

Mechanical Properties of Polylactic Acid (PLA) Green Composites Reinforced by Kenaf Bast and Core Fibers

S.N. Surip, W.N.R. Wan Jaafar, M.A. Tarawneh, N.N Azmi

Abstract—Polylactic acid (PLA) green composites were fabricated using melt compounding and compression moulding. Kenaf bast and core fibres had undergone chemi-mechanical treatment before use. PLA and kenaf fibres were mixed at different fibre loadings (2%, 4% and 6%) and extruded with three different rotation speeds (60, 70 and 80 rpm). The mechanical properties of kenaf bast composites (KBC) and kenaf core composites (KCC) were studied by performing flexural and impact testing. KBC and KCC treated with 1.0M acid treatment at 60 rpm speed had higher flexural and impact strength. KBC at 6% fibre loading had a higher flexural modulus, which was caused by the stiffness of the fibre incorporated in the PLA. However, KBC with 4% fibre loading has higher flexural strength than 6% fibre loading. In contrast, KCC at 2% fibre loading had the highest flexural modulus and strength. Meanwhile, for impact properties, 4% fibre loading had the optimum strength for both KBC and KCC.

Index Terms—Kenaf bast fibre, kenaf core fibre, polylactic acid, green composites.

I. INTRODUCTION

In recent years, large amounts of petroleum based plastic products have been discarded all around the world and the incineration of these wastes has caused increasing emissions to the environment [1, 2]. With the growing environmental awareness and regulations, biocomposite materials that are compatible with the environment and independent of fossil fuels have been critical issues in developing new products [2, 3].

Recently, bio-based polymers have been introduced as an alternative for petroleum based polymers. One major bio-based polymer is polylactic acid (PLA), which is a thermoplastic polymer that is obtained by either ring opening polymerization of the cyclic lactide dimer or by condensation of lactic acid [1, 3, 4]. Some environmental benefits of PLA are its biodegradability, low production energy, reduction of greenhouse gas production and renewability of the base material [5, 6]. In addition to these environmental benefits, PLA has been proven to have good mechanical properties, thermal plasticity and biocompatibility, and is readily

fabricated. However, the inherent brittleness, stiffness and low dimensional stability restrict the application of PLA in various applications [1, 7]. The incorporation of natural fibre as a reinforcing agent is one of the promising methods to improve these properties.

Natural fibres possess high specific strength and stiffness, have a lower cost, low density, are flexible, harmless, represent an abundant renewable source, reduce wear on machinery, are biodegradable and are capable of reinforcing the polymer matrix to produce composites with the desired properties [2, 8, 9]. Kenaf is one of the fastest growing plants that can be easily grown around the world. Despite these advantages, one major problem of dealing with natural fibre is the incompatibility between the hydrophilic natural fibres and the hydrophobic polymer matrix, which result in poor properties of the composites [2].

The compatibility between the reinforcing agent and polymer is a critical factor in determining the properties of the resultant products. Thus, in order to improve the properties of products, the interaction between the natural fibres and polymer must be improved. The treatments of natural fibres seem to improve the compatibility between the fibres and the polymer. A number of methods have been reported by various researchers for this, such as graft copolymerization, silane treatments, mercerization, esterification, use of a compatibilizer, plasma treatments, electric discharge, benzylation and cyano ethylation [2, 10]. The treatment techniques are used to improve the hydrophilic nature of the fibre and make it compatible with the hydrophobic nature of the polymer. Good interfacial bonding determines the good adhesion between the fibre and the polymer.

Fibre treatment was purposely done to remove the non-cellulosic material present on the surface of the fibre. The removal of these materials improves fibrillation, which, in turn, increases the number of bonding sites on the surface of the fibre. Alkali treatment removes the lignin, wax and oils that cover the fibre cell walls, depolymerises the cellulose and causes fibrillation [11]. It increases the surface area of the fibres to make polysaccharides more susceptible to hydrolysis [12]. Li et al. [13] and Mwaikambo and Ansell [14] added that alkali treatment interrupts the hydrogen bonding in the fibre structure that consequently increases the surface roughness of the fibre. The removal of the waxy layer on the fibre surface enhances the interfacial adhesion between the fibre and the polymer [15]. In addition, acid treatment hydrolyses the hemicelluloses and pectin by breaking down the polysaccharides to simple sugars, and hence, releases the cellulose fibre [12].

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Dilute acid treatment was carried out to remove acetyl group of hemicelluloses that hinder the cellulose structure. The acetyl group might be the barrier for the cellulose structure to form hydrogen bonding with other substances [16, 17]. Therefore, other than increasing the surface area of the fibre, deacetylation was necessary [18] for the preparation of the fibre polymer bonding site.

The alkali treatment of the kenaf fibre removed the natural and artificial impurities, thereby producing a rough surface. In addition, the removal of the acetyl groups by acidic treatment exposed the binding side of the cellulose [16, 17]. Thus, good interfacial bonding strength was achieved between the treated fibre and polymer, which determined the good impact properties of the composites. However, Rajeev [19] reported that the impact strength is not directly related to the adhesion between the fibre and the polymer as it depends on the perfection of the polymer-fibre bundles including the alignment of the fibres and the imperfections, such as voids. The impact strength also relates to the toughness of the composites that depends on the fibre, polymer and their interfacial adhesion [20]. The results suggest that the treatment of the fibre was effective in resisting the crack propagation during impact testing due to the improvement of the interfacial adhesion between the fibre and polymer.

This study deals with the preparation and evaluation of the properties of kenaf fibre reinforced PLA composites. The effect of acid concentration, rotation speed and fibre loading on the properties of bio-composites are studied. The mechanical performance is analysed to determine the optimum acid concentration, rotation speed and fibre loading on the properties of the composites. SEM reveals the surface morphology of the composites in failure.

II. EXPERIMENTAL

Materials

Poly lactic acid (PLA), ESUNTM, extrusion grade was supplied by Shenzhen Bright China Industrial Co., Ltd., China with a density of 1.31 g/cm³. Kenaf bast was collected from the Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia (UPM), Serdang. Chemical substances such as sodium hydroxide (NaOH) and hydrochloric acid (HCl) were supplied by the Merck Group Malaysia.

Chemi-Mechanical Treatment

The kenaf bast and core fibre were treated at a fibre to solution ratio of 1:25. The fibres were immersed into a 6% w/w sodium hydroxide (NaOH) solution and rinsed repeatedly to remove the excess NaOH solution. After that, the fibres underwent acidic treatment at three different acid concentrations (0.5M, 1.0M and 1.5M). Both the bast and core fibre were treated similarly. Following that, the bast and core fibre were mechanically beaten to reduce the fibre size. The dried fibres were ground using a Frisch pulverisette before being crushed using a freezer mill, Spex Sample Prep, model 6770 to reduce the fibre size.

