

# Improvement in the mechanical performance and interfacial behavior of kenaf fiber reinforced high density polyethylene composites by the addition of maleic anhydride grafted high density polyethylene

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**Abstract** The effects of compatibilizer on the tensile, flexural and interfacial adhesion behavior of kenaf fiber reinforced high density polyethylene composites were investigated. The addition of maleic anhydride grafted high density polyethylene (MA-HDPE) as compatibilizer into the composites was found to improve the mechanical properties and the adhesion behavior of the composites. These improvements were due to the improved compatibility between matrix and fiber. 8 % MA-HDPE loading provided maximum enhancement in tensile and flexural properties when compared to the other compatibilizer contents. Meanwhile, uncompatibilized composites showed poorer mechanical properties and interfacial behavior relative to the compatibilized composites. Fourier transformed infrared spectroscopy analysis confirmed the changed chemical structures by the appearance of stretching vibration of the ester carbonyl groups (C=O) around  $1725\text{ cm}^{-1}$  to  $1742\text{ cm}^{-1}$  and the peak of hydroxyl group at  $3327\text{ cm}^{-1}$  in the compatibilized composites. This indicates that the maleic anhydride has bonded to the kenaf fiber through esterification reaction, giving rise to strong interfacial bonding between the matrix and fiber. The improvement in the interfacial behavior was evident from the tensile fracture

surface morphology using a field emission scanning electron microscopy.

**Keywords** Mechanical properties · Interfacial behavior · Compatibilizer · Kenaf fiber

## 1. Introduction

Kenaf fiber is a type of lignocellulosic natural fiber which has the potential to be used as a replacement for the traditional reinforcement materials in composites. The addition of kenaf fiber is believed to confer strength and rigidity to the weak and brittle plastic matrix, reducing the high manufacturing cost of plastic application and increasing the value of natural fiber. Furthermore, the global demand for natural fibers is increasing due to a greater request for green industrial products.

However, drawback factors such as the tendency to form aggregates during processing, poor resistance to moisture, and weak interaction and adhesion in lignocelluloses fiber are some of the disadvantages when using this type of fibers. The weak interaction and adhesion between the fiber and matrix arise due to the hydrophilic nature of the fiber and the hydrophobic characteristic of the polymer. This leads to poor compatibility between the fiber and the matrix, thus resulting in unfavorable properties of the composites.

In order to enhance the compatibility between natural fiber and plastic, identification of a suitable compatibilizer is very important to improve the interfacial adhesion, thereby developing an effective interface structure with improved physical and mechanical properties of the composites. Several techniques ranging from chemical treatment, grafting of short-chain molecules and polymers onto the fiber surface by using

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coupling agents and radical induced adhesion promoters have been reported for improving interfacial bonding [1–3]. As reported [4, 5], grafting is one of the techniques to improve wetting between the fiber and matrix by promoting interfacial bonding through diffusion of the chain segments of the grafted molecules with the matrix. Coupling agents and radical induced adhesion enhance interfacial bonding by producing covalent bonds between the fiber and the matrix.

Maleated coupling agents have always been used in reinforced composite material. Two important functions of maleic anhydride – polyolefin in the composite is to produce economical products and to create good interaction between maleic anhydride (MA) surfaces that link polyolefin with reinforced fiber composites. Keener et al. [4] showed that the addition of 3 % maleic anhydride polyethylene (MA-PE) coupling agents in wood composites doubled the tensile strength when compared to the ones without coupling agents. Maleic anhydride grafted to polypropylene (MA-PP) was also reported [5] to show great efficiency in kenaf/polypropylene composite. MA-PP showed polarized interaction and bonded covalently with hydroxyl groups in lignocellulosic fiber. The chemical interactions that occur between the anhydride groups from the coupling agent and the hydroxyl groups of the natural fiber can help to overcome the unfavorable nature of both materials; hence an increase in the tensile and flexural strength results. Majid et al. [6] suggested that the reason for the usage of MA-PE as a compatibilizer in the composite are based on two factors. Firstly is the ability of the anhydride groups from the compatibilizer to undergo an esterification reaction with the hydroxyl groups of natural fiber, and secondly is due to good compatibility between grafted PE chains and the PE phase.

In this research, the investigation on the effects of addition of MA-HDPE on the mechanical and interfacial properties of kenaf fiber/high density polyethylene (HDPE) composites has been done at different percentages of MA-HDPE content. Fourier transformed infrared (FTIR) characterization was used to assign the functional groups of chemical species subjected to the various modifications. Field emission scanning electron microscopy (FESEM) was applied to investigate the morphology of the fracture surface and elucidate the interfacial adhesion between fiber and matrix. The purpose of this work is to prove the usefulness of MA-HDPE through improvement in mechanical properties and interfacial behavior of kenaf fiber reinforced HDPE composites and supported by the FTIR analysis.

## 2. Experimental

### 2.1. Materials

Kenaf bast fiber of 3 mm length with an average density of 134.3 kg/m<sup>3</sup> 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 HDPE, Titanex HI 1100 with a density of 961 kg/m<sup>3</sup> and melt flow index of 7 g/10 min, manufactured by Titan Petchem (M) Sdn. Bhd, Malaysia was used as the matrix. Maleic anhydride grafted high density polyethylene, MA-HDPE (NG 1002), with 1 % MA graft, density of 880 kg/m<sup>3</sup> and melt flow index 1.5 g/10 min was manufactured by Shanghai Zeming Plastic Co. Ltd China and used as the compatibilizer. All materials were used as received.

### 2.2. Extrusion

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 aspect ratio of 40. Extrusion was carried out at screw speeds of 80 rpm at 2 kg/h feeding rate with temperature settings of 165 °C, 170 °C, 175 °C, 180 °C and 185 °C from the hopper to the die. Pure HDPE and MA-HDPE were loaded 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. The strands leaving the circular extruder die with diameter of 3 mm were pelletized. The pelletized composites were then oven-dried at 80 °C for 24 h and stored in a sealed plastic bag inside desiccator for injection molding. Composites were prepared at two different fiber compositions of 8.5 wt.% and 17.5 wt.% and the MA-HDPE compatibilizer was added at different loadings of 0 %, 4 %, 8 % and 12 % of the total weight of the composites.

