noticeable that tensile strength decreases when the temperature increases. When comparing the pressure, the lower pressure gives the higher tensile strength. High temperature for a certain period of time with high pressure causes the quantity of PLA to overflow from the mould and thermal decomposition of the kenaf fiber [18]. The lack of matrix will also result in strength decrease. This indicates that the tensile properties are influenced by processing parameters such as temperature, pressure, and heating time.

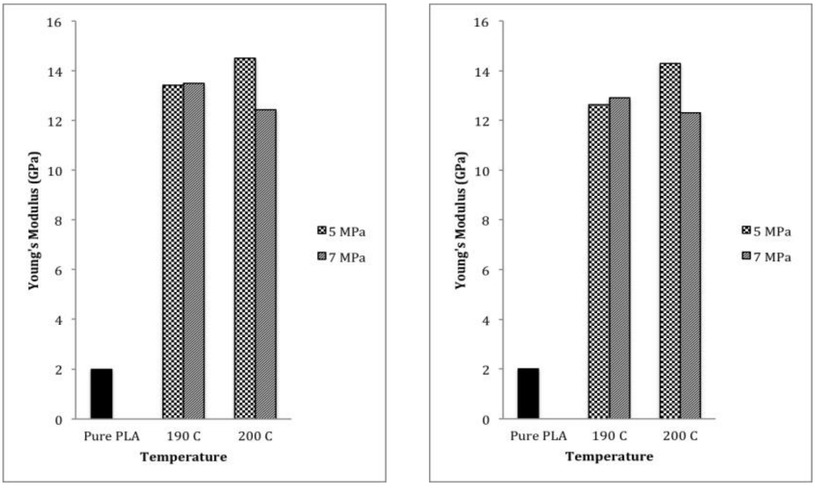


Figure 7: Results of Young's modulus for unidirectional LKF/PLA

Figure 7 (a) and 7 (b) represent the graph of Young's modulus at 5 minutes holding time and 10 minutes heating time respectively. The findings show the same trend even at different heating times. In the meantime, the highest Young's modulus can be obtained with a higher temperature at 200ºC but at lower pressure and heating time. According to Pickering et al. [19], the viscosity of the matrix during pressing and heating needs to be carefully controlled, to make sure the matrix is impregnated fully into the space between fibers. With the increase in wettability of the matrix, the interfacial adhesion between kenaf and matrix is improved. In addition, the Young's modulus may also be influenced by the long kenaf fiber which has a higher modulus than pure PLA polymer.

Conclusions

PLA could be reinforced with a maximum of 50 wt% fibers using the film stacking method. The composites tensile properties were improved up to 230% for strength and 650% for modulus compare to pure PLA polymer. The effects of hot pressing parameters on tensile properties of 50 wt% aligned LKF/PLA composites are also experimentally investigated. With a 50 wt% fiber content, the higher temperature would be required in order to improve the fiber/matrix impregnation. From the study, LKF/PLA composites produced with hot press at 200°C and 5 MPa for 5 minutes gives an excellent tensile strength and Young's modulus with 244% and 625% improvements respectively. These also indicate that kenaf fiber has suitable properties to be used in engineering as fiber reinforced polymer composites especially in structural applications at low and medium loads. For future work, a response surface methodology is recommended to determine the optimum process parameters for yielding good mechanical properties of the composite.

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References

[1] M. Z. A. Thirmizir, Z. a M. Ishak, R. M. Taib, S. Rahim and S. M. Jani,
"Natural Weathering of Kenaf Bast Fibre-Filled Poly(Butylen Succinate) Composites: Effect of Fibre Loading and Compatibiliser Addition," *J. Polym. Environ.*, vol. 19, no. 1, pp. 263–273 (2011).

[2] M. Jonoobi, J. Harun, A. P. Mathew and K. Oksman, "Mechanical properties of cellulose nanofiber (CNF) reinforced polylactic acid (PLA) prepared by twin screw extrusion," *Compos. Sci. Technol.*, vol. 70, no. 12, pp. 1742–1747 (2010).

[3] G. Romhany, J. Karger-Kocsis, and T. Czigany, "Tensile fracture and failure behavior of thermoplastic starch with unidirectional and cross-ply flax fiber reinforcements," *Macromol. Mater. Eng.*, vol. 288, no. 9, pp. 699–707 (2003).

