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Natural fiber-reinforced high-density polyethylene composite hybridized with ultra-high molecular weight polyethylene

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Abstract

A great deal of research and development work has been recently conducted on natural fiber-reinforced polymer matrix composite for its abundancy, low density, excellent damping characteristic, and good mechanical properties. However, the low strength of natural fiber composite has limited its use to only low stress applications. The purpose of this work is to develop a natural fiber hybrid material with both enhanced strength and failure strain using a novel approach and study the effect of the processing temperature on its microstructure and performance. High-strength ultra-high molecular weight polyethylene fabrics are co-molded onto the surfaces of a kenaf fiber high-density polyethylene-based composite material by single-step compression molding. The status of the ultra-high molecular weight polyethylene fabrics at different processing temperatures is investigated using microscopic analysis. The tensile strength and impact strength of the hybrid material are evaluated. It is found that its tensile strength is increased by more than 90% with only 8% ultra-high molecular weight polyethylene fiber reinforcement added and its low density is maintained.

Keywords

Polyethylene, natural fiber, composites, ultra-high molecular weight polyethylene, compression molding

Introduction

Natural fiber thermoplastic matrix composites have unique characteristics such as low density, high damping and specific mechanical properties, biodegradability, and recyclability. They have been widely used in applications including automotive and construction because of these advantages. The environmentally-friendly natural fiber thermoplastic composite consists of natural fibers, such as hemp, kenaf, and flax, and matrix such as polyethylene and polypropylene. The low densities of both natural fibers and thermoplastics often result in a composite with a density ranging from 1.04 to 1.45 g/cm^3 depending on the types of fiber and matrix and fiber percentages.[1,2]

In spite of the above-mentioned advantages of the natural fiber thermoplastic composites, their use has been generally limited to low stress applications because of its relatively low strength.[3–5] Natural fiber itself normally has good tensile strength. Most of natural fibers have a tensile strength of 400–930 MPa[6] while the strength of some natural fiber such as curaua, flax, and pineapple fiber can reach more than 1000 MPa.[7,8] However, the natural fibers are normally cut to smaller length intentionally during the manufacturing of prepreg mat, degraded to shorter length unavoidably during blending/compounding with thermoplastic matrix and/or molding process due to large shear stress involved in these processes. The attrition of the fiber length severely reduces the strength of natural fiber composites. In addition, the thermoplastic polymer matrices used in the composite are typically

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low temperature polymers such as polyethylene and polypropylene.[9–13] The low temperature used to melt these polymers during processing is able to avoid the degradation of the natural fiber and additionally result in easier processing. However, the low temperature polymer generally has a relatively low strength. As a result, the strength of the discontinuous natural fiber thermoplastic composite material with polyolefin matrix is relatively low, varying from 18 MPa to 74 MPa.[1,13–16] Most of the polypropylene matrix composites have a tensile strength of around 50 MPa.[16]

High-density polyethylene (HDPE) is a common type of polyethylene used in natural fiber composites,[9–13] including kenaf fiber-reinforced composites.[1,12,13] It has a low density less than 1 g/cm^3 and a melting temperature lower than 150°C.[1] The low melting temperature of the HDPE ensures good consolidation and impregnation but avoids degradation of the natural fibers. As such, the resultant natural fiber HDPE matrix composite has relatively good mechanical property along with low density. For example, Wang et al.[1] developed a high-performance kenaf fiber HDPE composite with a tensile strength of 52 MPa.

Ultra-high molecular weight polyethylene (UHMWPE) fiber, such as Spectra® and Dyneema®, is a high-performance fiber that has excellent mechanical properties in the fiber direction. The polymer chains in the UHMWPE fiber are highly crystallized and aligned in the fiber direction, which results in excellent tensile strength in that direction. For example, Spectra has a tensile strength of 2–2.9 GPa[17–19] and Dyneema has a tensile strength of 2.7 GPa.[20] UHMWPE has a low density that is 0.97 g/cm^3.[20] Additionally, its elongation at break is high and ranges from 3.5% to 4.5%.[19–21] Because UHMWPE is a polyethylene-based material, it has low melting temperature of approximately 142–150°C.[17,19,22–26] UHMWPE has been used to reinforce HDPE,[27,28] low-density polyethylene,[19,20,29–31] and thermoplastic starch.[32] All of these studies reported the significant strength improvement after UHMWPE fabric or fibers were added to the thermoplastic polymers. However, fewer studies have been found where UHMWPE fibers are used to reinforce natural fiber composite.

In this work, a natural fiber composite with significantly improved strength and strain to failure is developed by adding a minimal amount of UHMWPE fibers. The UHMWPE fiber is co-molded with the kenaf fiber HDPE composite at different processing temperatures to form a low-density PE-based hybrid material. Microstructural analysis is conducted through the thickness of the hybrid material and the effect of the processing temperature on its tensile and impact strength is evaluated.

Material and methods

Kenaf fibers with a fiber length of 25 mm supplied by CetoTech, Inc. and HDPE films with a thickness of 0.127 mm were used for this study. Spectra fabrics with 0/90 plain weave, as shown in Figure 1, were used to co-mold with the kenaf mats and HDPE films to form the hybrid material samples.

Compression molding was used to process the hybrid material samples. Six fiber mats were stacked with HDPE films and UHMWPE sheets. Each fiber mat has three HDPE films above and below. One UHMWPE sheet was placed on the bottom and top surfaces of the kenaf fiber and HDPE stack, respectively. Figure 1(a) and (b) shows the stacked fiber mats, HDPE films, and the UHMWPE sheets in a steel compression mold that has a dimension of 153 mm × 153 mm (6 inch × 6 inch). The fiber mats, the HDPE films, and the UHMWPE sheets were weighed individually before the layup. The two UHMWPE sheets have a mass of 5 g (2.5 g/sheet). The total mass of the kenaf fiber mats and the HDPE sheets are approximately 60 g, 40% of which is the mass of the kenaf fiber mats (24 g). The two UHMWPE sheets account for approximately 8% of the overall weight. The mold with the stack was placed in a hydraulic molding press (Pasadena Hydraulics, Inc.). A load of 10 tons was applied. It is noted that the UHMWPE sheets were stacked with the kenaf fiber mats and the HDPE sheets for molding altogether and no additional processing step is required to integrate the UHMWPE sheets onto the kenaf fiber HDPE composite (KF/HDPE).