Production of Composites

PLA and kenaf fibres were mixed at three different ratios (2%, 4% and 6% loading). Then, the mixture was pre-mixed before being placed in the Prism TSE System Type DSR 28 (No. 966195M02), twin-screw extrusion at a temperature

range of 150°C-160°C. At this point, the rotation speed was altered to 60, 70 and 80 rpm to find the optimum processing parameter. Subsequently, the extrudate was then pelletized using a Prism TSE System cutter (No. 2094). Then, the pellet was placed inside a desiccator before compression moulding. For the determination of treatment concentration, the rotation speed and fibre loading was fixed at 60rpm and 2% loading respectively, while, for rotation speed determination, the treatment concentration and fibre loading used were 1.0M concentration at 2% fibre loading respectively. In the determination of optimum fibre loading, 1.0M treatment concentration and 60rpm speed were used for the processing of the composite. A 150 mm x 150 mm x 3 mm polymer composite was manufactured for each of the properties. The collected pellet was compression moulded by Hung Ta Instrument Co. Ltd Type HT-8122 A (No. 2260) at a temperature of 175°C for 8 minutes. The first 5 minutes was pre-heated and the last 3 minutes was compression moulded at a pressure of 800 psi (50bar). Subsequently, the composites were left to cool under applied pressure. Afterwards, the composites were cut for sample testing. Table 1 shows the notation for the PLA/kenaf composites used in this study.

TABLE 1. Notation for PLA/kenaf composites

Composite	Notation		
		Bast	Core
Treatment concentrations [m]	0.5	KBC0.5	KCC0.5
	1.0	KBC1.0	KCC1.0
	1.5	KBC1.5	KCC1.5
Rotation speed [rpm]	60	KBC60	KCC60
	70	KBC70	KCC70
	80	KBC80	KCC80
Fibre loading [%]	2	KBC2	KCC2
	4	KBC4	KCC4
	6	KBC6	KCC6

Characterization

The flexural test of KBC and KCC was performed using an Instron 1195 Universal Testing Machine (capacity: 100kN) in accordance with ASTM D790-03 and with a crosshead speed of 5mm/min. The dimensions of the specimens were 127 mm x 12.7 mm x 3 mm. At least five samples were tested and analysed.

The impact strength of the KBC and KCC was done using a Ray-Ran RR2500 Universal Pendulum Impact System with references to ASTM D256. The dimensions of the specimens were 63.5 mm x 12.7 mm x 3 mm. At least five samples were tested and analysed. The fracture surface of the KBC and KCC were observed using a Field Emission Scanning Electron Microscope (FESEM) Philips XL40 with an acceleration voltage of 20 Kv at room temperature.

Percentage of crystallinity was determined by the differential scanning calorimeter (DSC) made on Perkin Elmer analyser with a Perkin Elmer thermal analysis system. Samples at approximately 8-10mg were weight and placed inside aluminum pan. Temperature was set from 20 to 200°C with one cycle heating rate of 10°C per minute. The percentage of crystallinity was calculated based on following equation:



$$X_c = [\Delta H_m / \Delta H_m^0 * (1-w)] * 100\%$$

Where ΔH_m is the enthalpy of melting (J/g), ΔH_m^0 is the enthalpy of melting of 100% crystalline PLA, 93.7 J/g and w is the weight percentage of PLA in the composites.

III. RESULTS AND DISCUSSION

Effect of Fibre Treatment on Mechanical Properties of PLA/Kenaf Green Composites

Figure 1 shows the flexural modulus of PLA/kenaf green composites at different treatment concentrations. As shown in Figure 1, the flexural modulus for the treated KBC was higher compared to the untreated KBC. The modulus for the treated KBC increased from KBC0.5 to KBC1.0 with a 16.4% improvement. However, increasing the treatment concentration to 1.5 M resulted in a reduction of the flexural modulus. This was due to the damage to the fibre structure [21] caused by the high concentration of acid used to remove the fibrils which possibly act as a binding site on the surface of the fibre.

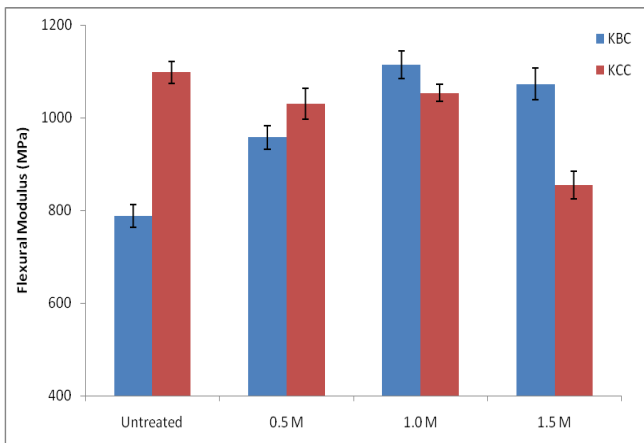


Figure 1. Flexural modulus of PLA/kenaf green composites at different treatment concentrations.

However, a different behaviour was shown for the flexural modulus of the KCC, in that the treated KCC had a slightly lower value than the untreated KCC regardless of the treatment concentration. Flexural modulus is dealing with the tendency of the fibre to bend and the bonding strength between atoms. Based on this result, it shows that untreated core has the more ability to bend than treated core due to its unaffected bonding between fibre-fibre that assists in bending of the fibre and also the composites. However, although the flexural modulus for the treated KCC increased from KCC0.5 to KCC1.0 with a 2.3% improvement, increasing the acid concentration to 1.5 M caused a reduction in the flexural modulus of the KCC, which was probably caused by the action of highly reactive hydrolysis of fibre that tend to damage the surface of the fibre [13, 21].

Figure 2 shows the flexural strength of PLA/kenaf green composites at different treatment concentrations. The flexural strength of the treated KBC had a higher value than the untreated KBC. The fibre surface treatment had an efficient effect on the flexural strength in that the concentration increased to 1.0 M, and the strength also improved by approximately 26.7% compared to the 0.5 M concentration. A further increment of the acid concentration reduced the flexural strength of the KBC. The 1.5 M treatment caused an 18.9% reduction in the flexural strength, which was probably

caused by the wiping-off effect on the fibril presented on the surface of fibre [22].

The KCC flexural strength of the treated fibre had a higher value than the KCC untreated fibre. Similar to the trend for the KBC, the flexural strength of the KCC increased when reinforced with 1.0M treated fibre compared to the 0.5 M and 1.5 M concentration. For KCC1.0, there was a 43.4% improvement in the flexural strength compared to KCC0.5; however, KCC1.5 reduced the strength properties by approximately 17% compared to the strength of the KCC1.0.

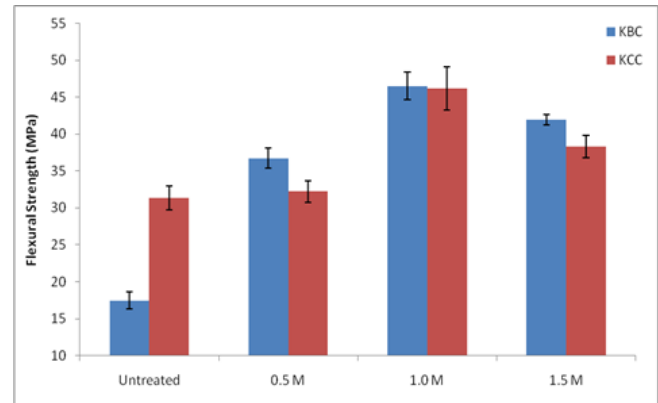


Figure 2. Flexural strength of PLA/kenaf green composites at different treatment concentrations.