### 2.3. Injection molding

The dried pelletized composites were then injection molded with a single gated, four cavities mold for tensile test specimens using injection molding machine (Boy® 55 M, Germany). The barrel was set at temperatures between 160 °C - 190 °C, an injection pressure of 100 bar - 120 bar, cooling time of 120 s and mold temperature of 20 °C.

### 2.4. Determination of the functional group of composites

Infrared absorption spectra of the uncompatibilized and compatibilized kenaf fiber composites were recorded using a fourier transform infrared, FTIR, spectrophotometer (PerkinElmer-Spotlight 400, USA) combined with a universal attenuated total reflectance, ATR accessory at a resolution of 4 cm<sup>-1</sup> with 64 sample scans for each spectrum in the wavelength of 4000–500 cm<sup>-1</sup>.

## 2.5. Determination of the tensile properties

Tensile tests were conducted according to ASTM D-638 using a universal testing machine (Instron 5569, USA) equipped with a load-cell of 50 kN at a constant cross-head speed of 5 mm/min, and a gauge length of 50 mm. For each test, a minimum of seven samples were tested and an average of at least five reproducible results were presented. The test was conducted under ambient conditions.

## 2.6. Determination of the flexural properties

A universal testing machine (Instron 5569, USA) was used to perform the flexural test under ambient condition. Three point bending flexural tests were set up according to ASTM D-790. Specimen support span,  $L$ , was fixed at 50 mm with maximum deflections of 30 mm and constant cross-head speed of 1.27 mm/min. For each test, a minimum of seven samples were tested and an average of at least five reproducible results were presented.

## 2.7. Fracture surface observation

The fractured surface of the tensile specimens was observed using the field emission scanning electron microscope, FESEM (Zeiss-Auriga,39-22, Germany) under an acceleration voltage of 1 kV. The non-coated samples were mounted on the aluminium sample holder and placed in the specimen chamber in a vacuum condition of 0.06 mbar at room temperature. Digital images were taken from the fractured surfaces of the tensile test samples at a magnification of 100 $\times$ , 500 $\times$  and 1000 $\times$ .

## 3. Results and Discussion

### 3.1 FTIR

FTIR spectra of pure HDPE and compatibilizer (MA-HDPE), are shown in Fig. 1. MA-HDPE showed the appearance of symmetric stretching of carbonyl absorption at 1712  $\text{cm}^{-1}$  and 1791  $\text{cm}^{-1}$ . The appearance of a small peak at 1791  $\text{cm}^{-1}$  is associated with the presence of C=O of 1 % anhydride functional group grafted onto the HDPE. However, this peak was not observed in the pure HDPE spectrum. Previous researchers observed the two absorbances near 1774  $\text{cm}^{-1}$  and 1790  $\text{cm}^{-1}$ , and these were attributed to MA symmetric C=O stretching of MA-PP and MA-PE, respectively [7–9].

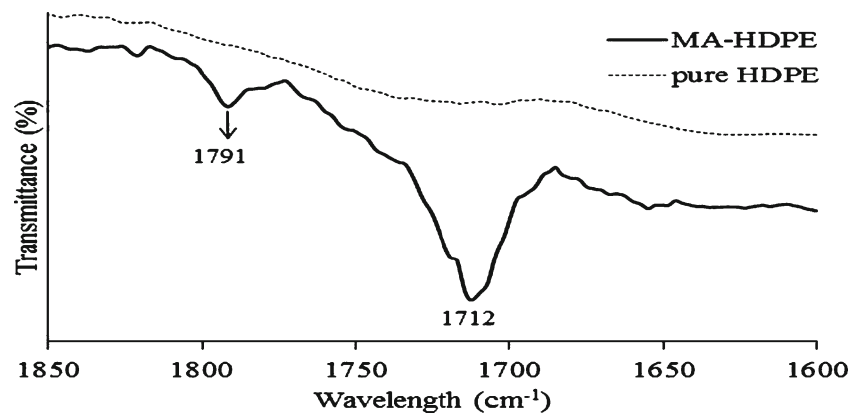
Figures 2 and 3 shows two key features of the FTIR spectra of uncompatibilized and compatibilized composites at 8.5 wt.% and 17.5 wt.% fiber loadings, respectively. The peak in the range of 3700  $\text{cm}^{-1}$  to 3100  $\text{cm}^{-1}$  indicates the presence of hydroxyl group and the peak in the range of 1800  $\text{cm}^{-1}$  to 1680  $\text{cm}^{-1}$  indicates the presence of carbonyl groups. From

Fig. 2, the appearance of symmetric stretching of carbonyl absorption of MA-HDPE at 1712  $\text{cm}^{-1}$  shifted to the range of 1725  $\text{cm}^{-1}$  - 1742  $\text{cm}^{-1}$  and disappearance of peaks at 1791  $\text{cm}^{-1}$  was observed in the case of compatibilized composites. This new appearance of the ester carbonyl groups shows that there could be interfacial bonding between the kenaf and matrix via the MA-HDPE. It was also observed that the new ester carbonyl groups peaks around 1725  $\text{cm}^{-1}$  - 1742  $\text{cm}^{-1}$  shifted with the addition of compatibilizer from 4 % to 12 % into the system, with the highest peak intensity obtained at 12 % compatibilizer content.