Processing temperature plays a key role in the consolidation and impregnation of the KF/HDPE and the bonding between the HPDE and the UHMWPE. Higher temperature will ensure good consolidation and impregnation of the KF/HDPE composite and good bonding between the UHMWPE and the HDPE. However, there is a risk that the UHMWPE could melt and lose its reinforcement function. In order to study the effect of the molding temperature, three temperatures, 135°C, 145°C, and 155°C, were used for the compression molding of the hybrid material. The sample with the same layup was heated to each temperature and held at that temperature for 30 min with a molding force of 10 tons. The samples processed at 135°C, 145°C, and 155°C with kenaf fiber, HDPE, and UHMWPE (UH), were denoted as 135C-KF/HDPE/UH, 145C-KF/HDPE/UH, and 155C-KF/HDPE/UH, respectively. The samples processed at these temperatures were tested, characterized, and compared in their performance in tension and impact. A sample without any UHMWPE sheets was also processed at 135°C (135C-KF/HDPE) for comparison purpose.

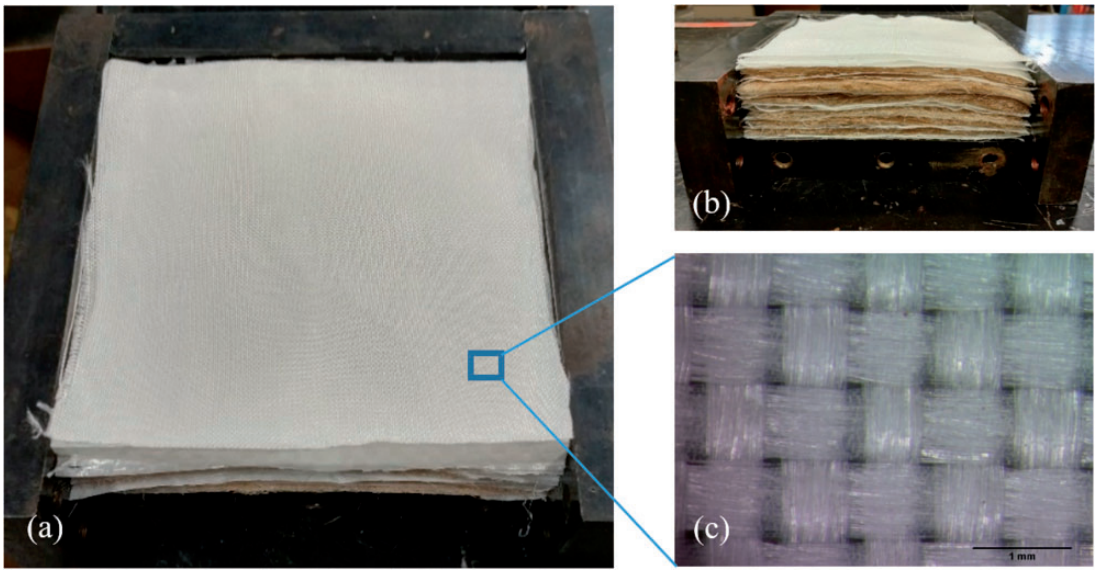

Figure 1. (a) Top view and (b) side view of the layup of the natural fiber mats, HDPE films and UHMWPE fabrics in a 153 × 153 mm^2 mold; (c) closer view of the UHMWPE fabric showing the 0/90 plain weave.

Results and discussion

Microstructural analysis

Microscopic analysis was conducted on the samples processed at different temperatures to characterize their consolidation and impregnation of the KF/HDPE, and the status of the UHMWPE. The samples were mounted, polished, and their thickness in cross sections examined under optical microscope and stereoscope. The microstructural images are shown in Figures 2, 3, and 4 for samples 135C-KF/HDPE/UH, 145C-KF/HDPE/UH, and 155C-KF/HDPE/UH, respectively. All of the samples exhibit adequate wet-out between the kenaf fiber and the HDPE matrix. Obvious UHMWPE fiber tows in both 0 and 90° directions can be noticed on the surfaces of both samples 135C-KF/HDPE/UH (Figure 2) and 145C-KF/HDPE/UH (Figure 3). HDPE flows and fills the spaces between UHMWPE fiber tows, which resulted in the smooth surface of the plates. However, there are no UHMWPE layers or fibers observed on the surface of sample 155C-KF/HDPE/UH, which verifies the result observed in Figure 4.

Sample 135C-KF/HDPE/UH shows obvious HDPE polymer rich areas in Figure 2. In some polymer-rich areas, weld lines between adjacent HDPE sheets are noticeable. These areas are labeled with "Weld line" in Figure 2(b) and (d). The weld lines between HDPE sheets are caused by limited polymer flow at low processing temperature. The positive effect of higher molding temperature on the flow and consolidation among the fiber layers can be observed in samples 145C-KF/HDPE/UH and 155C-KF/HDPE/UH. No weld line between HDPE sheets is noticed in either of these two samples. The fiber layers in sample 155C-KF/HDPE/UH (Figure 4(a)) are not as obvious as observed in samples 135C-KF/HDPE/UH and 145C-KF/HDPE/UH because of enhanced HDPE flow at high processing temperature.

Tensile testing

Tensile testing was carried out to evaluate the tension behavior of the PE-based hybrid material samples processed at the different temperatures. The tensile testing was conducted according to the ASTM D3039 – Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials. Tabbed specimens were prepared and tested at a loading rate of 1 mm/min on a MTS testing frame (MTS Systems Corp.). Sample 135C-KF/HDPE was also tested and the result was compared to those of the hybrid material samples.