Based on Figure 1 and 2, composites reinforced with treated bast and core fibre gives better flexural properties than untreated bast and core fibre with exception for modulus of untreated core fibre. From these results, it can be considered that alkali treatment followed by 1.0M acid concentration gives better flexural properties to the PLA/kenaf green composites. Adel et al. [23] explains that as acid concentration increase, the yield of cellulose also increased which providing higher possible number of bonding site. Increasing in number of bonding site leading to increasing in number of adhesion between fibre and polymer, which consequently improved the mechanical properties as proven by the flexural modulus and strength. However, increasing the acid concentration up to 1.5M, causing the reduction in flexural properties for both KBC and KCC. Bledzki et al. [21] explains that high concentration of acid lead to faster kinetic reaction which difficult to control, thus causing the damage to fibre surface. Difficulties in controlling the reaction lead to the degradation of cellulose caused by delamination effect [24] of the high concentration of acid. Other than that, higher acid concentration causing the gradually decreased in crystallite size and this is due to stronger hydrolysis of fibre at higher concentration has removed the amorphous domains more effectively and lowered the crystalline size [25].

The impact properties of KBC and KCC showed that the treated KBC and KCC had slightly higher values than the untreated KBC and KCC. As shown in Figure 3, both the KBC and KCC had a similar trend in that impact properties was increased from 0.5M treatment concentration to 1.0M treatment concentration of KBC and KCC with 100% and 93% improvement respectively. However, a further increment of concentration to 1.5M treatment caused a drop in the



strength for both the KBC and KCC.

The decrement of the impact strength for 1.5M of treatment concentration for KBC and KCC was found to be 59% and 58%, respectively.

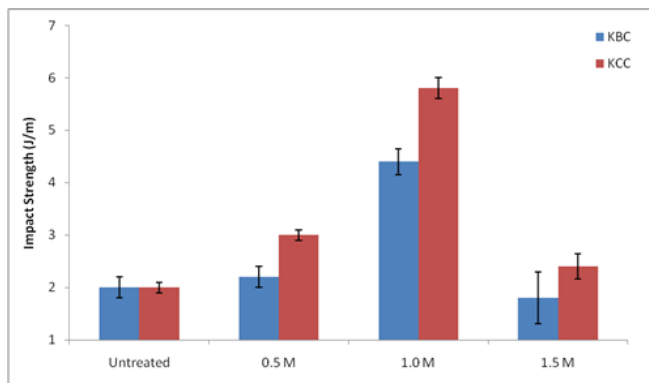


Figure 3. Impact strength of PLA/kenaf green composites at different treatment concentrations.

Notched Izod impact strength is the measurement of energy required for crack propagation. Based on the results in Figure 3, 1.0M treated bast and core fibre shows higher notched impact properties among others. This result is similar with flexural properties. As the fibres become shorter, the composites impact properties is depends on the interfacial adhesion of fibre and polymer, the aspect ratios, distribution and the orientation of fibres [26]. Thus, since the fibres were in micro size, the impact strength could be closely related to the adhesion between the fibre and polymer. With good adhesion, fibres are able to transfer the stress effectively throughout the composites. In addition, the dispersion of the reinforced fibres also assisted in increasing the impact strength of the composites.

Percentage of crystallinity

Table 2 shows the percentage of crystallinity of untreated and treated KBC and KCC at different treatment concentration. Untreated KBC has 30.29% of crystallinity within the composites. Incorporation of treated fibre has increased the percentage of crystallinity regardless of treatment concentration. As listed in Table 2, the percentage of crystallinity was gradually increase by increasing treatment concentration. Bast fibre treated with 0.5M and 1.0M has improves the percentage of crystallinity by 31% and 63% respectively as compared with untreated bast fibre. However, when the concentration is increased to 1.5M, the percentage of crystallinity has slightly reduced.

Similar observation was seen for KCC composites where treated core composites have higher percentage of crystallinity compared with untreated core composites. Untreated KCC shows a total of 34.38% of percentage of crystallinity. Incorporation of 0.5M and 1.0M treated core has increased the percentage of crystallinity by 8% and 38% of increment respectively. Conversely, increasing the concentration causing the reduction on the percentage of crystallinity that might be due to poor interaction between fibre and polymer.

This results was parallel to the mechanical properties of the composites where increased in percentage of crystallinity has increased the mechanical properties of the composites. Increased in compatibility between fibre and matrix by treatment of fibre indicates the reduction in matrix chain

mobility that consequently effect in the percentage of crystallinity [27, 28].

TABLE 2. Percentage of crystallinity of untreated and treated KBC and KCC

Samples	Percentage of crystallinity (%)	
	KBC	KCC
Untreated	30.29	34.38
0.5M	39.53	37.37
1.0M	49.46	47.28
1.5M	48.09	40.40

Effect of Rotation Speed on the Mechanical Properties of the PLA/Kenaf Green Composites

The flexural modulus of the PLA/kenaf green composites at different rotation speeds is shown in Figure 4. The processing of the KBC and KCC at various rotation speeds resulted in different flexural properties. The flexural modulus of the KBC slightly increased from KBC60 to KBC70 with a 7% increment; however, the modulus was reduced for KBC80 with a reduction of approximately 8.5%. The KCC flexural modulus showed a decrease in the modulus as the processing speed increased. The reduction in the modulus was found to be 3.3% and 16.9% from KCC60 for KCC70 and KCC80, respectively.

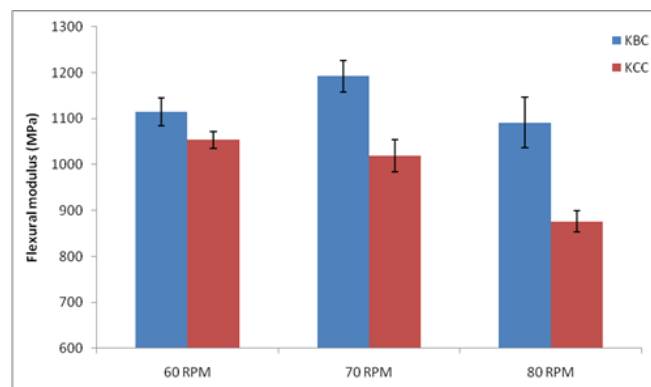


Figure 4. Flexural modulus of PLA/kenaf green composites extruded at different rotation speeds.

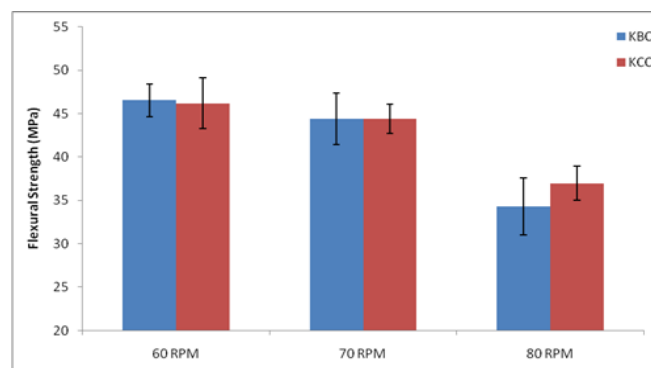


Figure 5. Flexural strength of PLA/kenaf green composites extruded at different rotation speeds.

