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The Role of Secondary Filler on Fracture Toughness and Impact Strength of HDPE/Clay Nanocomposites

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ABSTRACT

In this study, oil palm fruit bunch fiber (OPEFB) was used as a secondary filler in HDPE/clay nanocomposites. The composites were prepared by melt compounding, containing high density polyethylene (HDPE), OPEFB fibers, Maleic anhydride grafted polyethylene (MAPE) and four different clay loading (3, 5, 7 and 10 PE nanoclay masterbatch pellets per hundred HDPE pellets). Four OPEFB sizes (180 µm, 250 µm, 300 µm and 355 µm) were added in the composites to investigate its effects on the fracture toughness and impact strength. Fracture toughness of the composites was determined according to ASTM D5045 and single edge notch bending (SENB) was employed during the test while impact tests were performed according to ASTM D256. The effects of alkali treatment were also investigated in this study. The result indicates that the fracture toughness slightly increased as clay loading increased. The highest value of fracture toughness was 0.47 and 1.06 MPa.m1/2 at 5 phr for both types of composites. The presence of OPEFB fiber as a secondary filler in the matrix indicates significant enhancement of fracture toughness up to 133%. However, its impact strength seems to deteriorate with the presence of OPEFB fiber.

Keywords: Alkali treated, hybrid nanocomposite, oil palm empty fruit bunch (OPEFB) fiber, polyethylene, polymer

INTRODUCTION

Natural fibers have attracted the interest of material scientists, researchers, and industries to be used as reinforcement or filler in composites because of their specific advantages compared to conventional or synthetic fibers. Hence, a lot of research has been done to look into the potential of natural

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fiber as a reinforcement in composite materials. However, its mechanical properties are still not comparable to the synthetic fiber, mainly due to lower ultimate tensile strength of the fiber itself and poor bonding between the fiber and matrix. Properties of the natural fiber depend mainly on the nature of the plant, location in which it is grown, age of the plant, and the extraction method used. Several attempts have been successfully made to enhance the mechanical properties of the composites by treating the fibers to improve the bonding between fibers and matrix.

Oil Palm Empty Fruit Bunch (OPEFB) fiber extracted from an empty fruit bunch which is abundantly available is one of the best types of potential fiber to combine with thermoplastic to produce natural fiber composites. Mechanical properties of OPEFB fiber composites have been quite extensively investigated by many researchers (Shinoj, Visvanathan, Panigrahiand, & Kochubabu, 2011). Previous research works of studying and developing thermoplastic reinforced oil palm fruit bunch fibres composites have successfully proven their capability in competing with other natural fibre filled polymer composites (Kalam, Berhan, Ismail, & Razak, 2012) such as wood, kenaf, flax, hemp, coconut husk, pineapple leaf, oil palm, and sisal. However, the major drawback of promoting the usage of OPEFB fibre as reinforcement in polymer composites is always its poor bonding with the matrix. So, the aim of this paper is to study the effect of OPEFB fiber size on the fracture toughness and impact strength of OPEFB fiber filled polymer nanocomposites. This research also investigates the use of clay polymer nanocomposites as the matrix to improve the mechanical properties of composites, besides the adoption of fiber treatment.

MATERIALS AND METHODS

Oil palm fruit bunch fibers were supplied by Poly Region Sdn. Bhd. High density polyethylene (HDPE) was manufactured by Titan PP Polymers (M) Sdn. Bhd (melt flow index of 2.16 and a density of 0.946 g/cm^3, melting temperature between 120 - 130°C, processing temperature of 185°C). Polyethylene nanoclay (PE nanoclay) pellets were provided by Nanocor Inc. in the form of master batch, ready for injection moulding. Meanwhile, Maleic anhydride grafted polyethylene (MAPE) produced by Sigma Aldrich was used as a coupling agent.