Broad absorption bands of hydroxyl (–OH) stretching are present in both uncompatibilized and compatibilized composites. Figure 2 shows that the hydroxyl peak of uncompatibilized composites is at 3345  $\text{cm}^{-1}$ . After the addition of compatibilizer, only 8 % compatibilizer loading presented a shift in the hydroxyl peak to a higher wavelength with the highest peak intensity when compared to the other compatibilizer loadings. It is possible esterification reaction may have taken place and resulted in two products; one is the copolymer with diester structures and the other is the copolymer with half ester structure and half carboxylic structure [10]. The formation of half carboxylic structure after esterification reaction can give rise to intramolecular hydrogen bonding between hydrogen atoms of a hydroxyl group from kenaf fiber with the oxygen atom of a MA-HDPE from compatibilizer. This could also be the reason for the highest performance of 8 % compatibilizer at 8.5 wt.% fiber loading.

Figure 3 shows that there were no appearances of ester group peaks in the uncompatibilized composites. However, with the addition of compatibilizer, the ester group peaks were observable at 1737  $\text{cm}^{-1}$  (4 % MA-HDPE) and 1728  $\text{cm}^{-1}$  (8 % and 12 % MA-HDPE) where the peak intensity increased with increasing amount of MA-HDPE. These ester group peaks were also found to have shifted to the higher wavelength compared to the appearance of symmetric stretching of carbonyl absorption of MA-HDPE at 1712  $\text{cm}^{-1}$ . From Fig. 3, uncompatibilized composites showed the presence of hydroxyl group at 3373  $\text{cm}^{-1}$  due to the hydroxyl compound from the kenaf fiber. The peak of the hydroxyl group of all compatibilized composites shifted to lower wavelength (around 3345  $\text{cm}^{-1}$ ) with the addition of compatibilizer when compared to the uncompatibilized composites. Similar to the finding above, the addition of 8 % compatibilizer loading showed the highest peak intensity of hydroxyl group at 3327  $\text{cm}^{-1}$  suggesting that this percentage of compatibilizer loading gave the most effective esterification and intramolecular hydrogen bonding reaction inside the compatibilized system. At higher percentage fiber loading of 17.5 wt.%, the peak intensity of hydroxyl groups for compatibilized composites was much higher compared to the uncompatibilized composites. This was due to higher amounts of fibers contributing to higher existence of hydroxyl groups.

**Fig. 1** FTIR spectra for pure HDPE and MA-HDPE (compatibilizer)



Wang et al. [11] also reported quite similar behaviors for three compatibilizers, i.e. MA-LLDPE, acrylic acid (AA)-HDPE and MA-HDPE as coupling agents for wood floor/HDPE composites. The authors reported that the absorption peaks at  $1734\text{ cm}^{-1}$  for MA-LLDPE and AA-HDPE and  $1724\text{ cm}^{-1}$  for MA-HDPE compatibilized systems may be associated with the ester link between hydroxyl groups of wood and the anhydride or acid groups of polyolefin compatibilizers. The formation of ester linkage at the interface has also been reported by many researchers [7, 9, 12–15]. The better interaction between the components of the polymer composite has led to the correlation in peak shifts and change in peak shapes [11, 12].

### 3.2 Tensile properties

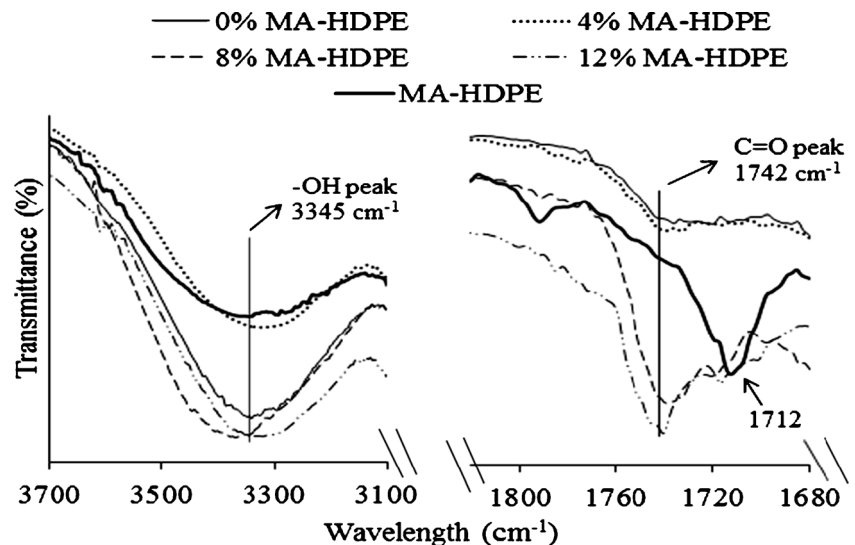
#### 3.2.1 Tensile strength

Tensile properties refer to the properties of a material that responds to the forces being applied in tension. In this paper the tensile modulus, tensile strength and tensile strain will be discussed. Figure 4 shows the effect of compatibilizer on the

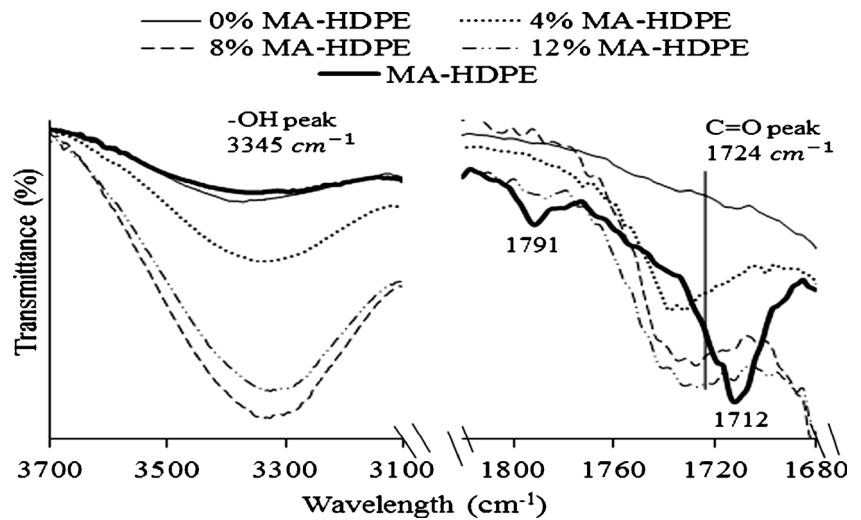
tensile strength and tensile modulus of composite at 8.5 wt.% and 17.5 wt.% fiber loadings. Composites with higher fiber loading (17.5 wt.%) presented higher tensile strength when compared to composites with lower fiber loading (8.5 wt.%). The tensile strength of the composites increased with the fiber fraction beyond the critical fiber content [16]. Increase in tensile strength is attributed to the increase in weight percent of fiber within the matrix, leading to an efficient stress transfer from the matrix to the fiber [17].