As shown in Figure 5, the flexural strength of the KBC and KCC processed at different speed shows a similar trend in that 60 rpm for both KBC and KCC was the optimum speed for the processing of composites. The strength of the KBC and KCC was reduced as the speed increased. The strength of KBC70 and KBC80 showed a reduction of 4.6% and 26.3%, respectively, while the strength reduction for KCC70 and KCC80 was found to be 3.9% and 20%, respectively.

Generally, processing with a higher speed will produce higher strength properties. This is due to the high viscosity of polylactic acid, which passes through the higher processing speed and is easily compounded with the fibre. Gamon et al. [29] reported that an increase in shear rate and temperature causes an increased in PLA viscosity, thus making the flow easier. The main cause of this behaviour is the breaking of the molecular chain by the strong shearing action during extrusion and the higher energy of the molecules present at a high viscosity of polymer. Villmow et al. [30] added that higher screw speeds lead to higher shear stress and thus improve the dispersion of the reinforcement agent within the polymer.

Regarding the above statement, a faster screw speed leads to a higher polymer viscosity due to the shear heating and heat conduction created within the extruder that assist in dispersion of the filler creating higher mechanical properties. However, based on Figure 4 and 5, a faster screw speed leads to a lower flexural modulus and flexural strength of the composites. The faster screw speed increases the polymer viscosity by setting temperature and heat created by the barrel. It needs to bear in mind that PLA has the tendency to be thermally degraded under high temperature. Taubner and Shishoo [31] has highlighted that PLA could be thermally degraded due to the process temperature and the residence time in the extruder. It is supported by Lim et al. [32] where processing of PLA at high temperature could lead to major dropping on molecular weight that consequently affect the strength of the composites. Processing with higher screw speed (i.e at 80rpm) creating high shear heating and heat conduction. This will contribute to increasing the temperature within the barrel that lead in degradation of PLA.

Another factor that might reduce the flexural properties concerns the pressure and fibre self-heating while being processed through twin-screw extrusion [29]. Yeh et al. [33] study the effect of screw speed on the mechanical properties of wood flour and polypropylene (PP). They found that increasing screw speed has reduced the mechanical properties of the composites and concluded that addition of energy to the composite during processing results in the fibre failure.

Figure 6 shows the impact properties of PLA/kenaf fibre extruded at different rotation speeds. The impact strength of KBC60 and KBC80 shows the same value which is 4.4 J/m, while KBC70 has a much lower value at about 27% less than that of KBC60. KCC had higher impact properties when extruded with 60 rpm speed with a value of 5.8 J/m. The impact strength was reduced when the rotation speed was increased, in that KCC70 had 3.4 J/m and KCC80 had 3.8 J/m of strength, respectively.

Hypothetically, composites showing fibre pull out will have lower impact strength because generally fibre pull out could dissipate more energy compared to fibre fracture. However, since treatment was done on fibre to improve the bonding properties between fibre and polymer, kenaf/PLA green composites is expected to have better bondability,

where more energy could be dissipated through stress transfer from matrix to the fibre, thus compensating for the energy absorption capability. The low impact toughness could be due to ineffective energy dissipation mechanism at the interface [34]. As shown in Figure 7(a), fibre were embedded into the polymer and fibre pull out was seen as the impact force was applied to the sample. This is a good indicator for composites where good compatibility makes strong interaction between fibre and polymer. The fibre network can be seen in Figure 7(b), where the network fibre acted as a bridge to hold the polymer together and was pulled out as an impact force was applied.

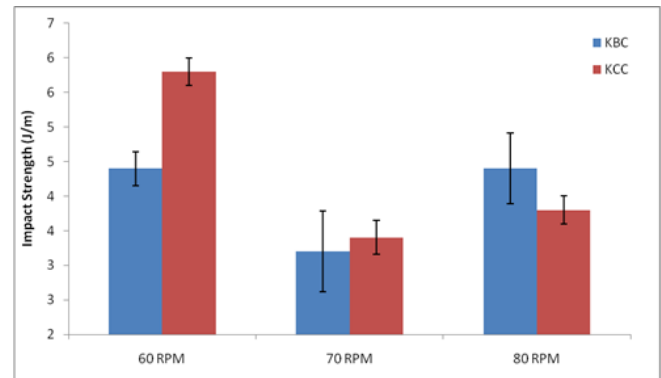


Figure 6. Impact strength of PLA/kenaf green composites extruded at different rotation speeds.

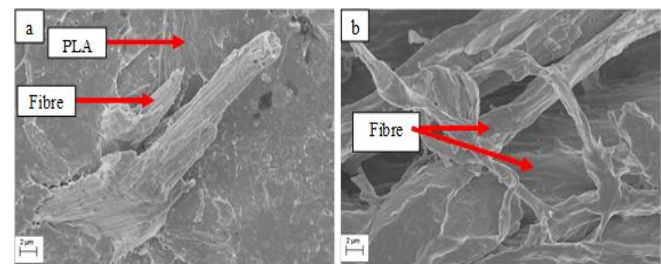


Figure 7. FESEM micrograph for failure surface of impact properties of a) KBC and b) KCC at 60 rpm and 3k magnification

Percentage of crystallinity

Table 3 listed the percentage of crystallinity of KBC and KCC at different rotation speed. The percentage of crystallinity was depending on the dispersion of the filler. As the fibre dispersed, the more percentage of crystallinity could be formed within the composites due to the nucleating effect of the filler.

Processing KBC at 60rpm produced 32.02% of percentage of crystallinity. Increasing speed to 70rpm causing 50% increment in percentage of crystallinity of KBC. Crystallinity production was also related to the dispersion of the filler within the composites where filler act as nucleating site for the crystallinity. From this result, 70rpm has dispersed the fibre within the composites and provide the site for crystallization to occur. However, further increases in speed cause the reduction in percentage of crystallinity. This could be due to the thermal degradation of the matrix while processing that interrupts the polymer molecular chain and crystallinity production.



Similar results were shown for KCC when process at different rotation speed where 70rpm shows higher in percentage of crystallinity. Processing KCC with 60rpm produce 30.83% of crystallinity which is only 8% lower than the crystallinity of 70rpm composites. As previously mention, higher speed caused the reduction of percentage of crystallinity, which can be related to the thermal degradation of the polymer as been described above. Within this parameter determination, dispersion of fibre was the critical factor where speed of the extruder much be carefully selected. Based on the percentage of crystallinity present in Table 3 and mechanical properties, 70rpm has given the best fibre dispersion where in improves its mechanical properties and also the percentage of crystallinity.

TABLE 3. Percentage of crystallinity of KBC and KCC at different rotation speed.