Pulverised OPEFB fibres were sieved to separate the particles using the sieve shaker of 180 µm, 250 µm, 300 µm and 355 µm based on the previous study (Kamarudzaman, Kalam, Ahmad, Razak, & Salleh, 2014). The composite formulations consisted of 68 wt% HDPE/Clay nanocomposites as the matrix (the composition of HDPE/clay were similar as described by Kamarudzaman, Kalam, Fadzil, & Ahmad (2014), 30 wt% of OPEFB fibres as the secondary filler and 2 wt% of maleic anhydride grafted polyethylene (MAPE) as the coupling agents. Details of the composite formulations are tabulated in Table 1. Some portions of OPEFB fibres were also treated with 5 % sodium hydroxide (NaOH) solution for 24 hours at room temperature to enhance the bonding between fibre and matrix. After treatment, OPEFB fibers were washed with distilled water to remove impurities and dried at 80°C for about eight hours.

Table 1
Composite formulations

OPEFB /Clay	180 µm	250 µm	300 µm	355 µm
3 phr	-	-	-	Untreated
5 phr	Untreated/treated	Untreated/treated	Untreated/treated	Untreated/treated
7 phr	-	-	-	Untreated
10 phr	-	-	-	Untreated

The compounding of HDPE/MAPE/PE nanoclay composites was performed in a sigma blade dispersion mixer at 185 °C and a rotor speed of 50 rpm for 30 minutes. After the compound was thoroughly mixed, OPEFB fibres were added and the compounding was continued for the next 30 minutes. The compound was removed from the mixer and allowed to cool at room temperature. The total mixing time was approximately one hour. The composite specimens were fabricated using injection moulding techniques into flat dog-bone shape specimens after the compounded material was crushed into granules. Notched Izod impact specimens were prepared and tested following the ASTM D256 to determine the impact strength of the composites.

On the other hand, fracture toughness of the composite was determined according to the ASTM D5045. Single edge notch bending (SENB) specimens were tested with a crosshead speed of 10 mm/min. The dimension of specimens were 52.8 mm × 12 mm × 6 mm (length × width × thickness) with a single notch 4 mm as shown in Figure 1. Five specimens were tested for each batch. The initial portion of the notch was machined with a V-blade at 4 mm and a starter crack was then introduced at the root of the notch by a razor blade, approximately at 2 mm. The ratio of crack length to width ($\frac{a}{w}$) was in the range 0.45 and 0.55.

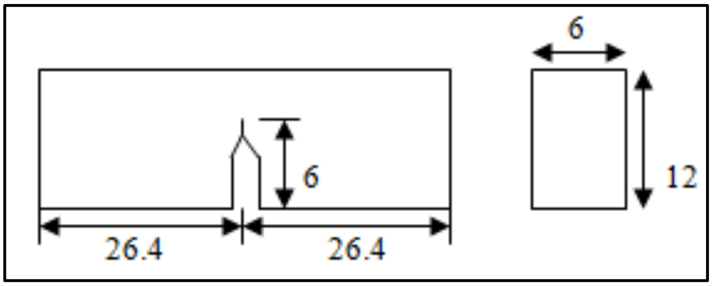


Figure 1. Sample specification of a Single Edge Notch Bend (SENB) specimen, all dimension (in mm)

Stress intensity (K_{1Q}) was calculated using the following equations:

$$K_{1Q} = \frac{P_Q S}{BW^{\frac{3}{2}}} \cdot f\left(\frac{a}{w}\right) \tag{1}$$

$$f\left(\frac{a}{w}\right) = \frac{3\left(\frac{a}{w}\right)^{\frac{1}{2}}\left[1.99 - \frac{a}{w}\left(1 - \frac{a}{w}\right)\left(2.15 - 3.93\left(\frac{a}{w}\right) + 2.7\left(\frac{a}{w}\right)^2\right)\right]}{2\left(1 + 2\frac{a}{w}\right)\left(1 - \frac{a}{w}\right)^{\frac{3}{2}}} \tag{2}$$

Validation of critical stress intensity K_{IC} valueor fracture toughness was done using equation 3.