All compatibilized composites showed higher tensile strength relative to the uncompatibilized composites. The low tensile strength of uncompatibilized composites might be due to the poor interfacial adhesion between the polymer matrix and fiber, resulting from poor compatibility between the hydrophobic polymer matrix and the hydrophilic fiber [9, 17]. From the micrograph of the tensile fracture surface, in the case of the uncompatibilized composites at lower fiber content of 8.5 wt.% (Fig. 5(a)), a plastic deformation of the host matrix during tensile test which created an irregular surface texture with voids is observed. However, at higher fiber loading of 17.5 wt.% (Fig. 6 (a)), more holes were observed although the texture of the matrix was present in irregular shapes. In Figs. 5

**Fig. 2** FTIR spectra of 8.5 wt.% kenaf fiber/HDPE composites at different MA-HDPE contents



**Fig. 3** FTIR spectra of 17.5 wt.% kenaf fiber/HDPE composites at different MA-HDPE contents



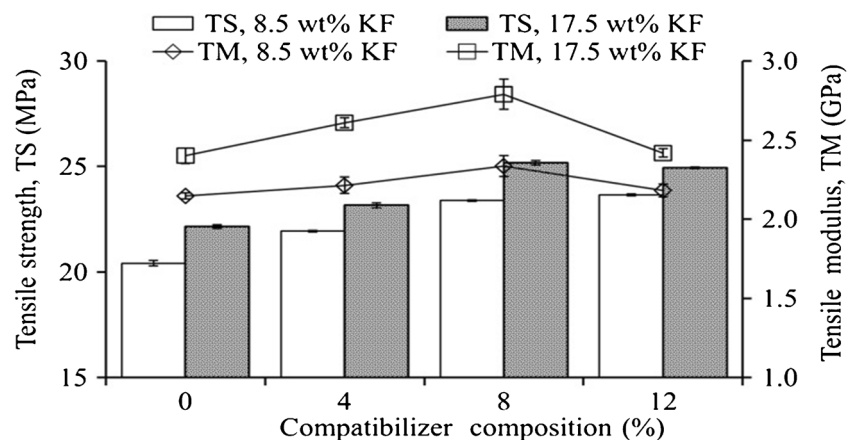
(b) and 6 (b), it is also evident that not much of the matrix adhered to the outer surface of the fibers. There are also cavities or gaps between the fiber and matrix (Figs. 5 (c and d) and 6 (c and d)), which are indications of weak interfacial adhesion between the fiber and matrix. In the micrograph of the composite without the compatibilizer, the fiber breakage and pull-out promoted by weak adhesion between the polymer and the fiber were observed [9, 16–20].

The tensile strength of composites (Fig. 4) increased by 7.4 %, 14.5 % and 15.9 % with the addition of 4 %, 8 % and 12 % compatibilizer respectively, when compared to the uncompatibilized composite (20.42 MPa) at 8.5 wt.% fiber loading. The increased tensile strength of composites indicates a good adhesion between kenaf fiber and HDPE when MA-HDPE was added to the composites. The improvement in tensile strength occurred in the presence of a compatibilizer, whereby improving the interfacial bonding between the fiber and the matrix, resulting in better stress transfer from the matrix to the fiber [5, 21–24]. Strengthening of the composite has been reported to occur through a better interfacial bonding between cellulosic fiber surfaces and PP, which was attributed to the esterification of the anhydride groups of MA-PP with

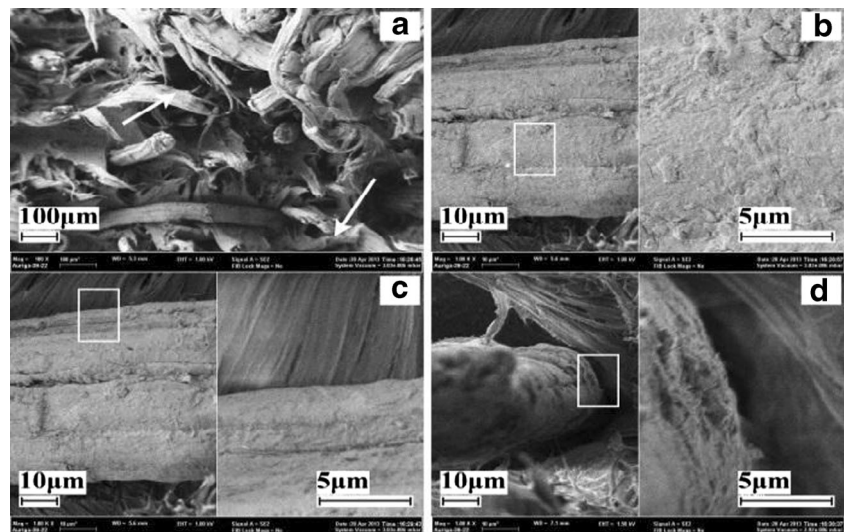
the hydroxyl groups of cellulosic fibers [24, 25]. This is supported by the FTIR results (Figs. 2 and 3), where effective esterification and intramolecular hydrogen bonding reaction was observed in the compatibilized composite system.