Samples	Percentage of crystallinity (%)	
	KBC	KCC
60rpm	32.02	30.83
70rpm	48.08	33.50
80rpm	23.81	29.67

Effect of Fibre Loading on the Mechanical Properties of the PLA/Kenaf Green Composites

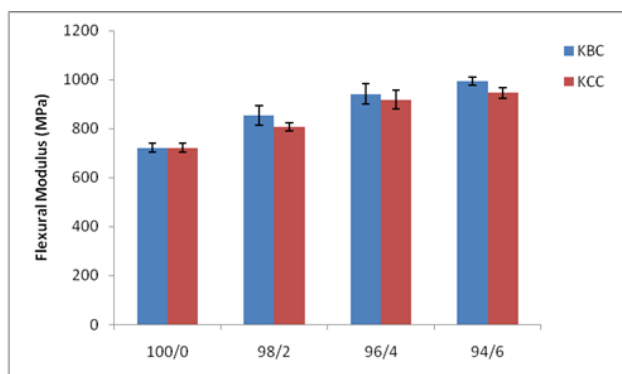


Figure 8. Flexural modulus of PLA/kenaf green composites at different fibre loadings.

Figure 8 shows the flexural modulus of PLA/kenaf fibre green composites at different fibre loading. Fibre reinforced PLA posses higher value in flexural modulus as compared to unreinforced PLA for both KBC and KCC. For KBC, increasing fibre loading caused gradually increasing in flexural modulus. KBC2, KBC4 and KBC6 shows approximately 18%, 31% and 38% increment as compared to unreinforced PLA.

Similarly, kenaf core reinforcement has improved the modulus of unreinforced PLA, increasing the fibre loading has increased the modulus of the composites. KCC2 has 12% improvement in modulus as compared to unreinforced PLA. While, KCC4 and KCC6 has increased the modulus at approximately 27% and 31% respectively from unreinforced PLA.

Natural fibre are originally stiff material [35]. The addition of kenaf fibre into PLA has improved the PLA's stiffness due to the effect of rigid natural fibre [36]. Shibata et al. [37] added that increasing in natural fibre loading causing the linearly enhancement of the modulus. Bledzki et al. [38] using MCC of abaca fibre in their research and concluded that fibres from natural fibres is more flexible which allows in achieving a higher modulus of natural fibre reinforced composites.

Moreover, kenaf fibre reduce the chain mobility of PLA and consequently produced more rigid and tough composites [39].

Figure 9 shows the flexural strength of PLA/kenaf green composites. As shown in Figure 9, the incorporation of fibre regardless of type of fibre used has reduced the flexural strength of the composites. There are gradually decrement of strength as the fibre loading increased which was clearly seen for KCC.

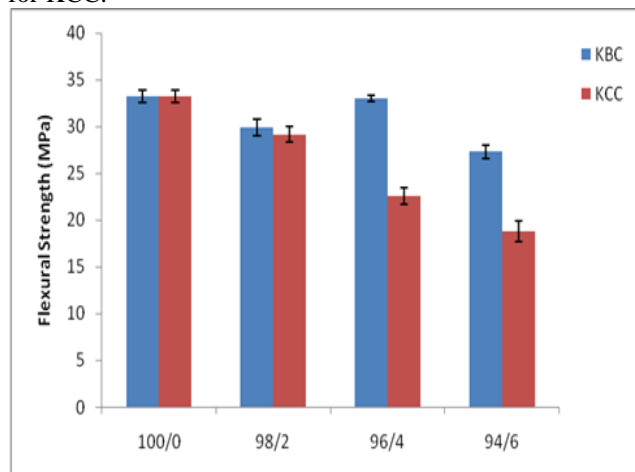


Figure 9. Flexural strength of PLA/kenaf green composites at different fibre loadings.

The decrement in strength of the reinforced composites can be explained by the fibre geometry within the composites. Kenaf fibre could possibly be in an agglomeration form which act as materials flaws [36] that trigger brittle response in the composites that consequently cause early material failure [36]. The addition of higher amount of kenaf fibre causing the linearly reduction on flexural strength. This behaviour was lead by the higher amount of fibre which cause the agglomeration of kenaf fibre within polymeric materials. As explained by Bledzki et al. [38], it was due to the preparation procedure where the compound was agglomerated during extrusion.

Although it is decreased as fibre incorporated into PLA, increasing fibre loading in KBC has increased the flexural strength of the composites until certain amount of loading (i.e 4%). As fibre loading increased, more fibre could be bonded with polymer and more interaction could be formed. Thus, more fibre could be bonded to polymer providing effeciently stress transfer throughout the composites. Nevertheless, further increased in the loading amount caused the reduction in strength where higher amount could lead to fibre agglomeration. Fibre agglomeration act as a stress concentrator that could initiate crack once stress is applied.

KCC has showed a gradual reduction in strength as the fibre loading increased. This behaviour was related to the fibre geometry that agglomerate even after processing. Thus, agglomerated fibre caused in decreasing in required energy for the composites to break and consequently decreasing the flexural strength. In addition to this behaviour, PLA are highly sensative to moisture in that PLA could be hydrolyzed with the present of moisture. Hydrolysis of PLA has much effect on its molecular weight that consequently effect the strength of the composites. Van den Over et al. [40] has studied the effect of moisture to the melocular weight of PLA.



They found that although dried PLA was used, while processing PLA could react with moisture and significantly decreased its molecular weight. The reduction of molecular weight was very visible when fibre were incorporated into PLA.

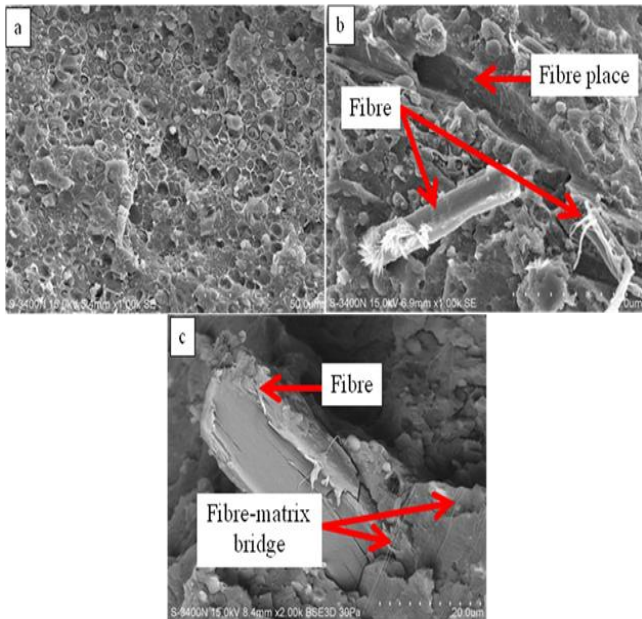


Figure 10. Flexural failure surface of a) neat PLA b) KBC and c) KCC.

Figure 10 shows the SEM micrograph of flexural surface failure for pure PLA, KBC and KCC at 2% fibre loading. Figure 10 (a) shows that unreinforced PLA having the uncompact surface unlike reinforced composites. In Figure 10 (b) and (c), the composites seem to have better adhesion between fibre and polymer and the failure surface visible more compact structure. Figure 10 (b) and (c) also shows the fibre pull out and fibre-matrix bridge after flexural testing. However, the strength of the reinforced composites is slightly lower than the pure PLA where the strength was also depends on the dispersion of the fibre. In addition, present of fibre-matrix bridge indicate that the composites posses ductility behaviour.

