Effects of extrusion temperature on the rheological, dynamic mechanical and tensile properties of kenaf fiber/HDPE composites

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ABSTRACT

The effects of extrusion processing temperature on the rheological, dynamic mechanical analysis and tensile properties of kenaf fiber/high-density polyethylene (HDPE) composites were investigated for low and high processing temperatures. The rheological data showed that the complex viscosity, storage and loss modulus were higher with high processing temperature. Complex viscosities of pure HDPE and 3.4 wt% composite with zero shear viscosity of \( \mu_{0} = 2340\ \text{Pa s} \) were shown to exhibit Newtonian behavior while composites of 8.5 and 17.5 wt% with zero shear viscosity \( \mu_{0} \geq 30,870\ \text{Pa s} \) displayed non-Newtonian behavior. The Han plots revealed the sensitivity of rheological properties with changes in processing temperature. An increase in storage and loss modulus and a decrease in mechanical loss factor were observed for 17.5 wt% composites at high processing temperature and not observed at low processing temperature. Processing at high temperature was found to improve the tensile modulus of composites but displayed diminished properties when processed at low processing temperature especially at high fiber content. At both low and high processing temperatures, the tensile strength and strain of the composite decreased with increased content of the fiber.

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1. Introduction

The interest in using natural fibers in polymer matrix composite (PMC) has increased in recent years. This is due to the lightweight, non-toxic, low cost and biodegradable properties of natural fibers. The use of natural fibers, derived from renewable resources, as reinforcing agent in both thermoplastic matrix composites provides positive environmental benefits with respect to disposability and raw material utilization [1].

Kenaf fiber has potential as reinforcement filler in PMC. The purpose of producing PMC is to create a new material that has better properties compared to their individual material. Kenaf fiber can generally be classified into two types. The first type is the outermost layer known as bast while the second type is the inner part, known as core. The core is very soft, hollow and suitable for application as organic filler in plastic, while bast fiber has hard properties and is suitable for blending with plastic, textile industry and also fiberglass technology applications. Kenaf fiber is also used as reinforcement for plastic and synthetic product, cosmetic product, organic filler and medicine. Besides that kenaf fiber is also environmentally friendly.

The performance of a composite material varies with the fiber–matrix bond strength and to some extent, depends on the choice of suitable processing techniques. There are various methods of processing natural fiber–polymer matrix composites. Methods such as extrusion, compression and injection molding are used to introduce fibers into the thermoplastic matrix. Twin-screw extruder system, with a feeding and a mixing zone, consists of screws with a multiplex shape. In this system, the fillers are very well blended with the matrix polymer because the mixtures pass through a number of mixing blocks [2]. Potente et al. [3] studied the impact of speed, melt throughput, continuous-phase viscosity, screw configuration, and disperse-phase content on the melting behavior and morphology development in the melting zone of a twin-screw extruder of polypropylene/polyamide-6 composite. Their result showed a finely dispersed morphology at the start of the melting section and the screw can feed and mix the melted mixture simultaneously. The use of unmodified and modified sugarcane bagasse cellulose/HDPE composite with zirconium oxychloride formed by compounding 10% by weight of fiber using extrusion and compresion molding showed that the modified composites present better tensile strength compared to unmodified composites [4].

Rheological measurement conducted in various steady state and dynamic environments is a widely used technique for determining the sensitivity of a material during processing [5]. From the findings conducted on multi-wall carbon nanotube (MWCNT)/poly(ether ether ketone) (PEEK) composites containing...
up to 17 wt% filler compounded by using a twin screw extruder, the complex viscosity and moduli as determined by the linear viscoelastic measurements increase with increasing MWCNT concentration and the storage modulus, \(G'\) exhibits a dramatic seven order increase in magnitude around 1 wt%, leading to a solid-like behavior at low frequency especially at higher filler loadings [6]. Rana et al. [7] have reported that the storage and loss moduli of short jute fiber reinforced PP composites increased with fiber content. However injection molding, extrusion or mixing in an internal mixer and then molding involves high shear and therefore it might damage the natural fiber [8].

In this research, the processing parameter of kenaf fiber/HDPE composites using co-rotating twin screw extruder was investigated at two different temperatures defined as low processing temperature (LPT) and high processing temperature (HPT). The main aim of this work is to establish the effect of this extrusion parameter on the rheological, dynamic mechanical and tensile properties.