Figure 5 illustrates the superimposed load and displacement curves of all of the four samples and Figure 6 compares their tensile strength. Sample 155C-KF/HDPE/UH has the lowest tensile strength among all of the samples with UHMWPE added. It actually has a tensile strength as low as that of sample 135C-KF/HDPE, the sample without any UHMWPE. It strongly indicates that the UHMWPE on sample 155C-KF/HDPE/UH has completely lost its reinforcement effect due to melting. This result also confirms the finding from the microstructure analysis that shows the disappearance of the UHMWPE fibers.

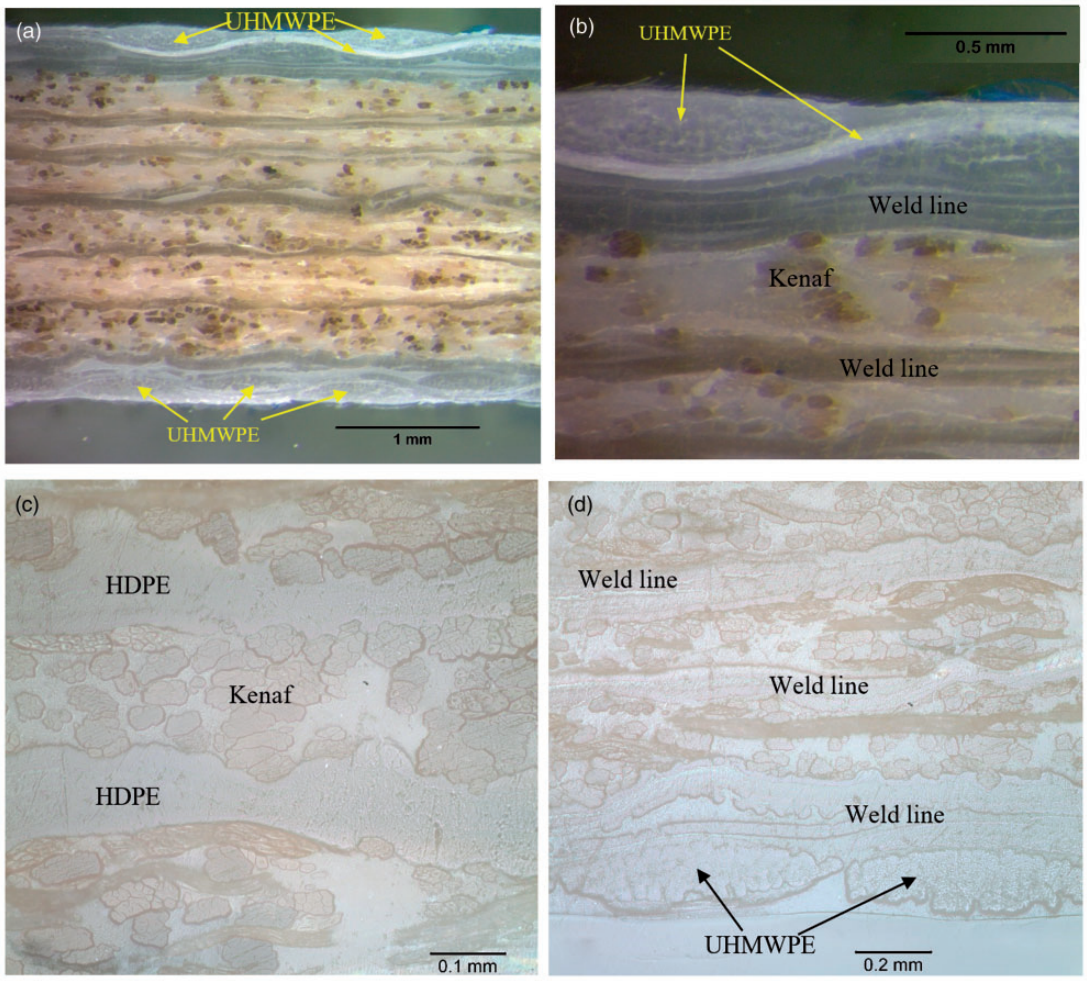

Figure 2. (a) The overall through-thickness microstructure of sample 135C-KF/HDPE/UH processed at 135°C; (b) a closer view of the sample surface showing the UHMWPE fiber tows in both 0 and 90 direction; (c) a close view of the layered kenaf and HDPE; (d) an optical microscopic image of the weld lines between the kenaf fiber layer and the HDPE layer, indicating inadequate flow of the HDPE at 135°C.

The addition of the UHMWPE sheets on the surfaces of KF/HDPE results in a tremendous increase in the tensile strength by comparing samples 135C-KF/HDPE/UH and 135-KF/HDPE. The tensile strength increased from 48 MPa to 93 MPa. The increase in tensile strength is more than 90%. The failure mode for sample 135-KF/HDPE/UH was progressive failure, starting from delamination between the UHMWPE and the KF/HDPE and the delamination within the KF/HDPE layers. Fracture of the UHMWPE fibers and the KF/HDPE occurred consequently. The load–displacement curve of 135C-KF/HDPE/UH in Figure 5 shows the progressive failure.

Both samples 135C-KF/HDPE/UH and 145C-KF/HDPE/UH show nearly identical tensile strength, 93 MPa and 89 MPa, respectively. In spite of their similar tensile strength, their failure modes are quite different. The hybrid sample molded at 135°C showed gradual failure with initial delamination followed by fracture as aforementioned. After the load reached the peak, delamination between the KF/HDPE composite and the UHMWPE sheets resulted in the sudden load drop from 6000 N to 3000N. However, the sample still has residue load bearing capability because of no fracture. Subsequently, more delamination occurred to the sample and eventually both of the UHMWPE fibers and the KF/HDPE layers fractured. In contrast, the hybrid material sample molded at higher temperature 145°C showed complete catastrophic fracture after reaching peak load, which is indicated by the sudden load drop to zero as shown in Figure 5. The higher processing temperature 145°C has resulted in better bonding of the HDPE matrix to the UHMWPE fibers and within the kenaf fiber HDPE layers. The better bonding strengthens the UHMWPE/HDPE interface and the HDPE/HDPE interface, and