Figure 11 shows the impact strength of PLA/kenaf fibre green composites at different fibre loading. PLA impact strength shows an increment after reinforced with kenaf bast fibre. KBC2 has an increment of 17% from the value of virgin PLA impact strength. Increasing the amount of bast fibre loading to 4% loading has improved the impact strength of KBC with a 61% increment from PLA impact strength. However, the KBC6 seemed to drop the impact strength of the composites.

The improvement in impact strength was due to the increasing number of fibres lead to increment in total fibre surface in contact with matrix. Thus, the applied stress could be transferred more effectively due to higher number of surface area [34]. In addition, more fibres wetting by the matrix could present in the composites that consequently enhance to stress transfer across the fibre-matrix interface [34]. Despite that, 4% bast fibre loading are able to compensate the loss in impact strength caused by the presence of fibre producing composites with balanced stiffness and toughness [36]. Reduction on impact strength after being reinforced with higher fibre loading could be caused by the

agglomeration of fibre. Agglomeration became the stress concentrator that brings on the micro-spaces between fibre and polymer and causes several micro-cracks when impact occurs, thus it induces crack propagation and lowering the impact strength [8, 41, 42, 43].

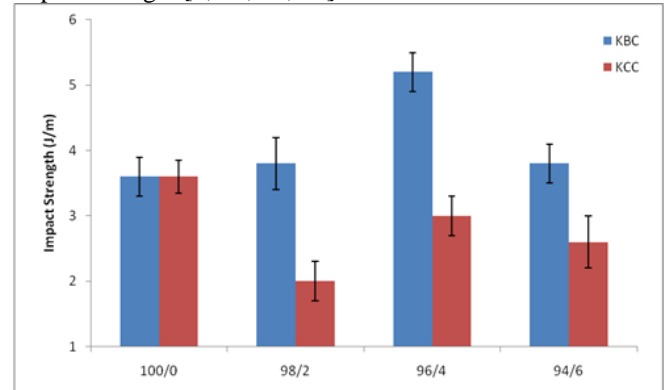


Figure 11. Impact strength of PLA/kenaf fibre green composites with different fibre loadings.

On the other hand, PLA reinforced kenaf core fibre show a declining trend in impact strength. Nevertheless, increasing the core fibre loading has improved the impact strength properties where 50% and 30% improvement have been seen for the KCC4 and KCC6 respectively as compared to the KCC2.

The decrement of KCC in impact strength compared with unreinforced PLA was due to the layered structure of kenaf core. Although the size of the fibre were in micro size, the layered structured were still visible and the space between the layer were unable to reach by the polymer. This section act as a void that consequently failed to behave as a medium for stress transfer. Thus, lowering the impact strength of the composite. Nevertheless, increasing the fibre loading resulting in increasing impact strength. Increasing in fibre loading will consequently increased the number of fibre surface in contact with the matrix, thus effective stress transfer could be achieved [34] and improved the impact strength of the composites. Similar to KBC, higher fibre loading (i.e 6% fibre loading) causing in dropping off the impact strength. Higher fibre loading has a high risk of fibre agglomeration that act as stress concentrator that induce crack propagation and lowering the impact strength [8, 41, 42, 43].

Percentage of crystallinity

Table 4 shows the percentage of crystallinity of KBC and KCC at different fibre loading. Unreinforced PLA has 22.51% of crystallinity. The incorporation of fibre into PLA has increased its percentage of crystallinity regardless of the amount of loading. This is due to the fact that fibre has become the nucleating site for the crystallization to occur.

For KBC, increasing fibre loading until 4% loading has increased the percentage of crystallinity while 6% fibre loading has caused the reduction in the percentage of crystallinity. In contrast, for KCC increasing the amount of fibre loading has reduced its percentage of crystallinity. The reduction in percentage of crystallinity within this parameter was due to the agglomeration of fibre. As the fibre loading increased, the fibre-fibre interaction was likely to form leading to fibre agglomeration within composites.



Agglomeration of fibre provide less nucleating site for crystallization to occur that consequently reduce the percentage of crystallinity. This result was parallel with the mechanical properties which is higher in crystallinity gives higher mechanical strength.

TABLE 4. Percentage of crystallinity of PLA/kenaf green composites.

Samples	Percentage of crystallinity (%)	
	KBC	KCC
100/0	22.51	22.51
98/2	38.50	48.93
96/4	39.01	40.33
94/6	28.55	29.64

IV. CONCLUSION

As a conclusion, in the determination of the optimum acid concentration for fibre treatment, NaOH treatment followed by 1.0M acid treatment has shown higher mechanical properties for both KBC and KCC. The increment from untreated fibre composites was 140% for KBC and 22% for KCC on flexural strength. For impact strength, 1.0M of fibre treatment showed an improvement of up to 120% for KBC and 190% for KCC from the untreated fibre reinforced PLA. For the determination of the optimum rotation speed, both KBC and KCC had better mechanical strength when processed at 60 rpm. On the other hand, KBC at 6% fibre loading showed a 38% improvement in flexural modulus. While, KCC at 2% fibre loading gave 31% increase in flexural modulus compared to unreinforced PLA. However, the flexural strength decrease as the fibre loading increased. The impact strength of KBC and KCC had a higher impact value when reinforced with 4% fibre loading.

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REFERENCES

- Jandas PJ, Mohanty S and Nayak SK. Renewable resource-based biocomposites of various surface treated banana fibre and poly lactic acid: Characterization and biodegradability. *Journal of polymer and environment*. 2012; 20: 583-595.
- Goriparthi BK, Suman KNS and Rao NM. Effects of fiber surface treatments on mechanical and abrasive wear performance of polylactide/jute composites. *Composites: Part A*. 2012; 43: 1800-1808.
- Zhao YQ, Cheung HY, Lau KT, et al. Silkworm silk/poly(lactic acid) biocomposites: Dynamic mechanical, thermal and biodegradable properties. *Polymer degradation and stability*. 2010; 95: 1978-1987.
- Yu T, Ren J, Li S, et al. Effect of fibre surface-treatments on the properties of poly(lactic acid)/ramie composites. *Composites: Part A*. 2010; 41: 499-505.
- Shukor F, Hassan A, Islam MS, et al. Effect of ammonium polyphosphate on flame retardancy, thermal stability and mechanical