$$B,\ W - a,\ a \geq 2.5\left(\frac{K_{1Q}}{\sigma_{ys}}\right)^2 \tag{3}$$

Where;

B = specimen thickness

W = specimen width

a = the crack length

W - a = the ligament length

σ_{ys} = the yield strength (Based on the data obtained from previous works by (Fadzil, Kalam, & Kamarudzaman, 2014)

RESULTS AND DISCUSSION

Load versus displacement curves of tested specimens are presented in Figure 2. The curves indicate that most of the specimens have similar trend, which is type I according to ASTM D 399. This trend of load-displacement graph is expected for polymer composites. A linear region can be observed at the beginning of the graph followed by slight nonlinearity before the maximum load is reached. The nonlinearity portion indicates that crack initiation is in progress. The presence of secondary filler (OPEFB fibre) has significantly increased the maximum load of the Clay/HDPE nanocomposites. This trend signifies that the OPEFB fibres are good reinforcement for Clay/HDPE nanocomposites.

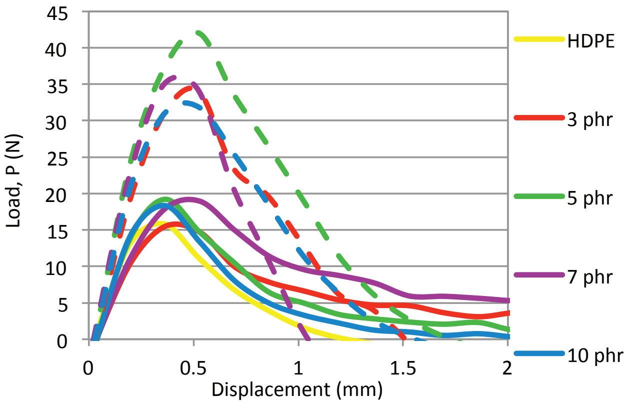


Figure 2. Load Versus displacement of tested specimens, solid curves indicate clay/HDPE and dashed curves indicate OPEFB/Clay/HDPE nanocomposites

Figure 3 shows the fracture toughness of pure HDPE, Clay/HDPE and OPEFB/Clay/HDPE nanocomposites. A slight increase occurs when various clay loadings are added into pure HDPE. The highest value of fracture toughness is 0.47 MPa.m$^{1/2}$ at 5 phr and the lowest value is 0.42 MPa.m$^{1/2}$ at 10 phr. The increase is about 1% to 4%, compared to the fracture toughness of pure HDPE which is 0.37 MPa.m$^{1/2}$. This behaviour is due to the presence of nanoclay which is confirmed by XRD test reported elsewhere (Kamarudzaman et al., 2014).