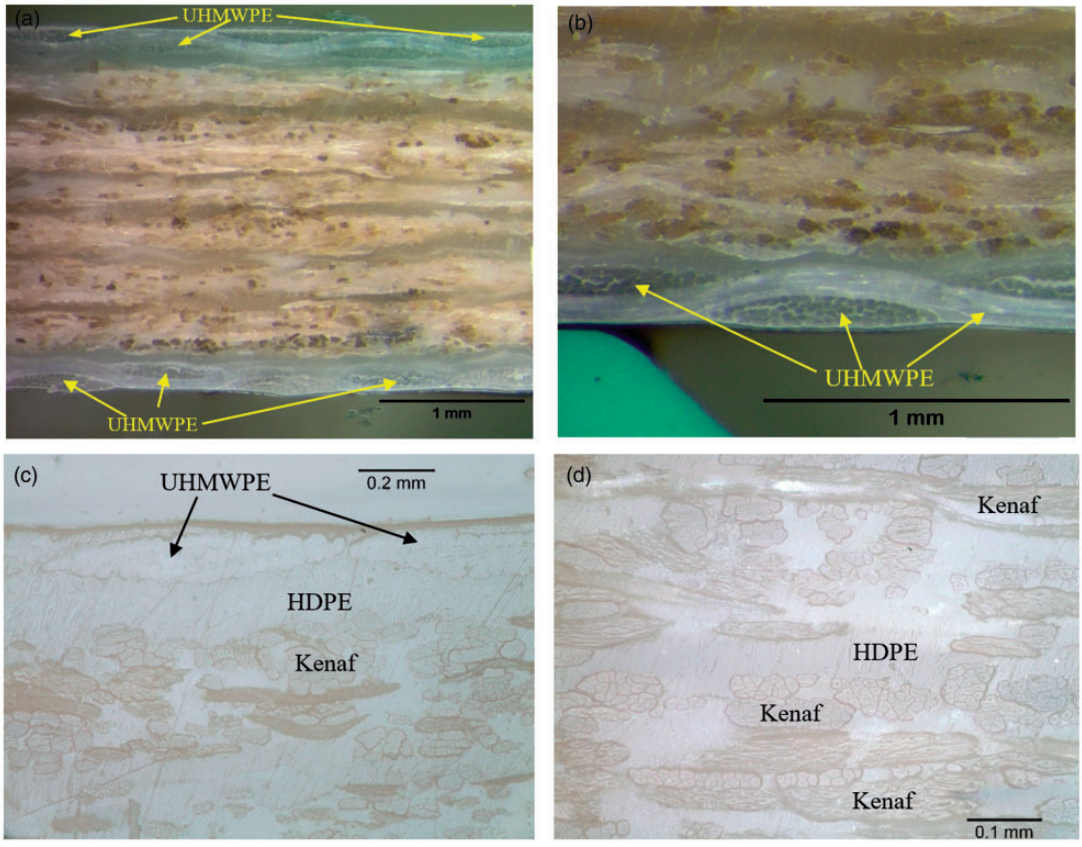

Figure 3. (a) The overall through-thickness microstructure of sample 145C-KF/HDPE/UH processed at 145°C; (b) and (c) a closer view of the sample surface showing the UHMWPE fiber tows in 0° and 90° direction; UHMWPE fiber tows are highlighted; (d) an optical micrograph showing relatively uniform kenaf fiber distribution.

therefore prevents any interlaminar delamination. Figure 7 compares the failure modes between tested 135C-KF/HDPE/UH and 145C-KF/HDPE/UH samples. Obvious delamination is noticed for 135C-KF/HDPE/UH while 145C-KF/HDPE/UH shows fracture failure mode.

The load-bearing capacity of the 135°C hybrid sample is similar to that of the 145°C hybrid sample in spite of weld lines between HDPE sheets observed. The slight higher strength of 135C-KF/HDPE/UH might be attributed to the lower processing temperature that allows UHMWPE sheets maintain its strength. Previous study has shown that the crystalline state of UHMWPE could be affected when it is exposed to temperature above 135°C.[19] The benefit of better HDPE flow, consolidation, and bonding gained from higher processing temperature at 145°C was probably counterbalanced by the affected crystalline state and reduced mechanical properties of UHMWPE.

The strain at peak load was significantly increased by the addition of UHMWPE processed at lower temperature. Figure 8 compares the strain at peak load for different samples. Sample 135C-KF/HDPE has a strain to peak load of 1.3% while the addition of UHMWPE resulted in a strain at peak load of 3.8%. The drastic increase of the strain is attributed to the excellent strain to failure of UHMWPE. However, that effect was reduced when the processing temperature was raised to 145°C and eventually vanished when the processing temperature reached 155°C. The probable reason is that the processing temperature 155°C is well above the melting temperature of UHMWPE, which is approximately 142–150°C,[17,19,22–26] and complete melting of the UHMWPE was resulted. The microstructural analysis in Figure 4 also verifies the disappearance of the UHMWPE. Therefore, the strain of sample 155C-KF/HDPE/UH is identical to that of sample 135C-KF/HDPE due to the melting of UHMWPE at 155°C.

Impact testing

One of the significant advantages of the UHMWPE fiber is its excellent impact resistance. It has been extensively used as the ballistic resistant material for its superior energy absorption capability.[33] In this work, Izod impact testing was used to study the impact resistance