- properties of alkali treated kenaf fibre filled PLA biocomposites. *Materials and design*. 2014; 5 (4): 425-429.
- Suprakas SR and Mosto B. Biodegradable polymers and their layered silicate nanocomposites: In greening the 21st century materials world. *Progress in material science*. 2005; 50: 962-1079.
- Pettersson I, Kvien K and Oksman K. Structure and thermal properties of poly(lactic acid)/cellulose whiskers nanocomposites materials. *Composites science and technology*. 2007; 67: 2535-2544.
- Huda MS, Mohanty AK, Drzal LT, et al. "Green" composites from recycled cellulose and poly(lactic acid): Physico-mechanical and morphological properties evaluation. *Journal of materials science*. 2005; 40: 4421-4429.
- Islam MS, Pickering KL and Foreman NJ. Influenced of alkali treatment on the interfacial and physic-mechanical properties of industrial hemp fibre reinforced poly(lactic acid) composites. *Composites: Part A*. 2010; 41: 596-603.
- Masud S, Huda A, Drzal LT, et al. Effect of fibre surface treatments on the properties of laminated biocomposites from poly(lactide acid) (PLA) and kenaf fibres. *Composites science and technology*. 2008; 68: 424-432.
- Mohanty AK, Mishra M and Drzal LT. Surface modification of natural fibre and performance of the resulting biocomposites: An overview. *Composites Interf*. 2001; 8-5: 313-343.
- Alemdar A and Sain M. Isolation and characterization of nanofibers from agricultural residues – Wheat straw and soy hulls. *Bioresource technology*. 2008; 99: 1664 – 1671.
- Li X, Tabil LG and Panigrahi S. Chemical treatment of natural fibre for use in natural fibre-reinforced composites: A review. *Polymer Environment*. 2007; 15(1): 25-33.
- Mwaikambo L and Ansell M. Chemical modification of hemp, sisal, jute and kapok fibre by alkalization. *Journal of applied polymer science*. 2002; 84: 2222-2234.
- Yousif BF, Shalwan A, Chin CW, et al. Flexural properties of treated and untreated kenaf/epoxy composites. *Materials and design*. 2012; 40: 378-385.
- Chang VS and Holtzaple MT. Fundamental factors affecting biomass enzymatic reactivity. *Applied Biochemistry and Biotechnology*. 2000; 84–86:5–37.
- Teixeira LC, Linden JC and Schroeder HA. Simultaneous saccharification and cofermentation of peracetic acid-pretreated biomass. *Applied Biochemistry and Biotechnology*. 2000; 84–86:111–127.
- Yang B, Dai Z, Ding SY, et al. Enzymatic hydrolysis of cellulose biomass. *Biofuels*. 2011; 2:421–450.
- Rajeev K. Kenaf-reinforced polypropylene composites. Department of Chemical Engineering. Michigan State University. 1996.
- Masud SH, Lawrence TD, Amar KM, et al. Chopped glass and recycled newspaper as reinforcement fibers in injection molded poly(lactic acid) (PLA) composites: A comparative study. *Composites science and technology*. 2005; 66: 1813-1824.
- Bledzki, AK, Mamun, AA, Lucka-Gabor, M, Gutowski, VS. The effects of acetylation on properties of flax fibre and its polypropylene composites. *eXPRESS polymer letters*. 2008; 6: 413-422.
- Mohd Edeerozey, AM, Akil, HM, Azhar, AB, Zainal Ariffin, MI. Chemical modification of kenaf fibers. *Materials letters*. 2007; 61: 2023-2025.
- Adel, AM, Abd El-Wahab, ZH, Ibrahim, AA, Al-Shemy, MT. *Bioresource technology*. 2010; 110: 4446-4455.
- Guo, J, and Catchmark, JM. Surface area porosity of acid hydrolyzed cellulose nanowhiskers and cellulose produced by *Gluconacetobacter xylinus*. *Carbohydrate polymers*. 2012; 87: 1026-1037.
- Samir, MASA, Alloin, F, Paillet, M, Dufresne, A. Tangling effect in fibrillated cellulose reinforced nanocomposites. *Macromolecules*. 2004; 37: 4313-4316.
- Corrales, F, Vilaseca, F, Llop, M, Gironos, J, Mandez, JA, Mutje, P. Chemical modification of jute fibres for the production of green composites. *Journal of hazard materials*. 2007; 144: 730-735.
- Jandas, PJ, Mohanty, S, Nayak, SK. Surface treated banana fiber reinforced poly (lactic acid) nanocomposites for disposable application. *Journal of cleaner production*. 2013; 52: 392-401.
- Sarasini, F, Puglia, D, Fortunaati, E, Kenny, JM, Santulli, C. Effect of fibre surface treatments on thermo-mechanical behaviour of poly (lactic acid)/Phormium Tenax composites. *Journal of polymer environment*. 2013; 21: 881-891.