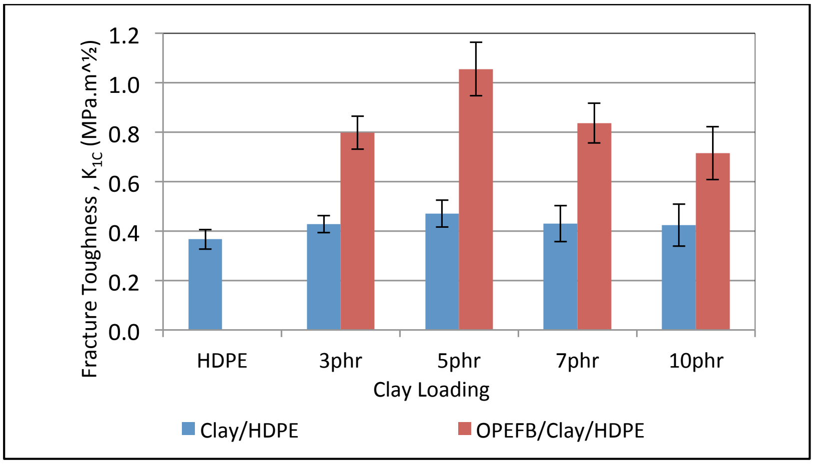


Figure 3. Fracture toughness of nanocomposites at various clay loadings

The addition of OPEFB fibre into Clay/HDPE nanocomposites has significantly increased the fracture toughness and the trend is also the same where it increases when there is an increase in the clay loading. The highest value of fracture toughness for OPEFB/Clay/HDPE is 1.06 MPa.m$^{1/2}$ at 5 phr while the lowest value is 0.72 MPa.m$^{1/2}$ at 10 phr. The covalent bonds between fiber and coupling agents which react with the hydroxyl groups of cellulose or hemicelluloses (two main constituents in oil palm empty fruit bunch fibre) has improved their physical interlock, which consequently leads to fracture toughness enhancement of the composite (Suradi, Yunus, & Beg, 2011). However, beyond 5 phr clay loading, the fracture toughness decreases due to weak interfacial interaction between matrix and filler, leading to weak interfacial bonding (Kamarudzaman et al, 2014). This is possibly due to insufficient coupling agent as the clay loading increases.

The reinforcing ability of the fillers does not just depend upon the mechanical strength of the fillers, but also on many other features, such as polarity (types of functional group) of the filler, surface characteristics and particle size of the fillers (Lewis & Nielsen, 1970). Hence, based on the highest fracture toughness recorded at 5 phr, further investigation was done on several sizes of OPFB fibres to investigate the effect of fibre size. The results are shown in Figure. 4, where the result of the alkali treated OPEFB fibre at 5% NaOH concentrations is also included. The figure indicates that the fracture toughness has increased as the OPEFB fibre size increases towards maximum increase of about 40%. This trend is in line with the improvement of its mechanical properties that are reported in another article (Facile et al., 2014). Alkali treatment has a significant effect on the smaller OPEFB size (180 μm) showed by a 38 % increase in fracture toughness, compared to larger OPEFB fiber (355 μm), that shows insignificant changes. This suggests insufficient alkali concentration for larger OPEFB fibre drug during treatment.

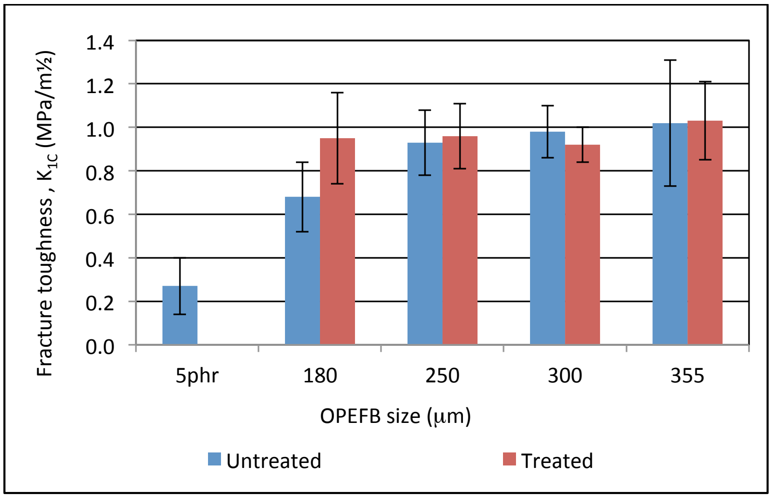


Figure 4. Fracture toughness of OPEFB/Clay/HDPE nanocomposites at various OPEFB fiber size

The addition of OPEFB fibers abruptly decreases the impact strength of composites as displayed in Figure 5. However, as the OPEFB fiber size increases, the impact strength of the nanocomposite also increases up to 27% at 300 μm, then it decreases again to 355 μm OPEFB fibers. It is also noted that alkali treatment on OPEFB fibres produce the same trend towards impact strength as well. For smaller OPEFB fiber size, the impact strength shows significant increase upon 5% alkali treatment. This evidence supports the suggestion that larger OPEFB fibre size needs higher alkali concentration as indicated in fracture toughness test.