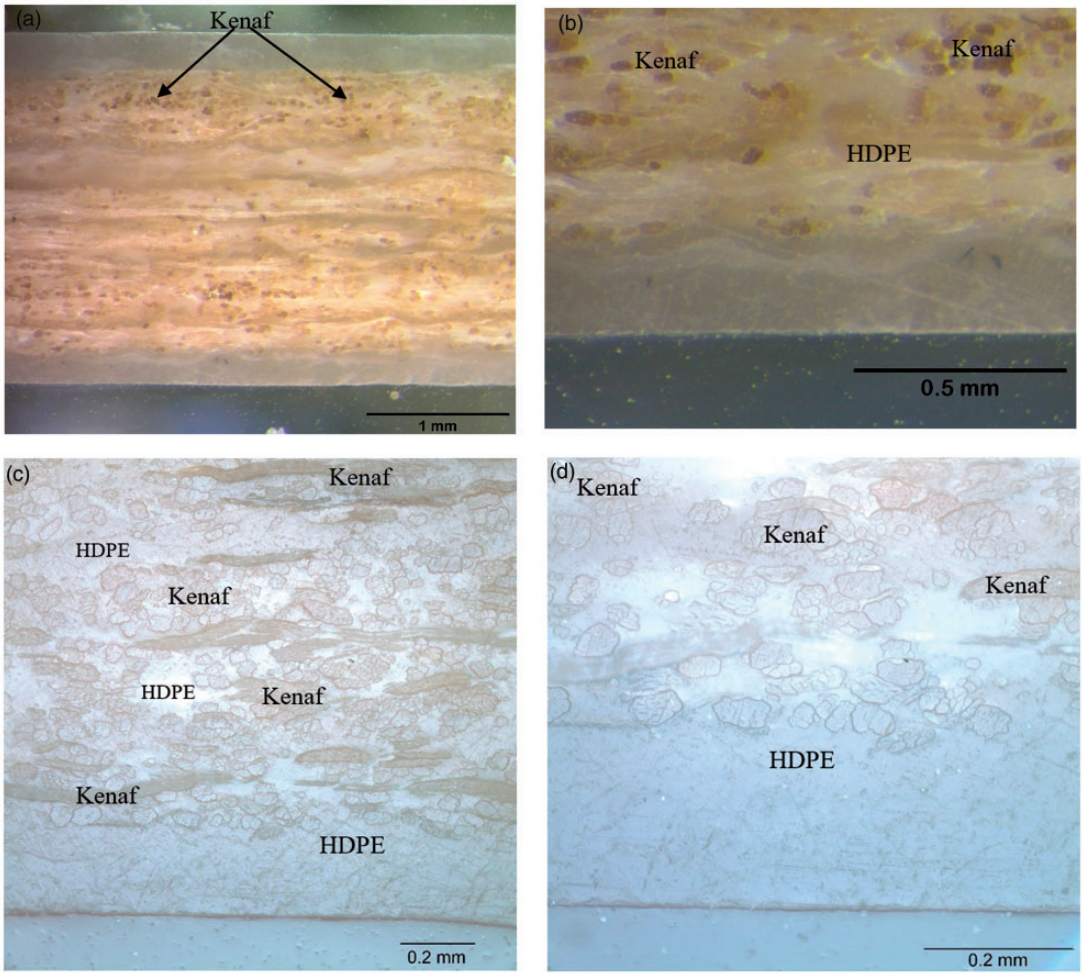

Figure 4. (a) The overall through-thickness microstructure of sample 155C-KF/HDPE/UH processed at 155°C; (b) a stereo microscopic image of the sample surface showing no noticeable UHMWPE fiber tows; (c) optical micrograph showing relatively uniform kenaf fiber distribution. (d) A closer view of the sample surface showing no noticeable UHMWPE fiber tows.

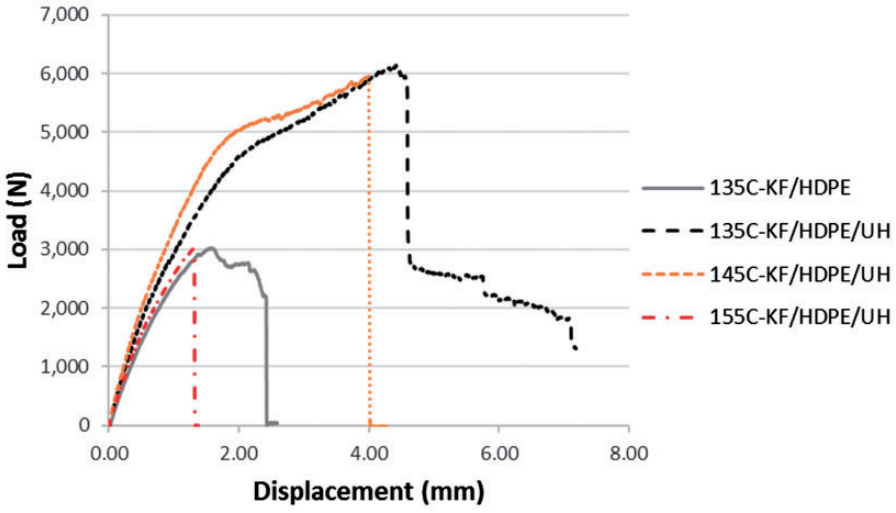

Figure 5. The superimposed load–displacement curves for the four types of samples.

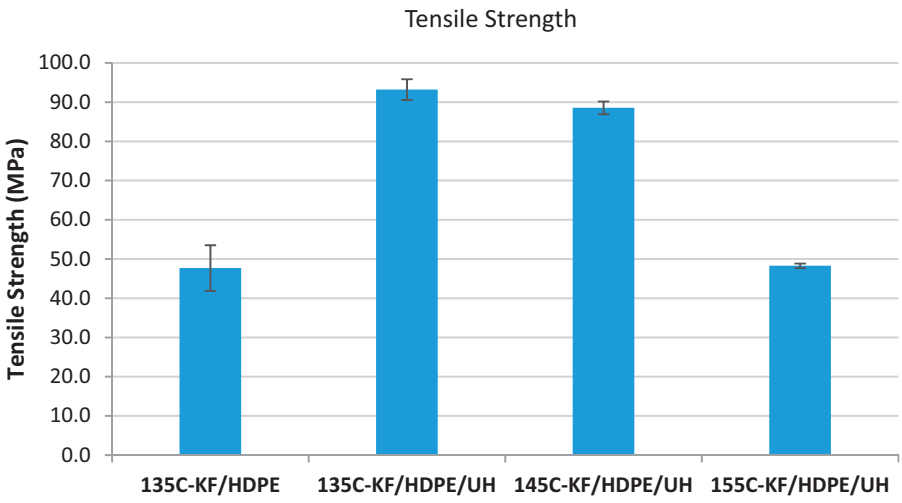

Figure 6. The comparison of tensile strength of the four samples.