The presence of the compatibilizer changed the morphology of the materials. The use of MA-HDPE as compatibilizer greatly improved the interfacial bonding between the fiber and the matrix, as observed through the fractured surface of compatibilized composites (Fig. 7). As seen in the magnified micrograph of Figs. 7a (i)–c (i), the strong bonding between the matrix and fibers are clearly displayed, proving the strong interfacial adhesion as well as good wettability of fibers in the matrix. It is also noticeable that the degree of polymer coverage on the pulled-out fibers and their adhesion with the host matrix at all percentages of compatibilizer contents are very good; whereby, the fibers are still attached or in contact with the matrix after failure during tensile test (Figs. 7a (ii)–c (ii)). It was also observed that most of the fibers were coated with the matrix and pulled-out together with the bulk matrix, thus revealing the efficiency of the interfacial adhesion between the fibers and polymers.

**Fig. 4** The tensile strength and tensile modulus of uncompatibilized and compatibilized composites



**Fig. 5** FESEM micrograph of tensile fractured specimen of uncompatibilized composites at 8.5 wt.% fiber loading, a) irregular fractured surface, b) fiber uncoated by matrix, c) no adhesion and d) cavities



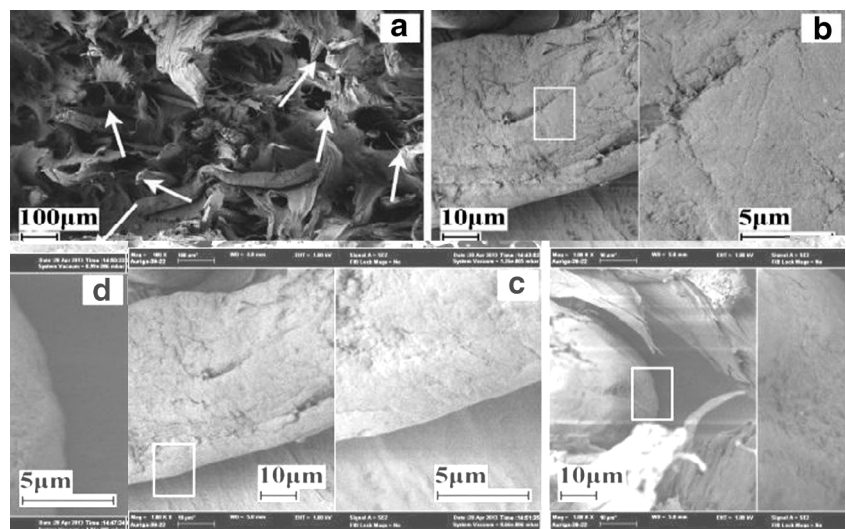
At 17.5 wt.% fiber loading, the tensile strength of composite (Fig. 4) increased with increasing compatibilizer loading up to 8 %. The tensile strength improved by 13.6 % relative to the uncompatibilized composites (22.15 MPa) with addition of 8 % compatibilizer. Further addition of 12 % compatibilizer into 17.5 wt.% fiber loading shows a slight reduction in the tensile strength. According to Sathe et al. [26], most of fibers are located at the interphase between the matrix and the dispersed phase up to a saturation level of the compatibilizer. However, when the concentration of the compatibilizer is above the saturation level, only a part of the molecules remains in the interfacial area and the excess is dispersed in the matrix, affecting its homogeneity and consequently the mechanical properties of the blends. The addition of 8 % and 12 % MA-HDPE (Figs. 7b (iii-vi) and 7c (iii-iv)), respectively, showed that the fiber was embedded in the HDPE matrix phase and there were no cavities in the interfacial area between fibers and the polymer matrix. In addition, fiber breakage is

clearly observed in Fig. 7b (v-vi), an evidence of better interfacial adhesion. All these indicate that a good adhesion has been achieved. This proves that MA-HDPE had a positive effect on the interfacial adhesion between the fiber and matrix. This explains the improvement in tensile strength and modulus of the compatibilized composites compared to the uncompatibilized composites. An improvement in the interfacial adhesion between fiber and matrix were also reported [11, 14, 16–18, 22], where the addition of compatibilizer into the composites system showed less extensive fiber pull-outs because the anhydride group present in compatibilizer strongly adheres to the –OH on the natural fiber surface.