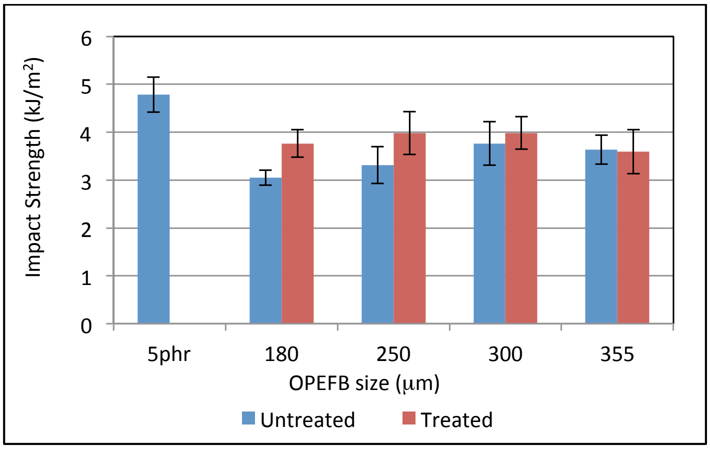


Figure 5. Impact Strength of OPEFB/Clay/HDPE Nanocomposites at Various OPEFB Fiber Size

The presence of clay in the composites is quite detrimental for high speed load, as indicated by the impact strength at various clay loadings shown in Figure 6. The addition of 3 phr of clay loading in HDPE has reduced the impact strength to about 7% and continues to decrease with the increase of clay loading. The same trend is seen by composites with OPEFB fibres as secondary filler. Lower impact strength of composite as compared to pure HDPE is most probably due the brittleness of the composite, where the addition of filler normally will transform the ductile polymer into brittle composite, which can be identified from their stress-strain curves reported in other work (Kamarudzaman et al., 2014). The impact resistance of ductile materials, is mostly contributed by shearing of polymer chain.

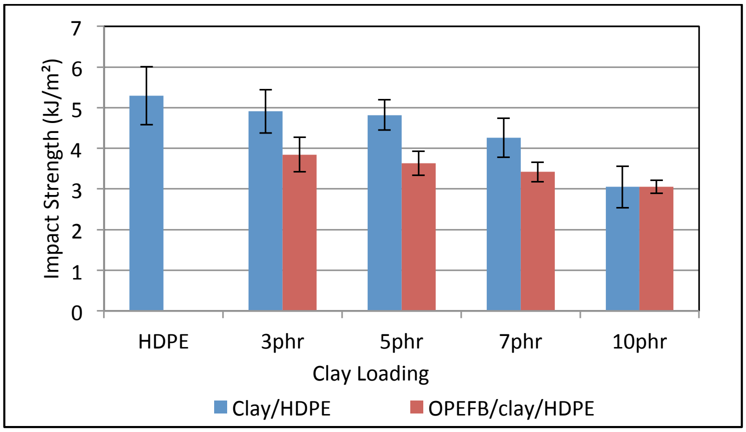
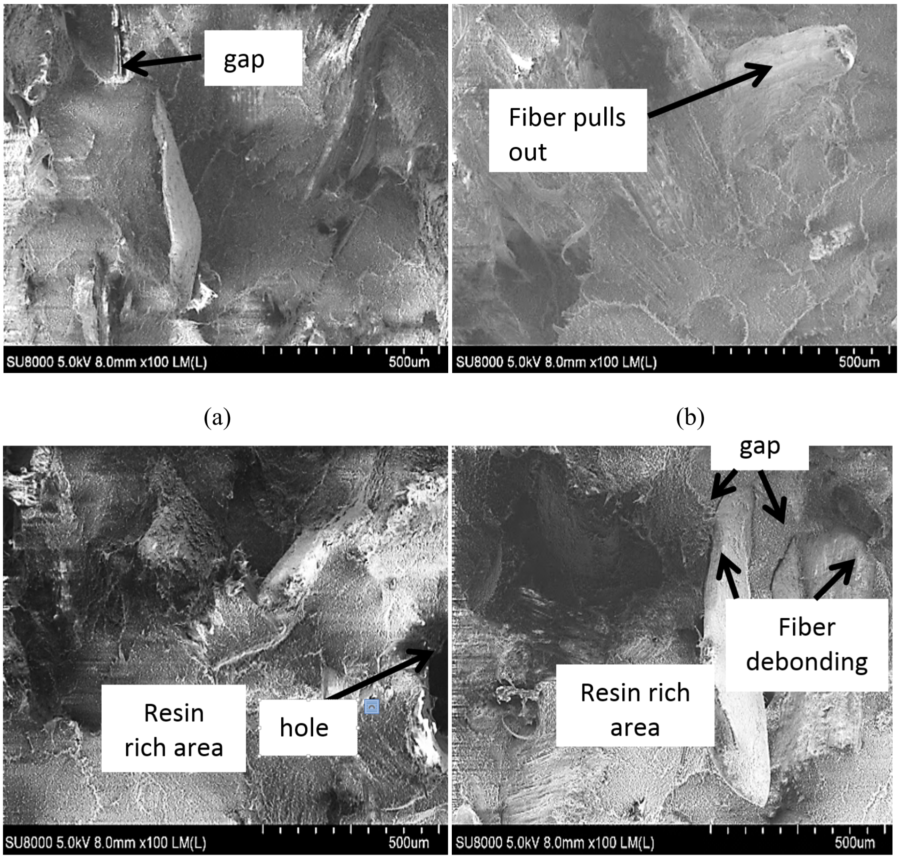