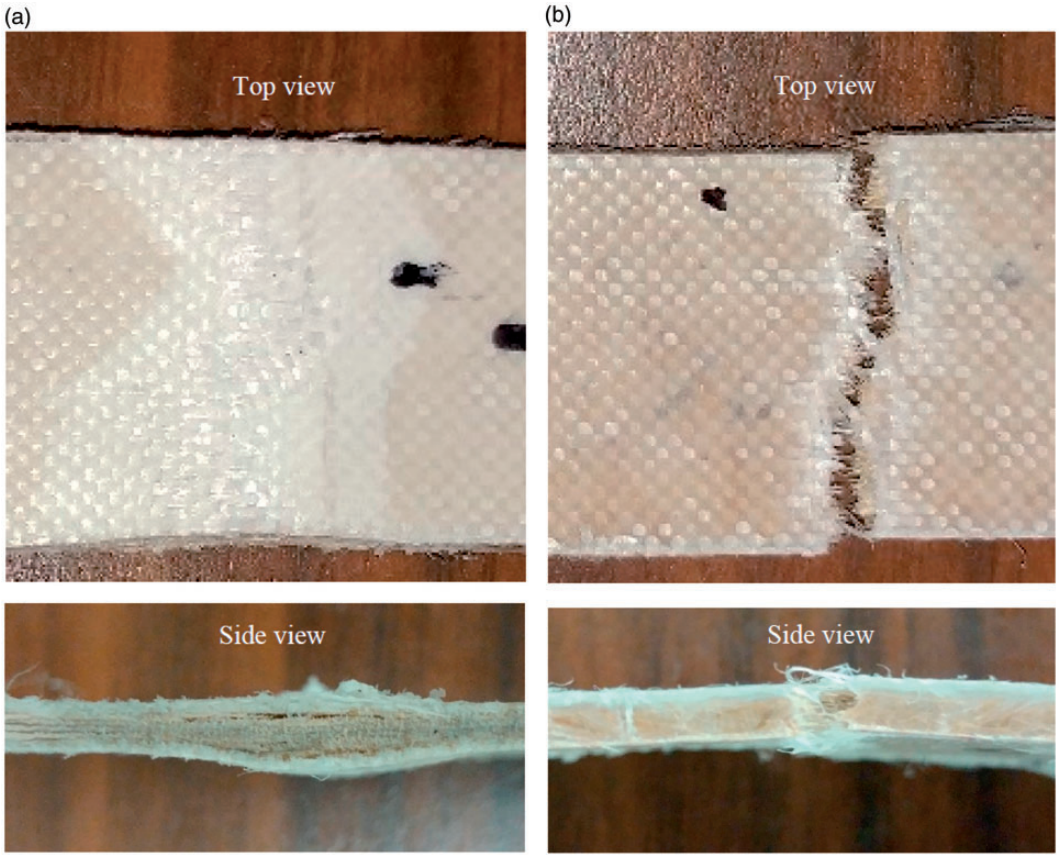

Figure 7. Different failure modes occurred: (a) sample 135C-KF/HDPE/UH and (b) sample 145C-KF/HDPE/UH. Sample 135C-KF/HDPE/UH shows obvious delamination while sample 145C-KF/HDPE/UH fracture.

of the hybrid material according to the ASTM D256 – Standard Test Methods for Determining the Izod Pendulum Impact Resistance of Plastics. Unnotched specimens for each type of sample were prepared and tested on an Izod impactor (Tinius Olsen Material Testing Machine Company). The impact strength of unnotched specimens was averaged and compared in Figure 9(a) for all of the four types of samples.

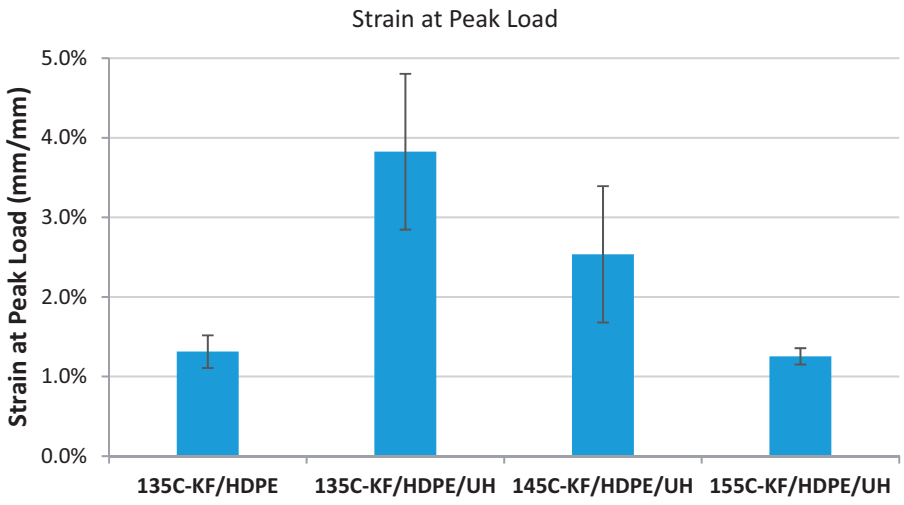

Figure 8. The comparison of strain at peak load of the four samples.