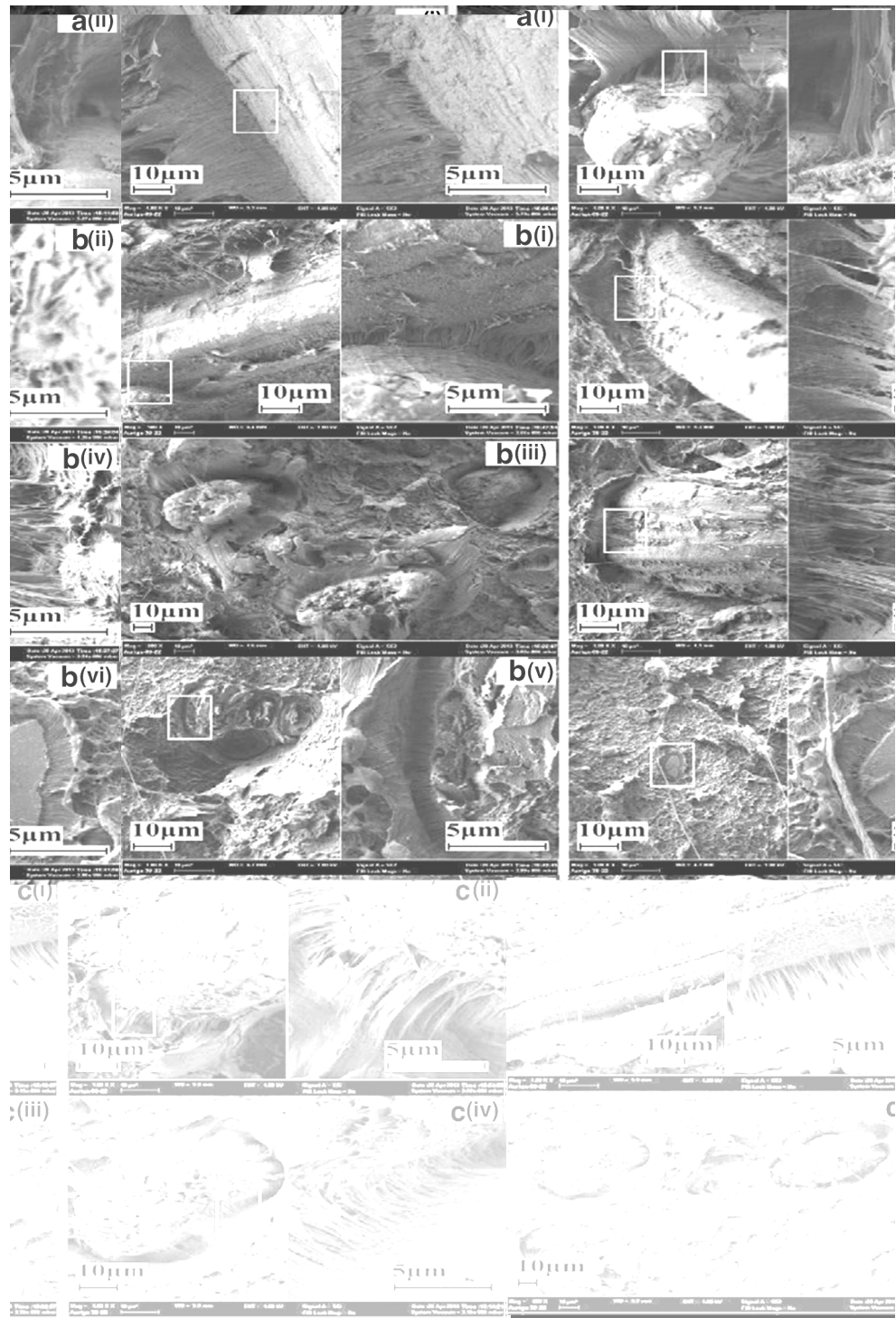
### 3.2.2 Tensile modulus

The addition of compatibilizer in composites containing 8.5 wt.% fiber loading showed that the tensile modulus increased with increasing MA-HDPE content up to 8 %.

**Fig. 6** FESEM micrograph of tensile fractured specimen of uncompatibilized composites at 17.5 wt.% fiber loading, a) irregular fractured surface and holes, b) fiber uncoated with matrix, c) no adhesion and d) cavities



**Fig. 7** FESEM micrograph of tensile fractured specimen of 17.5 wt.% compatibilized composites, a) 4 % MA-HDPE, b) 8 % MA-HDPE and c) 12 % MA-HDPE, i) attach to matrix, ii) pull out from matrix and iii-vi) embedded into matrix



Compatibilized composite with 8 % MA-HDPE showed 8.8 % improvement in tensile modulus relative to the uncompatibilized composite (2.15 GPa) at 8.5 wt.% fiber loading (Fig. 4). The improvement in tensile modulus is only achieved with certain coupling agents at specific loading [27]. A similar trend was observed at 17.5 wt.% fiber loading, where increasing the percentage of compatibilizer from 4 to

8 % showed an increase in the tensile modulus up to 8.7 % and 16.2 % respectively. In relation to the compatibilizer, it can be observed that increasing the composition of MA-HDPE as coupling agent in the composite, a further improvement in stiffness results. Only 12 % compatibilizer showed a reduction in tensile modulus both at 8.5 wt.% and 17.5 wt.% fiber loading. The reduction of tensile modulus at higher

composition of MA-HDPE could be attributed to the changes of molecular morphology of the polymer near the fiber surface or in the bulk of the plastic phase similar to that reported by Sanadi et al. [5].

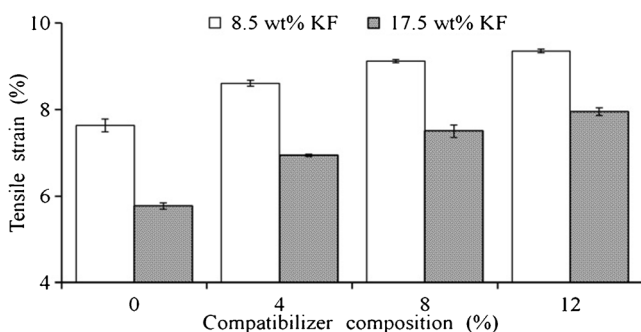
### 3.2.3 Tensile strain

The bar chart of tensile strain of the composites at 8.5 wt.% and 17.5 wt.% kenaf fibers at different percentages of compatibilizer is shown in Fig. 8. Uncompatibilized composites showed a lower tensile strain when compared to the compatibilized composites for both 8.5 wt.% and 17.5 wt.% fiber loadings. Li and Matuana [16] and Liao, et al. [28] reported that without coupling agents, the addition of wood into the PE matrix significantly reduced the elongation at break of the composites due to the incorporation of brittle filler into the polymer matrix. Composites at lower fiber loading of 8.5 wt.% showed a higher tensile strain compared to the composites at higher fiber loading of 17.5 wt.%. Increasing the amount of fiber decreases the amount of polymer available for the elongation [29]. Decrease in failure strain with an increase in fiber content is due to restriction posed by the fiber [20]. However, addition of the compatibilizer helps in stress transfer from matrix to the fiber and is observed from the increase in tensile strain with increase in amount of compatibilizer in the kenaf fiber/HDPE composites. Maleated PEs (MA-HDPE and MA-LLDPE) enhanced the elongation at break of the composites probably because of strong compatibility between the phases [16]. The addition of less stiff components like compatibilizer, shows significant improvement on elongation at break, which increases with increasing loading of the compatibilizer [11].