2. Experimental

2.1. Materials

Kenaf bast fiber of 3 mm length with the average density of 134.3 kg/m\(^3\) was obtained from the National Kenaf & Tobacco Board, Malaysia. It was sieved and fibers with diameter of less than 0.5 mm were collected. A semi-crystalline high density polyethylene (HDPE), Titanex HI 1100 with a density of 961 kg/m\(^3\) and melt flow index of 7 g/10 min, supplied by Titan Petchem (M) Sdn. Bhd., Malaysia was used as the matrix.

2.2. Extrusion and compression molding

Kenaf fiber/HDPE composites were prepared by melt-compounding using a co-rotating twin screw extruder with gravimetric metering device feeder (Brabender KETSE 20/40 Lab Compounder, Germany). The screw has a diameter of 20 mm and length to diameter ratio of 40. Extrusion was carried out at screw speed of 80 rpm at 2 kg/h feeding rate with two different temperature settings, i.e. 160, 165, 170, 175 and 180 °C from the hopper to the die and 165, 170, 175, 180 and 185 °C, designated as LPT and HPT respectively. These processing temperatures produced the actual melt temperatures of between 190–195 °C and 194–198 °C for LPT and HPT respectively. Pure HDPE was loaded automatically into the feed hopper by using gravimetric metering device while kenaf fiber was introduced to the barrel at the side feeder between zones 3 and 4 (decompression zone). The strands leaving the extruder die were pelletized and molded using the compression molding machine at temperature of 155 °C and pressure of ca 98 MPa (100 kgf/cm\(^2\)).

2.3. Rheological study

For the rheological study, materials were subjected under dynamic frequency sweep (DFS) mode using a Physica MCR 301 rheometer (Anton Paar, Germany). A cone-plate measuring system was used with geometry of 25 mm diameter, 0.051 mm gap and angle 2°. Measurements were performed over frequency range from 0.05 to 500 rad/s with 5% strain amplitude at temperature of 190 °C.

2.4. Dynamic mechanical properties

The dynamic mechanical properties of composites were analyzed using a dynamic mechanical analyzer model Q800 (TA Instruments, USA) in thin film mode. DMA specimens were cut from the compression molded sheet to a dimension of 30 mm length, 6.3 mm width and 0.13 mm thick. Measurements were conducted over a temperature range from −135 °C to 100 °C with a heating rate of 3 °C/min and constant frequency of 1.0 Hz.

2.5. Tensile properties

Tensile tests were conducted according to ASTM D-638 standard by using a universal testing machine (Instron 5567, USA) equipped with a load cell of 10 kN at a constant cross-head speed of 5 mm/min, and a gauge length of 50 mm. Dumb-bell shaped tensile test specimens were cut to the dimension of 75 mm length by 6 mm width by 1 mm thick. For each test, a minimum of seven samples were tested.

3. Results and discussion

Since there is no application of fiber surface treatment or coupling agent materials during compounding to improve fiber–matrix interfacial adhesion, it is assumed that any changes in materials properties are due to the quality of fiber dispersion within the matrix, compounded at LPT and HPT.

3.1. Rheological properties (DFS)

Rheology is a study of flow behavior of liquids or deformation behavior of solid material when it is being processed. Many factors including temperature, pressure, screw speed, type of material, amount of filler loading and die diameter of die influence the quality of the extrusion product. Extrusion process requires that the polymer composite be subjected to temperatures above its melting point to enable deformation and flow while the screw speed is responsible for feeding and mixing the melted material simultaneously so as to be forced into the extruder barrel and die. Rheological properties can reflect the internal structure and process ability of materials [9]. DFS, an oscillatory test, has been performed at variable angular frequencies, keeping the amplitude at a constant value to give the complex viscosity, storage and loss moduli behavior of the kenaf fiber/HDPE composites.

3.1.1. Complex viscosity

The complex viscosity, \(\eta^{*}\) is defined by,

\[
|\eta^{*}| = \frac{G'}{\omega} \tag{1}
\]

where \(G'\) and \(\omega\) are the complex shear modulus and angular frequency respectively. Fig. 1 shows the complex viscosity curves as a function of angular frequency for the kenaf fiber/HDPE composites obtained at LPT and HPT with different fiber loadings. \(\eta^{*}\) of pure HDPE and 3.4 wt% fiber composite shows a Newtonian plateau until angular frequency of 10\(^5\) rad/s, beyond which non-Newtonian behavior prevails. On the contrary, composites with 8.5 and 17.5 wt% fiber display non-Newtonian behavior over the entire range of frequencies for both at LPT and HPT.