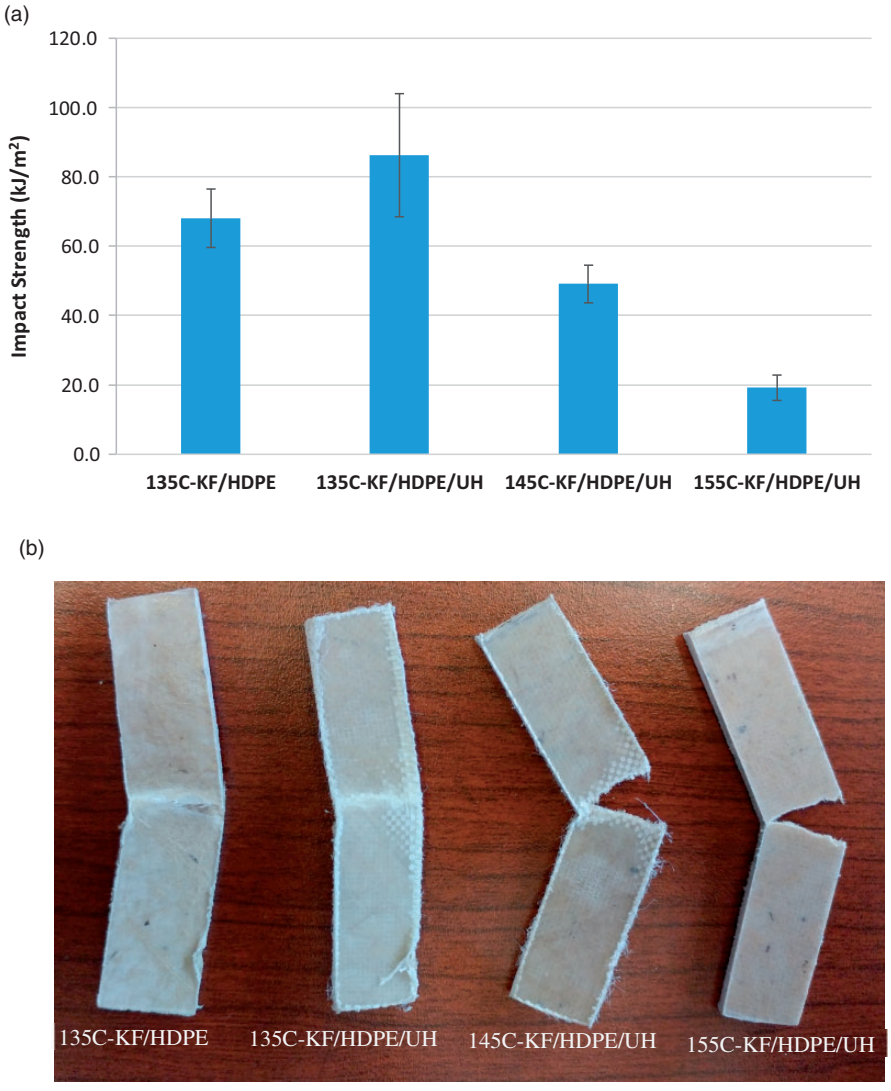

Figure 9. (a) The comparison of impact strength showing that sample 135C-KF/HDPE/UH has the highest impact strength; (b) the tested samples from left to right: 135C-KF/HDPE, 135C-KF/HDPE/UH, 145C-KF/HDPE/UH, and 155C-KF/HDPE/UH.

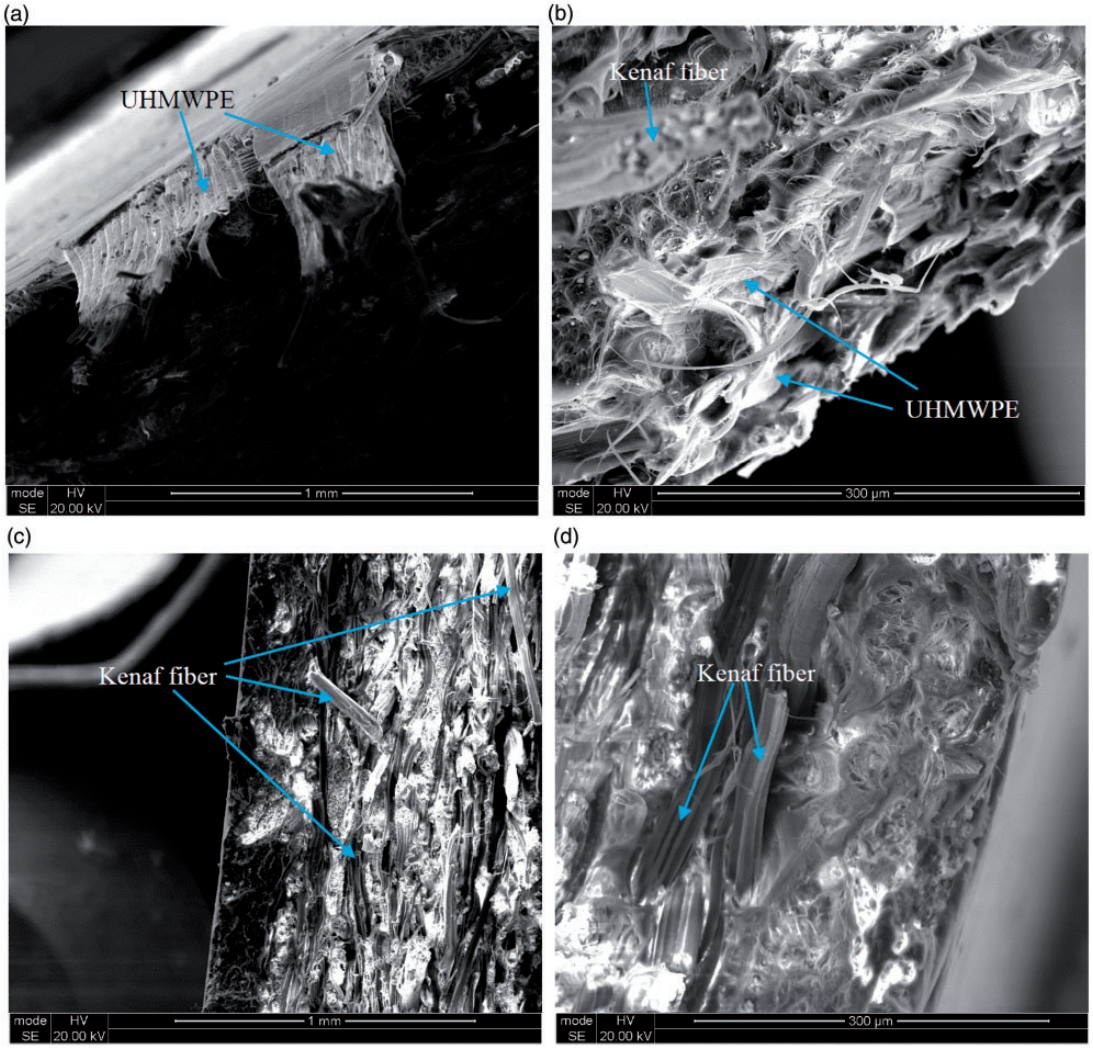

Figure 10. SEM images show the UHMWPE fibers in sample 145C-KF/HDPE/UH in (a) and (b); and no obvious UHMWPE fibers was noticed in sample 155C-KF/HDPE/UH in (c) and (d).