## 3.3 Flexural properties

### 3.3.1 Flexural strength

Three point bending flexural test measures the force required to bend a material under three point loading conditions. The variation of flexural strength and flexural modulus of



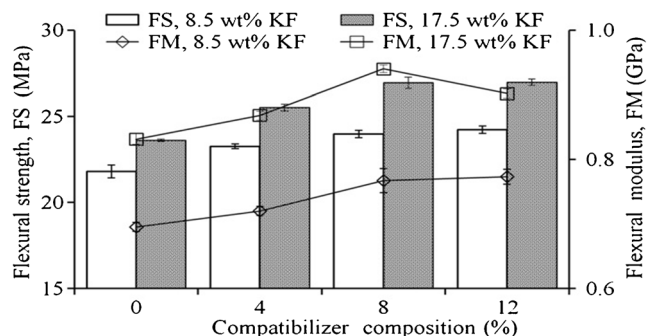
**Fig. 8** The tensile strain of uncompatibilized and compatibilized composites

composites at 8.5 wt.% and 17.5 wt.% kenaf fiber loading with different percentages of compatibilizers is presented in Fig. 9. From the figure, the flexural strength of composites is higher at higher fiber loading of 17.5 wt.% composites relative to the lower fiber loading of 8.5 wt.% composites.

All compatibilized composites showed improvement in flexural strength when compared to the uncompatibilized composites. Better interfacial adhesion between the fiber and matrix could be responsible for this behavior. Karmarkar et al. [30] reported that, increased adhesion between the wood fiber and the matrix provides increased stress transfer from the matrix to the fiber, resulting in an increased stress at failure and higher values of flexural strength in compatibilized system. The flexural strength of 8.5 wt.% and 17.5 wt.% fiber loading increased steadily with the addition of 4 %, 8 % and 12 % compatibilizer into the system. The highest improvement was achieved by addition of 12 % compatibilizer into 8.5 wt.% fiber loading, with the flexural strength value of 24.2 MPa. Addition of 12 % compatibilizer into the composites with 17.5 wt.% fiber loading improved the flexural strength up to 14.2 % when compared to the uncompatibilized system. This proves that the flexural strength is affected by the presence of MA groups attached to HDPE. Previous researchers found that sufficient number of MA groups attached onto PP chains causes strong interfacial interaction, probably due to the formation of chemical bonds between MA groups and hydroxyl groups of bio-flours [9, 31].

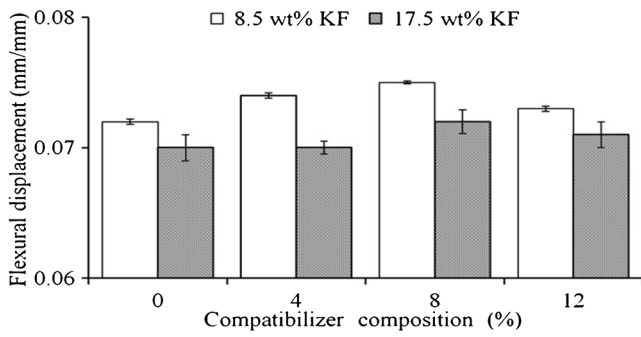
### 3.3.2 Flexural modulus

The flexural modulus of compatibilized composites were higher relative to the uncompatibilized composites at 8.5 wt.% and 17.5 wt.% fiber loadings, as presented in Fig. 9. The improvement in flexural modulus could be due to the improved wetness between the fiber and matrix with the presence of compatibilizer. The flexural modulus of 8.5 wt.% fiber loading increased throughout the range of MA-HDPE content. As the MA-HDPE content increased from 4 % to 8 % loading in composites with 17.5 wt.% fiber loading, a steady



**Fig. 9** The flexural strength and flexural modulus of uncompatibilized and compatibilized composites





**Fig. 10** The flexural displacement of uncompatibilized and compatibilized composites

improvement in flexural modulus was observed. However, incorporation of 12 % MA-HDPE did not contribute to further improvement in flexural modulus. This behavior may be attributed to the migration of excess compatibilizer around the fibers, causing self-entanglement among themselves rather than the polymer matrix, resulting in slippage [12, 20]. The highest improvement at 17.5 wt.% kenaf fiber in flexural modulus was achieved by addition of 8 % compatibilizer with improvement of 13.1 %.

### 3.3.3 Flexural displacement

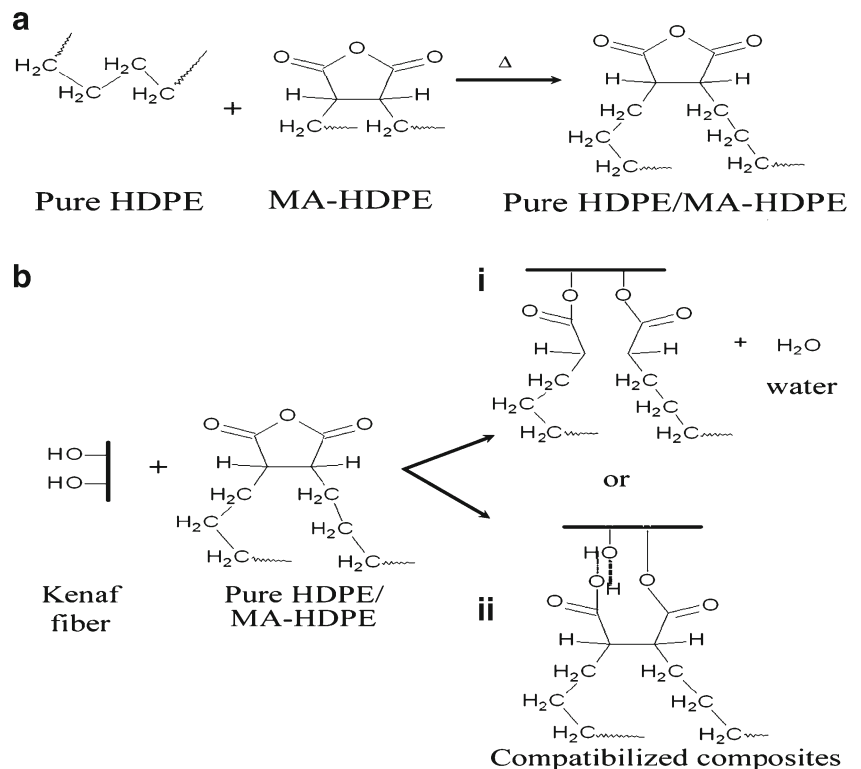
Figure 10 presents the effect of MA-HDPE content on the flexural displacement of the composites. From the figure, the flexural displacement of composite decreased with increasing