The zero-shear viscosity, \(\eta_0\) is a very important value since it is proportional to the average molar mass, \(M_m\). The Carreau-Yasuda 1 regression from the complex viscosity curve was used to determine the \(\eta_0\) of the composites. The Carreau-Yasuda 1 regression is defined by,

\[
\eta = \frac{\eta_0 - \eta_\infty}{1 + (\lambda/\omega)^{\eta_0 - \eta_\infty}} + \eta_\infty, \quad \eta_0 - \eta_\infty > 0 \tag{2}
\]

where \(\eta, \eta_0, \eta_\infty, \lambda, a\) and \(n\) are the viscosity, zero shear viscosity, infinite-shear viscosity, relaxation time, power law exponent and
width of the transition range respectively. Data of \( \eta_0 \) from Carreau-Yasuda 1 regression is tabulated in Table 1. In relation with Table 1 and Fig. 1, it shows that pure HDPE and 3.4 wt% fiber composites with \( \eta_0 \leq 2340 \text{ Pa s} \) shows Newtonian behavior, while composites with 8.5 and 17.5 wt% fibers display non-Newtonian behavior with \( \eta_0 > 30,970 \text{ Pa s} \). From the \( \eta_0 \) data, average of molar mass value, \( M_w \) was calculated using equation,

\[
\eta_0 = k_e M_w^a
\]

(3)

where the parameters \( k_e \) and \( M_w \) are the material constant and average of molar mass respectively. The relationship between molecular features and rheological properties is a topic of long-standing discussion in polymer science, especially the \( \eta_0-M_w \) dependence, where the power law exponent, \( a \), takes values varying between 3.36 and 3.64 for polyethylene, and the material constant, \( k_e \), depends on temperature \([10,11]\). Recent work has shown that the relationship between \( \eta_0 \) and \( M_w \) for the two species is indistinguishable at 175 °C, and those results with a very minor temperature adjustment to 190 °C using \( E_a \) as 6.4 kcal/mol for linear polyethylene \([12-14]\) lead to,

\[
\eta_0 = 3.4 \times 10^{-14} M_w^{1.6}
\]

(4)

for \( M_w \geq 3200 \) \([15]\). The \( k_e \) and \( a \) value from the above literature was used to calculate the average molar mass and tabulated in Table 1. From the table, it can be seen that the average molar mass increases with increasing fiber loading.

<table>
<thead>
<tr>
<th>Fiber fraction (wt%)</th>
<th>Barrel setting</th>
<th>Zero shear viscosity, ( \eta_s ) (Pa s)</th>
<th>Average molar mass, ( M_w ) (g/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>LPT</td>
<td>710</td>
<td>34,152</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>960</td>
<td>37,125</td>
</tr>
<tr>
<td>3.4</td>
<td>LPT</td>
<td>2339</td>
<td>47,545</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>2020</td>
<td>45,648</td>
</tr>
<tr>
<td>8.5</td>
<td>LPT</td>
<td>30,971</td>
<td>97,441</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>36,750</td>
<td>102,184</td>
</tr>
<tr>
<td>17.5</td>
<td>LPT</td>
<td>123,980</td>
<td>143,244</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>159,620</td>
<td>153,659</td>
</tr>
</tbody>
</table>

Extrusion at HPT also shows a higher \( \eta_0 \) and higher \( M_w \) for all samples compared to the one processed at LPT except for the composites at 3.4 wt% fiber loading (Table 1). Moreover, composites extruded at HPT show higher \( \eta'' \) compared to those at LPT as observed in 0.0, 8.5 and 17.5 wt% fiber composites (Fig. 1). The composites with 3.4 wt% fiber did not show significant difference when the processing temperature was changed. Extruding the composites at HPT shows higher \( \eta'' \) compared to the ones at LPT measured at both low and high frequencies. At HPT, there is enough heat and energy to melt the matrix and at the same time have the sufficient melt strength to produce a more homogeneous dispersion of fiber within the matrix. It is also believed that at this processing temperature, the optimum stress relaxation occur and therefore shows a higher \( \eta'' \) compared to that at LPT. However, LPT does not seem to have sufficient heat to fully melt the matrix. The matrix becomes more viscous and the fiber will be difficult to disperse in the matrix. This consequently creates inhomogeneous composites, resulting in reduction of melt strength and complex viscosity. At low temperature, the viscosity as well as the shear stress generated in the mixture is very high, causing the breakdown of the fibers during mixing \([16]\).