29. Gamon G, Evon P and Rigal L. Twin-screw extrusion impact on natural fibre morphology and material properties in poly(lactic acid) based biocomposites. *Industrial crops and products*. 2013; 46: 173-185.
30. Villmow T, Pötschke P, Pegel S, et al. Influence of twin-screw extrusion conditions on the dispersion of multi-walled carbon nanotubes in a poly(lactic acid) matrix. *Polymer*. 2008; 49: 3500-3509.
31. Taubner, V, Shishoo, R. Influence of processing parameter on the degradation of poly(L-lactide) during extrusion. *Journal of applied polymer science*. 2001: 79: 2128-2135.
32. Lim, LT, Auras, R, Rubino, M. Processing technologies for poly(lactic acid). *Progress in polymer science*. 2008: 33: 820-852.
33. Yeh, SK and Gupta, RK. Improved wood-plastic composites through better processing. *Composites: Part A*. 2008: 39: 1694-1699.
34. Wong, KJ, Nirmal, U, Lim, BK. Impact behaviour of short and continuous fibre-reinforced polyester composites. *Journal of reinforced plastic and composites*. 2010: 29 (23): 3462-3474.
35. Zampaloni M, Pourboghraf F, Yankovich SA, et al. Kenaf natural fibre reinforced polypropylene composites: A discussion on manufacturing problems and solutions. *Composites: Part A*. 2007; 38: 1569-1580.
36. Balakrishnan H, Hassan A, Wahit MU, et al. Novel toughened polylactic acid nanocomposites: Mechanical, thermal and morphological properties. *Materials and design*. 2010; 31: 3289-3298.
37. Shibata, M, Ozawa, K, Teramoto, N, Yosomiya, R, Takeishi, H. Biocomposites made from short abaca fibre and biodegradable polyester. *Macromolecule material engineering*. 2003: 288: 35-43.
38. Bledzki, AK, Jaszkwicz, A, Scherzer, D. Mechanical properties of PLA composites with man-made cellulose and abaca fibers. *Composites: Part A*. 2009: 40: 404-412.
39. Ismail H, Abdullah AH, and Abu Bakar A. Kenaf core reinforced high-density polyethylene/soya powder composites: The effect of filler loading and compatibilizer. *Journal of reinforced plastics and composites*. 2010; 29: 2489-2497.
40. Van den Oever, M.J.A., Beck, B., Müssig, J., 2010. Agrofibre reinforced poly(lactic acid) composites: Effect of moisture on degradation and mechanical properties. *Composites: Part A*. 41: 1628-1635.
41. Shyang CW and Kuen IS. Flexural, morphological and thermal properties of polylactic acid/organo-montmorillonite nanocomposites. *Polymer composite*. 2008; 16 (4): 263.
42. Zhao Q, Tao J, Yam RCM, et al. Biodegradable behaviour of polycaprolactone/rice husk ecocomposites in simulated soil medium. *Polymer degradation and stability*. 2008; 93 (8): 1571-1576.
43. Sawpan MA, Pickering KL and Fernyhough A. Improvement of mechanical performance of industrial hemp fibre reinforced polylactide biocomposites. *Composites: Part A*. 2011; 42: 310-319.
- thermoplastic natural rubber hybrid nanocomposites. *Composite Interfaces* 2014. Vol. 21 (4): 281-286.
2. Aein Afina, M.R., Noor Najmi, B., Siti Shakirah, S., Surip, S.N. Mechanical And Thermal Properties Of Rubber Toughened Carbon Black-filled Polyester Composite. *Advanced Materials Research* 2013. 812:163-168
3. Aisah Mohamad, Siti Norasmah Surip & Wan Asma Ibrahim. Renewable Sugars Hydrolyzed from Banana Pseudo-Stem Using Different Chemical Pretreatments. *Applied Mechanics and Materials* 2013. 372: 97-100.
4. Azmi, N.N., Surip, S.N., Wan Jaafar, W.N.R. Flexural strength of PLA/kenaf green composite *Advanced Materials Research* 2013. 748:11-15
5. Surip, S.N., Zhang, Z.Y., Dhakal, H.N., Bonnia, N.N., Ahmad, S.H. Thermal properties and crystallisation behaviour of thermoplastic natural rubber (TPNR) nanocomposites. *Advanced Materials Research* 2013. 812:125-130
6. Wan Nor Raihan Wan Jaafar, Siti Norasmah Surip and Nur Naziha Azmi .Effect of Screw Rotation Speed on Mechanical Properties of Extruded PLA/kenaf Nanocomposites. *Advanced Materials Research* 2013.748:61-64
7. A.A Redzuan, N.N Bonnia, N.S Shuhaimen, M.R.H Mohamed & S.N Surip. Mechanical Properties of Rubber Toughened Polyester Filled Carbon Black. *Advanced Materials Research* 2013. 701: 37-41.
8. H.N. Dhakal, Z.Y. Zhang , P.N.B Reis & S.N. Surip. Evaluation of Water Absorption Damage In Hemp Fibre Reinforced Unsaturated Polyester Composites by the Nanoindentation Testing. *Journal of Biobased Materials and Bio-energy* 2012. 6(4): 1-7.
9. Surip, S.N., Bonnia, N.N., Anuar, H., Hassan, N.A., Yusof, N.M. Nanofibers from oil palm trunk (OPT): Preparation & chemical analysis ISBEIA 2012 - IEEE Symposium on Business, Engineering and Industrial Applications 2012. 809-812 6423003
10. H.A. Lutpi, H. Anuar, N. Samat, S.N. Surip, N.N. Bonnia. Evaluation of Elastic Modulus and Hardness of Polylactic Acid-Based Biocomposite by Nano-Indentation. *Advanced Materials Research* 2012. 576:446-449.
11. Anuar, H., Azlina, H.N., Suzana, A.B.K., Kaiser, M.R., Bonnia, N.N., Surip, S.N. and Razak, S.B.A. Effect of PEG on impact strength of PLA hybrid biocomposite. *Business, Engineering and Industrial Applications (ISBEIA), 2012 IEEE Symposium* 2012. 473-476 doi: 10.1109/ISBEIA.2012.6422930
12. N. N. Bonnia, S. N. Surip, and M. M. Mahat. Mechanical Performance Of Hybrid Polyester Composites Reinforced Cloisite 30B and kenaf Fibre. *AIP Conf. Proc.* 2012 1455, 136; doi: 10.1063/1.4732482
13. Azlina, H.N., Norita, H., Anuar, H., Surip, S.N., Bonnia, N.N. Permeability Properties Of Thermoplastic Natural Rubber (TPNR) Nanocomposites. *Business, Engineering and Industrial Applications (ISBEIA), 2012 IEEE Symposium*. 714,718 doi: 10.1109/ISBEIA.2012.6422983
14. N. N. Bonnia, S. H. Ahmad, S. N. Surip, S. S. Nurul, H. N. Azlina nd H. Anuar. Mechanical Properties and Environmental Stress Cracking Resistance of Rubber Toughened Polyester/Clay Composite. *Advanced Materials Research* 2012. 576: 318-321
15. Surip S. N, Wan Jaafar W. N. R , Azmi N. N,& Anwar U.M.K. Microscopy Observation on Nanocellulose from Kenaf Fibre. *Advanced Materials Research* 2012. 488-489:72-75.
16. Wan Nor Raihan Wan Jaafar , SN Surip , Nur Naziha Azmi . Thermal Properties of PLA/Kenaf Green Nanocomposite: Effect of Chemi-Mechanical Treatment . *Advanced Materials Research* 2012. 576:342-344
17. S. Ratim, N. N. Bonnia, and S. N. Surip. The Effect of Woven and Non-woven Fiber Structure On Mechanical Properties Polyester Composite Reinforced Kenaf. *AIP Conf. Proc.* 2012. 1455:131 doi: 10.1063/1.4732481.
18. Z.Y. Zhang, H.N. Dhakal, S.N. Surip, I. Popov, N. Bennett. Characterisation of roof tile coating degradation using nano-indentation test & surface profilometry. *Polymer Degradation & Stability* 2011. 96(5):833-838.
19. Surip, S.N, Halos, N., Bonnia, N.N and Mohamed, R. Nano Modified Epoxy Adhesive: Mechanical & Thermal Properties. *Solid State Science and Technology Letters* . 2010. 17 (1&2) :146-151.
20. S.N. Surip, S.H. Ahmad, R. Rasid, H. Anuar, T. Czigany and S.B. Abdul Razak. Mechanical and Fracture Toughness Behavior of TPNR Nanocomposites. *Journal of Composite Materials*. 2007. 41(17): 2147-2159.



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21. H. Anuar, S.H. Ahmad, R. Rasid, S.N. Surip, T. Czigany and G. Romhany. Essential Work of Fracture and Acoustic Emission Study on TPNR Composites Reinforced by Kenaf Fiber. *Journal of Composite Materials*. 2007. 41(25): 3035-3049
22. Siti Norasmah Surip, Sahrim Ahmad, Rozaidi Rasid, Tibor Czigany, Hazleen Anuar, Noor Azlina Hassan & Norita Hassan. Impact Properties of Thermoplastic Natural Rubber (TPNR) filled Nanoclay Composite. *Journal of Solid State Science and Technology Letters*. 2007. 14(1): 17-21.
23. S.N. Surip, S.H. Ahmad, R. Rasid , T. Czigany & H. Anuar. Instrumented Falling Weight Impact Properties of Thermoplastic Natural Rubber (TPNR) Filled Organoclay Nanocomposites. *Journal of Solid State Science and Technology Letters* 2006. Vol.13 (2):8.
24. S.N. Surip, S.H. Ahmad, R. Rasid & W. N. W. Busu. The Mechanical Properties of Thermoplastic Natural Rubber (TPNR) – Montmorillonite Nanocomposites. *Journal of Solid State Science and Technology Letters* 2004. Vol.2 (83):36.