fiber content. This indicates that at higher fiber loading, the fiber dominates the fracture process of failure at the interface between the fiber and matrix. The addition of compatibilizer to 8.5 wt.% composites showed an improvement in flexural displacement relative to the uncompatibilized composite. Increase in the compatibilizer content from 4 % to 8 % in the composites containing 8.5 wt.% fiber loading led to increased flexural displacement of composites. Beyond this MA-HDPE content, no appreciable improvement could be seen. The addition of compatibilizer at lower fiber loading seems to help in stress transfer, thereby increasing the flexural displacement. At lower fiber loading the amount of fiber inside the composite is less, so reduced restriction in the mobility of the polymer chain in the composites happens, thereby increasing the flexural displacement of composites. This is quite similar with the tensile strain results as mentioned earlier. At 17.5 wt.% fiber loading, the addition of 4 % compatibilizer did not show any positive effect on the flexural displacement when compared to composite without compatibilizer. However, at 8 % compatibilizer loading, the flexural displacement was at its highest. On the other hand, increasing the compatibilizer content to 12 % in the composite containing higher fiber loading caused a decrease in flexural displacement.

### 3.4 Effect of MA-HDPE treatment (Adhesion mechanism)

The hydroxyl and the other polar groups in the kenaf fiber components act as active sites for water absorption, which

**Fig. 11** Hypothetical adhesion mechanism, (a) interaction between pure HDPE matrix and MA-HDPE, and (b) esterification reactions between kenaf fibers with MA-HDPE, (i) diester structures (ii) half ester and carboxylic structure with intramolecular hydrogen bonding



resulted in incompatibility with the hydrophobic HDPE matrix, leading to reduced properties of the composites. In order to improve the compatibility between the non-polar parts of HDPE and reduce the surface hydrophilicity of kenaf fiber, MA-HDPE was used as compatibilizer in the system. The hypothetical adhesion mechanism is illustrated in Fig. 11.

MA-HDPE improves the interfacial bonding between the kenaf fiber and HDPE matrix by two simultaneous reactions. First, the long molecular chain of MA-HDPE is responsible for the interaction with the non-polar HDPE matrix inside the twin screw extruder created by shear from rotating screws. Secondly, the MA-HDPE groups chemically interact with the functional groups on the kenaf fiber surface. Upon esterification, the polyolefin backbone chains of the MA-HDPE and entangled HDPE were exposed on the surface of fiber, creating a bridge at the interface between the fiber and the matrix. The esterification reaction may result in two products; one is the copolymer with diester structures and another one is the copolymer with half ester and carboxylic structure [10]. Half carboxylic structure will further form intramolecular hydrogen bonding by a reaction between hydrogen atom of a hydroxyl group of kenaf fiber and the MA-HDPE group in the same molecules. Besides that, intermolecular hydrogen bonding also is another possible reaction that will occur. It is believed that exposed polyolefin chains from the compatibilizer diffused into the HDPE matrix phase and entangled with HDPE molecules during processing [32–34]. These interactions provide mechanical integrity to the host material [5]. In the case of natural fiber reinforced polyolefin composites, the chemical interaction has been attributed to the formation of covalent bonds between hydroxyl groups on the fiber surface and the anhydride groups of the coupling agent via an esterification process [16, 35]. Therefore, the compatibility between kenaf fiber and HDPE matrix can be improved, which in turn enhances the wettability within the matrix, leading to improved mechanical and interfacial behavior of composites.

#### 4. Conclusion

The effects of MA-HDPE compatibilizer loading on the mechanical and interfacial properties of kenaf fiber reinforced HDPE composites were investigated in this study. In the absence of a compatibilizer, kenaf fiber reinforced HDPE composites showed poor mechanical and interfacial properties due to poor interfacial bonding between the fiber and the polymer matrix. The addition of MA-HDPE as compatibilizer improved the tensile and flexural properties of kenaf fiber reinforced HDPE composites. These are clearly revealed through a fractured surface characterization that showed that the compatibilized composites have a better dispersion on surface structure and improved interfacial adhesion. 8 % MA-HDPE loading provided maximum enhancement in all

mechanical properties listed above and better adhesion between kenaf fiber and the matrix when compared to the other compatibilizer contents. Presumably, there is a good compatibility between HDPE matrix and kenaf fiber due to the strong interfacial bonding resulting from the chemical reaction between maleic anhydride on the compatibilizer and hydroxyl groups of kenaf fiber. FTIR analysis further supported the improvement in mechanical properties and interfacial properties of the composites. The FTIR spectroscopy indicated that the maleic anhydride was bonded to the kenaf fiber through esterification reaction by observation of stretching vibration of the ester carbonyl groups (C=O) around  $1725\text{ cm}^{-1}$  to  $1742\text{ cm}^{-1}$  in the compatibilized composites and hydroxyl group at  $3327\text{ cm}^{-1}$ . These results indicate that the mechanical performance and interfacial properties of the kenaf fiber reinforced HDPE improved with the addition of MA-HDPE composites due to the enhanced interfacial adhesion between kenaf fiber and HDPE matrix.

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