Composites at 3.4 wt% fiber loading exhibit an almost similar pattern as pure HDPE, i.e. Newtonian plateau at the low frequency region. This is believed to be due to the low loading level of kenaf fiber in HDPE matrix. In the higher filled composite systems, a significant increase in \( \eta'' \) can be observed with increasing fiber loading at all frequency range, i.e. non-Newtonian behavior. Appearance of shear-thinning behavior of \( \eta'' \) for 8.5 and 17.5 wt% fiber composites at low frequencies indicates the transformation from liquid-like to pseudo-solid like response. The presence of fiber with higher fiber loading perturbs the normal flow of polymer and therefore hinders the mobility of polymer chain segments. This phenomenon is attributed to shear thinning effect, which is predominant in the composite materials compared to a pure matrix. This behavior is due to the fact that at low oscillatory frequencies, disoriented fiber collisions dominate the flow compared to the situation at high frequencies, where matrix behavior is the dominant factor \([5]\).

### 3.1.2. Storage and loss moduli

The storage and loss moduli as a function of frequency of kenaf fiber/HDPE composites are presented in Figs. 2 and 3 respectively. Both storage, \( G' \) and loss, \( G'' \) moduli curves of pure HDPE and 3.4 wt% fiber composites increase progressively with increasing frequency, showing a terminal behavior. Meanwhile at 8.5 and 17.5 wt% fiber loadings, the slope of \( G' \) and \( G'' \) level off especially at the low frequency range indicating the nonlinear pattern. Fig. 2 shows the slope values of \( G' \) for pure HDPE at LPT and HPT are almost the same (\( G' \sim \omega^{0.87} \)) (Fig. 3). From the above results, the relaxation exponents of storage and loss moduli for pure HDPE are less than what is expected from the linear viscoelasticity theory, where \( G' \sim \omega^2 \) and \( G'' \sim \omega^3 \) respectively. According to the linear viscoelasticity theory, the storage and loss moduli of homogeneous polymer systems obey scaling law behavior in the low frequency region, with slopes equal to 2 and 1 respectively. The linear viscoelasticity at low frequencies (terminal zone) reflects a fully relaxed polymer chain \([6]\). Similar to \( \eta'' \) finding, composites with 3.4 wt% fiber did not show a significant difference on \( G' \) and \( G'' \) with changing of processing temperature. The development of plateau at low frequencies can be seen for the 8.5 and 17.5 wt% fiber composites, where \( G' \) and \( G'' \) values deviate from that of pure HDPE due to the effect of fiber loading.
At higher fiber loading, the magnitudes of \( G_0 \) and \( G''_0 \) of composites compounded at HPT are greater than those compounded at LPT, suggesting a synergistic effect on the elastic behavior of the composites. Sample extruded at HPT with 17.5 wt% fiber loading shows the highest magnitude of \( G_0 \) and \( G''_0 \), thus revealing the reinforcing effect imparted by the fiber. Bangarusampath et al. [6] reported that for MWCNT/PEEK composites, \( G_0 \) and \( G''_0 \) often deviate from the linear viscoelasticity behavior. The viscosity of the matrix polymer increases with the incorporation of fiber, which can be seen with the increase of the storage modulus. At loading above 1 wt%, the terminal behavior disappears and instead tends to a plateau-like region that is indicative of a transition from liquid-like to solid-like viscoelastic behavior. This critical composition (around 1 wt%), correlates with the transitions observed in the other rheological data and can be identified as a rheological percolation threshold concentration. An investigation on fiber-filled polymeric melt claimed that formation of a plateau at a very low applied frequency indicates that relaxation of the polymer composites is concentrated in the low frequency region. The authors also observed that an apparent yield stress is exhibited due to the formation of a network structure within the matrix [6]. However, in the present study, the storage modulus increases considerably in the composites at low frequency and no yield stress is observed. This phenomenon is attributed to an increase in the rigidity of the matrix with the incorporation of the fibers [5].