[4] E. Nassiopoulos and J. Njuguna, “Thermo-mechanical performance of poly(lactic acid)/flax fibre-reinforced biocomposites,” *Mater. Des.*, vol. 66, pp. 473–485 (2015).

[5] M. S. Huda, L. T. Drzal, A. K. Mohanty, and M. Misra, “Effect of fiber surface-treatments on the properties of laminated biocomposites from poly(lactic acid) (PLA) and kenaf fibers,” *Compos. Sci. Technol.*, vol. 68, no. 2, pp. 424–432 (2008).

[6] S. Ochi, “Mechanical properties of kenaf fibers and kenaf/PLA composites,” *Mech. Mater.*, vol. 40, no. 4–5, pp. 446–452, (2008).

[7] D. U. Shah, “Developing plant fibre composites for structural applications by optimising composite parameters: A critical review,” *J. Mater. Sci.*, vol. 48, pp. 6083–6107 (2013).

[8] H. Ku, H. Wang, N. Pattarachaiyakoop and M. Trada, “A review on the tensile properties of natural fiber reinforced polymer composites,” *Composites Part B: Engineering*, vol. 42. pp. 856–873 (2011).

[9] B. S. Kumar and S. Balachandar, “A Study on the Influence of Hot Press Forming Process Parameters on Flexural Property of Glass/PP Based Thermoplastic Composites Using Box-Behnken Experimental Design,” *ISRN Mater. Sci.*, vol. 3, no. 4, pp. 1–6 (2014).

[10] M. I. M. Kandar and H. M. Akil, “Application of Design of Experiment (DoE) for Parameters Optimization in Compression Moulding for Flax Reinforced Biocomposites,” *Procedia Chem.*, vol. 19, pp. 433–440 (2016).

[11] G. B. Kiran, K. N. S. Suman, N. M. Rao and R. U. M. Rao, “A study on the influence of hot press forming process parameters on mechanical properties of green composites using Taguchi experimental design,” *Int. J. Eng. Sci. Technol.*, vol. 3, no. 4, pp. 253–263 (2011).

[12] M. Bernard, A. Khalina, A. Ali, R. Janius, M. Faizal, K. S. Hasnah, and A. B. Sanuddin, “The effect of processing parameters on the mechanical properties of kenaf fibre plastic composite,” *Mater. Des.*, vol. 32, no. 2, pp. 1039–1043 (2011).

[13] T. Mukherjee and N. Kao, “PLA Based Biopolymer Reinforced with Natural Fibre : A Review,” *Polym. Environ.*, vol. 19, pp. 714–725 (2011).

[14] Nor Azowa Ibrahim, Kamarul Arifin Hadithon and K. Abdan, “Effect of Fiber Treatment on Mechanical Properties of Kenaf Fiber-Ecoflex Composites,” *J. Reinf. Plast. Compos.*, vol. 29, no. 14, pp. 2192–2198 (2010).

[15] R. Hu and J.-K. Lim, “Fabrication and Mechanical Properties of Completely Biodegradable Hemp Fiber Reinforced Polylactic Acid Composites,” *J. Compos. Mater.*, vol. 41, no. 13, pp. 1655–1669 (2007).

[16] N. Graupner and J. Müssig, "A comparison of the mechanical characteristics of kenaf and lyocell fibre reinforced poly(lactic acid) (PLA) and poly(3-hydroxybutyrate) (PHB) composites," *Compos. Part A Appl. Sci. Manuf.*, vol. 42, no. 12, pp. 2010–2019 (2011).

[17] M. A. Sawpan, K. L. Pickering and A. Fernyhough, "Flexural properties of hemp fibre reinforced polylactide and unsaturated polyester composites," *Composites Part A: Applied Science and Manufacturing*, vol. 43. pp. 519–526 (2012).

[18] S. Kobayashi and K. Takada, "Processing of unidirectional hemp fiber reinforced composites with micro-braiding technique," *Compos. Part A Appl. Sci. Manuf.*, vol. 46, no. 1, pp. 173–179 (2013).

[19] K. L. Pickering, M. G. A. Efendy and T. M. Le, "A review of recent developments in natural fibre composites and their mechanical performance," *Compos. Part A Appl. Sci. Manuf.* vol. 83, pp. 98-112. (2015)