Figure 6. Impact strength of HDPE and its composites at various clay loadings

The improvement of mechanical properties by the addition of coupling agent and fillers is likely due to better bonding between fibre and the matrix. Some evidence of fibre debonding from the matrix that leaves empty holes, creates larger flaws and hence worsens the fracture resistance ability, which can be observed from the FESEM micrograph is depicted in Figure 7(a). In Figure 7(b), it can be seen that the OPEFB fibre is nicely embedded in the matrix that supports the evidence of highest fracture toughness of nanocomposites with 5 phr clay loading. It is clearly shown that the interfacial region between filler and the matrix is continuous, indicating a good interfacial bonding between the constituent phases.



(a) (b)

(c) (d)

Figure 7. Fracture Surface of OPEFB/Clay/HDPE at various clay loadings: (a) 3 phr; (b) 5 phr; (c) 7 phr; and (d) 10 phr

A small hole as shown in Figure 7(c) due to air bubble during composites preparation also serves as a weak point during testing. A significant improvement can be achieved if the fibres are to be dispersed uniformly in the matrix (Ishak, Aminullah, Ismail, & Rozman, 1998), hence no resin rich areas are observed on the fracture surfaces. Some occurrences of OPEFB fiber debonding are visible, which is dominant on the fracture plane as illustrated in Figure 7(d). This is believed to be more detrimental to the mechanical performance of the composites.

CONCLUSION

The addition of OPEFB fiber as a secondary filler in the matrix indicates significant enhancement of fracture toughness of up to 133%. The highest fracture toughness, K_{1C} of clay/HDPE nanocomposites are found to be 0.47 MPa.m$^{1/2}$ at 5 phr clay loading. The addition of 355 µm OPEFB fibres in th eclay/HDPE nanocomposite has doubled the K_{1C} with the maximum value recorded at 1.15 MPa.m$^{1/2}$. Further investigation on the effect of OPEFB fibre size revealed that K_{1C} value increases in tandem with the increase in OPEFB fibre size. Fibre treatment with 5% NaOH solution appears to work well on the small OPEFB size, where it is proven to dramatically increase the K_{1C} value up to 40%. Further investigation is needed for higher concentration of NaOH and larger OPEFB size. However, its impact strength seems to deteriorate with the presence of OPEFB fiber concentration of NaOH and larger OPEFB size.

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