The change in rheological properties with fiber content was also studied using a log–log plot of \( G' \) versus \( G'' \) (Han plot). This plot indicates the sensitivity of rheological properties to composition and temperature [17,18]. Fig. 4 shows the Han plot of the kenaf fiber/HDPE composites, plotted with the data extracted from Figs. 2 and 3. As can be seen, the Han plot shows that at these two different processing temperatures, the curves are not fitted into a single curve. HPT shows an upward shift compared to the LPT for pure HDPE, 8.5 and 17.5 wt% fiber loadings composites. This indicates that the rheological behavior of these samples is affected by the extrusion temperature significantly. The strong temperature dependence of \( \log G' \) versus \( \log G'' \) plot over a range of temperatures is attributable to the occurrence of a thermally induced transition from an ordered microdomain structure to a disordered homogeneous phase [19]. From this, it is also clear that rheological properties change with increase in fiber loading, given the Han plot shows an upward shift from pure HDPE to the highest filler content of 17.5 wt% fiber. Unfortunately all the Han plots have a slope of much less than 2 compared to the linear viscoelasticity theory. It was reported [20] that the \( \log G' \) versus \( \log G'' \) for homopolypropylene/maleated composite (PPVC-PP-g-MA) and copolymer ethylene propylene/maleated composite (PPSC-PP-g-MA), the curves shift upwards as the PP-g-MA content increases. In the oscillatory test, the deformation of the dispersed phase with very low viscosity should induce and improve the deformation of the continuous PP phase. The deformation will become more prominent with higher PP-g-MA content. Consequently, the ratio of the amount of energy stored (\( G' \)) to the amount of energy dissipated (\( G'' \)) increases with an increase of the PP-g-MA content in the blend, that is the ratio varies with the blend composition.

### 3.2. Dynamic mechanical analysis

#### 3.2.1. Storage modulus, \( E' \)

Storage modulus is the elasticity properties that describes the energy stored in the system. Change in \( E' \) indicates the changes in rigidity and hence strength of the materials. Fig. 5 shows the storage modulus (\( E' \)) curves of composites compounded at LPT and HPT. The storage modulus decreases with increase in temper-
Fig. 5. Storage modulus curves of composites compounded at (a) LPT and (b) HPT.

3.2.2. Loss modulus, $E''$

Loss modulus is the viscous component that describes the energy dissipated during a process and is susceptible to molecular motions [7,22]. Variations of loss modulus as a function of temperature for composite specimens at LPT and HPT are graphically plotted in Fig. 6. From Fig. 6(a), pure HDPE, 8.5 and 17.5 wt% fiber composites exhibit three transition peaks ($\alpha$, $\beta$ and $\gamma$) while 3.4 wt% fiber composite exhibits only two transition peaks ($\alpha$ and $\gamma$). The $\alpha$-transition temperature within the temperature range of 38.5–66.5 °C (Table 3) is associated to the motion of long-chain ($–CH_2–$)$_n$ segments in the crystalline region of the HDPE. The $\beta$-transition temperature, which is around -32.0 to -26.0 °C, is related to the segmental motion of ($–CH_3$) relaxation in the amorphous region of the HDPE and the $\gamma$-transition between -120.0 and -117.0 °C, is attributed to long chain ($–CH_2–$)$_n$ crankshaft relaxation in the amorphous polyethylene segments of the HDPE chain. Khanna et al. [28] reported that the $\beta$ and $\gamma$-transitions for HDPE were at -45 °C and -107 °C (±1 °C) respectively. Transitions at $\beta$-relaxation are associated with the branch point. This relaxation is therefore generally attributed to segmental motions of methyl groups in the non-crystalline phase. At LPT, the $\beta$-transition peak of the composites was shifted from about 5.5 and 6.0 °C to a lower temperature compared to the pure HDPE as observed in the case of 8.5 and 17.5 wt% fiber composites. A lower $\beta$-transition temperature implies the presence of some processes, which have possibly
led to the softening of the matrix and increasing the ability of the
matrix chains to move more freely [29].

Fig. 6(b) shows a variation of the loss modulus of composites
compounded at HPT. At HPT, only the 17.5 wt% fiber composite
exhibits three transition peaks (α, β and γ) while composites
with 3.4 and 8.5 wt% fiber exhibit only two transition peaks
(α and γ). The α, β and γ-transition temperatures are found
to be within 43.5–62.0 °C, −22.5 to −2.5 °C and between
−119.0 and −116.0 °C respectively (Table 3). Processing at higher
temperature with 17.5 wt% fiber composite shows better
properties of loss modulus compared to the pure HDPE. At
17.5 wt% fiber composites, processing at this condition gives
the highest magnitude of $E_0^0$ (625 MPa), $E_2^0$ (727 MPa) and $E_3^0$ (665 MPa).