Sample 135C-KF/HDPE/UH showed the highest impact strength, 87 kJ/m^2 and sample 155C-KF/HDPE/UH has the lowest impact strength, 21 kJ/m^2. Both samples 135C-KF/HDPE and 135C-KF/HDPE/UH mainly showed delamination, similar failure behavior observed in the tension testing. The UHMWPE sheets showed no fracture but only delamination on sample 135C-KF/HDPE/UH. Samples 145C-KF/HDPE/UH and 155C-KF/HDPE/UH had fracture failure mode, which also matches with the failure mode noticed in the tensile testing. Both 135C-KF/HDPE/UH and 145C-KF/HDPE/UH showed the "chess board" pattern around the impact area (high stress area) caused by the pulling/debonding of the UHMWPE fiber tows in the sample length direction from HDPE matrix during test. Figure 9(b) shows the tested specimen from each type of sample. Figure 10 shows the scanning electron microscopic (SEM) images of the tested sample.

Density measurement

One of the advantages for natural fiber PE matrix composite is its low density because of the low density of both natural fiber and PE. The UHMWPE fibers also have low density, which would expectedly result in a low density for the KF/HDPE/UH hybrid material samples. The densities of the hybrid samples were measured according to the ASTM D792 – Standard Test Methods for Density and Specific Gravity (Relative Density) of Plastics by Displacement. Dry mass of the sample was measured and wet mass was consequently obtained by immersing the sample in distilled water. The average density of each KF/HDPE/UH sample is in the range of 1.04–1.09 g/cm^3 as shown in Figure 11. The hybrid sample density increases slightly with processing temperature. The low density of the hybrid material enables the material to possess superior

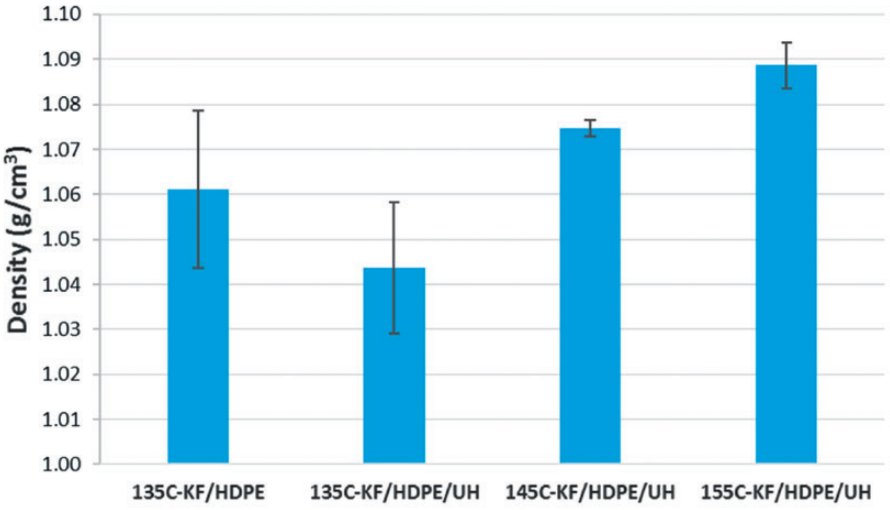

Figure 11. The density comparison among the four samples. Note the *y*-axis starts from 1.00 g/cm^3.

specific strength. Sample 135C-KF/HDPE/UH has an averaged tensile strength of 93 MPa, therefore, its specific tensile strength is calculated to be 89 MPa/(g/cm^3). It is comparable to aluminum alloy 3xxx series[34] and some discontinuous glass fiber-based composites.[35] The superior specific strength provides the avenue for the hybrid material replacing traditional materials without compromising the performance. However, it is noted that the cost of the UHMWPE material is relatively high. Two 153 mm × 153 mm (6 × 6") UHMWPE sheets used for processing one panel costed approximately $2.80 based on its retail price in 2017, which could be a major hurdle for its widespread usage.

The SEM images of the fractured impact samples are shown in Figure 10. The 145C-KF/HDPE/UH shows the fracture of the UHMWPE fiber and the kenaf fibers. There is no obvious UHMWPE noticed in sample 155C-KF/HDPE/UH (Figure 10(c) and (d)). Fiber fracture and matrix fracture are the failure mechanisms observed in samples 145C-KF/HDPE/UH and 155C-KF/HDPE/UH.

Conclusions

A polyethylene-based hybrid material was developed by adding UHMWPE woven fabrics to the surfaces of the KF/HDPE composites with a single-step compression molding process. The interrelationship among the processing, structure, and property of this newly developed material is studied. The effect of three different processing temperatures on the tension and impact performance of the hybrid material is investigated. The sample processed at 155°C shows the melting of UHMWPE fibers, which results in the complete loss of its reinforcement function. The lower processing temperatures at 135°C and 145°C enable adequate melting of HDPE and consolidation and impregnation of the HDPE onto the kenaf fiber but avoid the melting of the UHMWPE based on the microstructure analysis. Both of the samples have similar tensile strength of approximately 90 MPa but with different failure modes. The integration of 8% UHMWPE could result in an increase in the tensile strength of the KF/HDPE composite by 90% and its strain at peak load by nearly three times. The optimal processing temperature for this hybrid material is considered to be between 135°C and 145°C to gain the maximum reinforcement from the addition of UHMWPE. The low density of the hybrid material, ranging from 1.04 to 1.09 g/cm^3, results in a superior specific strength.

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Declaration of Conflicting Interests

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