3.2.3. Tan Delta
Mechanical loss factor curves of composite specimens com-
pounded at LPT and HPT are shown in Fig. 7. The tan $\delta_{\text{max}}$ peak can also provide information on the glass transition temperature, $T_g$, and energy dissipation of composite materials [30]. From
Fig. 7(a), pure HDPE presents the highest magnitude of tan $\delta_{\text{max}}$ (6.23 × 10^{-2}) at LPT. Composites with 3.4 wt% fiber loading show the highest magnitude of tan $\delta_{\text{max}}$ (5.82 × 10^{-2}) compared to other fiber loadings. The damping or tan $\delta$ in the transition region measures the imperfections in the elasticity of a polymer [31]. The higher damping factor relates to a poor fiber dispersion within
the matrix. At 17.5 wt% fiber loading, the tan $\delta_{\text{max}}$ value is found to be higher compared to the composite at 8.5 wt% fiber loading. Its $T_g$ value also shifted slightly to the lower temperature (−115.5 °C)

### Table 3

<table>
<thead>
<tr>
<th>Fiber fraction (wt%)</th>
<th>Barrel setting</th>
<th>Loss modulus, $E^0$ (MPa)</th>
<th>β-Transition</th>
<th>γ-Transition</th>
<th>Tan δ</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$E^0$ max (MPa)</td>
<td>$T_a$ (°C)</td>
<td>$E^0$ max (MPa)</td>
<td>$T_g$ (°C)</td>
</tr>
<tr>
<td>0</td>
<td>LPT</td>
<td>173</td>
<td>55.0</td>
<td>107</td>
<td>−26.0</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>297</td>
<td>57.0</td>
<td>183</td>
<td>−22.5</td>
</tr>
<tr>
<td>3.4</td>
<td>LPT</td>
<td>217</td>
<td>66.5</td>
<td>58.0</td>
<td>−31.5</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>289</td>
<td>58.0</td>
<td>158</td>
<td>−32.0</td>
</tr>
<tr>
<td>8.5</td>
<td>LPT</td>
<td>288</td>
<td>58.0</td>
<td>158</td>
<td>−31.5</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>359</td>
<td>62.0</td>
<td>140</td>
<td>−32.0</td>
</tr>
<tr>
<td>17.5</td>
<td>LPT</td>
<td>152</td>
<td>38.5</td>
<td>43.5</td>
<td>727</td>
</tr>
<tr>
<td></td>
<td>HPT</td>
<td>625</td>
<td>43.5</td>
<td>727</td>
<td>2.5</td>
</tr>
</tbody>
</table>
compared to the others (Table 3), which indicates an increase in molecular mobility of composite with LPT.

From Fig. 7(b) composites compounded at HPT show that the magnitude of $\tan \delta_{\text{max}}$ values reduce with increasing fiber loading except for the 3.4 wt% fiber composite. The higher reduction of $\tan \delta_{\text{max}}$ values for composites with higher fiber loading is believed to be due to the strengthening effect imparted by the fibers. This is caused by the limited mobility of polymer matrix which was affected by the improvement in fiber dispersion in the composites [24]. In composites, as the amount of incorporated fiber increases, the contact area between the fiber and the polymer matrix also increases, leading to a stronger interaction between the fiber and matrix. Hence, the chain mobility of HDPE around the fiber is restrained, resulting in the improvement in the hysteresis of the system and a reduction in the internal friction [21]. Therefore, with the presence of kenaf fiber, the molecular mobility of the polymeric materials decreases and the mechanical loss to overcome intermolecular chain friction is reduced [32]. The increasing magnitude of $\tan \delta_{\text{max}}$ at 3.4 wt% fiber composite is expected because the high volume of matrix is able to dissipate the vibrational energy and gives higher damping at the interface.

3.3. Tensile properties

3.3.1. Tensile modulus

The tensile modulus values and tensile strengths of pure HDPE and kenaf fiber/HDPE composites are presented in Fig. 8. At HPT, pure HDPE and kenaf fiber/HDPE composites show a higher tensile modulus compared to those processed at LPT. The tensile modulus values increase with increasing fiber loading when processed at HPT. The increment of modulus values with increasing amount of fiber loading is in agreement with what was reported by Girun et al. [33], where a higher fiber loading composites are able to withstand more load and can fabricate the composites to become stiffer [34]. In comparison to the pure HDPE at HPT, the tensile modulus of composites containing 3.4, 8.5 and 17.5 wt% kenaf fiber increase by 8.7%, 10.9% and 12.2% respectively. This shows that the incorporation of kenaf fiber at HPT improves the tensile modulus of HDPE, indicating that transferring of stress from the polymer matrix to the stiffer fiber has occurred [35]. On the contrary, LPT composites show a decrease in tensile modulus compared to the HPT composites for all samples, especially for the highest fiber loading of 17.5 wt%. The tensile modulus of composite at 17.5 wt% kenaf fiber decreases by 29% when processed at LPT compared to the HPT.

3.3.2. Tensile strength

The tensile strength of the composites is shown to decrease with increasing content of kenaf fiber at both LPT and HPT (Fig. 8). This could be attributed to the poor adhesion between fiber and matrix, resulting in the poor interfacial interaction and debonding of the matrix from the fiber during the tensile deformation. The debonding results in void formation, which lowers the tensile strength because cracks can easily propagate through regions containing the voids [21,31]. Nevertheless, the HPT composites shows better tensile strength compared to the LPT composites. LPT results in inconsistent melt of the resin that can lead to non-uniform dispersion of the fibers in the composites and eventually lowers the tensile strength [36].

3.3.3. Tensile strain

Fig. 9 shows tensile strain of pure HDPE and kenaf fiber/HDPE composites. Generally, the addition of fiber reduces the tensile strain of composites. The tensile strains of the LPT composites are higher compared to the HPT composites and greatly decrease with fiber loading. The elongation probably arises from the polymer matrix because the kenaf fiber is rigid relative to HDPE. Increasing the amount of filler decreases the amount of polymer available for the elongation [31]. Moreover, adding higher amount of fiber increases the possibility of fiber agglomeration. Such an agglomeration can lead to the formation of stress concentrated region where less energy is required for elongating the crack propagation [37].

4. Conclusion

Rheological data on DFS measurements of HPT specimens showed higher complex viscosity, zero shear viscosity and average molar mass for all samples compared to the LPT composites, except for the low fiber loading of 3.4 wt%. Complex viscosity of pure HDPE and 3.4 wt% fiber composite with $\eta_o \leq 2340$ Pa s showed Newtonian behavior. However, composites with higher fiber loadings of 8.5 and 17.5 wt%, the complex viscosity curves with $\eta_o \geq 30,970$ Pa s displayed non-Newtonian behavior. In addition, storage ($'G'$) and loss ($'G''$) moduli curves of pure HDPE and 3.4 wt% fiber composites increased progressively with increasing angular frequency. Meanwhile at 8.5 and 17.5 wt% fiber loadings, the slope of $'G'$ and $'G''$ were found to level off especially at the low frequency range, indicating the nonlinear pattern due to shear thinning. The Han plots showed that at different processing temperatures the curve were not fitted into a single curve but an
upward shift with HPT for pure HDPE, 8.5 and 17.5 wt% fiber loadings compared to the LPT. Moreover, increasing the fiber loading showed an upward shift of Han plot from pure HDPE to the highest fiber content of 17.5 wt%.

DMA showed that the storage and loss moduli of composites increased with increasing fiber loading and 17.5 wt% fiber composite showed the highest value of storage and loss moduli at HPT. However, composite with the highest fiber loading (17.5 wt%) compounded at LPT showed the lowest storage and loss moduli curves. The incorporation of the kenaf fiber at HPT reduced the magnitude of tan\(\delta\)\(_{\text{max}}\) values with the increasing fiber loading except for the low loading fiber of 3.4 wt%. Storage and loss moduli as a function of temperature appeared to be more sensitive to the fiber loading at LPT while this was not observed in the storage and loss moduli as a function of angular frequency.

By increasing the compounding temperature, the tensile modulus presented an increasing pattern compared to the ones processed at LPT. The tensile modulus also increased with increasing fiber loading when processed at HPT. However, the tensile strength of the composites decreased with increasing content of the fiber at both LPT and HPT. Nevertheless, composites processed at HPT showed better tensile strength compared to the ones at LPT. The tensile strains of the composites decreased with the increasing amount of fiber loading while composites processed at LPT presented a higher tensile strain compared to those at HPT. From the above, it can be concluded that the composites compounded at HPT provides best performance of rheological, thermo-mechanical and tensile properties in comparison with composites compounded at LPT.

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References


