Effect of Lysine Treatment on the Properties of Linear Low-Density Polyethylene/Poly (Vinyl Alcohol)/Kenaf Composites

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Kenaf (KNF) was treated at room temperature with the amino acid lysine. The surface treatment of KNF using lysine was studied for the first time on prepared composites with linear low-density polyethylene, poly(vinyl alcohol), and kenaf. The LLDPE/PVOH/KNF composites with different loadings of lysine-treated and untreated KNF were studied at levels of 10, 20, and 40 parts per hundred resin (phr). The melt mixing of all the composites were prepared by using an internal mixer for 10 min at a temperature and rotor speed of 150 °C and 50 rpm, respectively. The results showed that tensile strength and tensile modulus were improved for the lysine-treated KNF composites compared to the untreated KNF composites. Fourier transform infrared (FTIR) spectroscopy revealed the presence of protonated amino (NH₃⁺) and carboxylate (COO⁻) groups in the LLDPE/PVOH/KNF composites after the lysine treatment. Scanning electron microscopy analysis showed good adhesion between the lysine treated KNF and the LLDPE/PVOH matrices. Thermogravimetric analysis (TGA) showed that the lysine treated KNF composites possessed a higher thermal stability than the untreated KNF composites.

Keywords: Lysine; Amino acid; Kenaf composites; Tensile properties; Chemical treatment

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INTRODUCTION

In the past decade, the use of natural fibers as a potential substitute for synthetic fibers such as glass and aramid fibers used in the development of composites materials has received considerable interest (Morales-Cepeda *et al.* 2015; Pai and Jagtap 2015; Pang *et al.* 2015). Natural fibers are advantageous to synthetic fibers because they are biodegradable, inexpensive, non-toxic, and lightweight (El-Shekeil *et al.* 2014; Islam *et al.* 2015; Zamri *et al.* 2016). Natural fibers such as kenaf (KNF), jute, sisal, hemp, coir, bamboo, wood, and cotton fibers are commonly used in natural fiber-polymer composites (NFPCs) (Morales-Cepeda *et al.* 2015; Väisänen *et al.* 2017). Among natural fibers, KNF is advantageous due to its low cost, short plantation cycle, ability to grow under various climates, and minimal need for pesticides and herbicides (Kamal *et al.* 2014; Saba *et al.* 2015; Ramesh 2016). Nevertheless, the incompatibility between hydrophilic KNF and hydrophobic thermoplastic matrices (polyethylenes (PEs) and polypropylene (PP)) generally results in poor fiber-matrix interfacial adhesion, which leads to a decrease in the mechanical properties of the composites (Pang and Ismail 2014; Pang *et al.* 2016). The mechanical properties of the composites depend strongly on the fiber-matrix

interfacial adhesion, as the applied stress is transferred between the matrix and the fibers across the interface (Pickering *et al.* 2016). Therefore, a good fiber-matrix interfacial adhesion is very important to achieve optimum reinforcement (George *et al.* 2015; Pickering *et al.* 2016).

The surface modification of fibers by chemical treatment is a widely used approach to improve the interfacial interaction and properties of NFPCs (Asumani et al. 2012; Ramesh, 2016; Väisänen et al. 2017). Chemical treatments such as silane treatment, alkali treatment, acetylation, isocyanate treatment, the use of maleated coupling agents, and graft copolymerization are the common approaches used to modify KNF (Datta and Kopczyńska 2015; Akhtar et al. 2016; Pang et al. 2016). For instance, Datta and Kopczyńska (2015) investigated the effect of acetylation, blocked isocyanate, maleic anhydride, and potassium permanganate on the properties of KNF/thermoplastic polyurethane composites. They noted improved tensile properties, hardness, and water resistance, particularly for the treated composites with low fiber loading. Compared to the other chemical treatments, it was found that the composites with the potassium permanganate treatment showed better interfacial adhesion between the KNF and the thermoplastic polyurethane matrix. In another study, Akhtar et al. (2016) found that alkaline-treated KNF/PP composites showed higher tensile and flexural properties in comparison to untreated KNF/PP composites. Meanwhile, chemical treatment by means of amino acids has also become an alternative to enhance the mechanical properties of natural fiber polymer composites. The use of amino acid (lysine) has gained interest for its high biocompatibility, non-toxic, and widespread availability (Liuyun et al. 2016; Villegas et al. 2017). For instance, Alam et al. (2015) and Liuyun et al. (2016) successfully enhanced the mechanical properties of flax fiber composites and g-nanohydroxyapatite/poly(lactic acid-co-glycolic acid) nano composites with the assistance of lysine. To the best of our knowledge, there has been no research on the utilization of lysine amino acid in modifying KNF-based LLDPE/PVOH composites.

The study aimed to investigate the effect of lysine treatment on the mechanical and thermal properties of LLDPE/PVOH/KNF composites. The morphological aspects and structural information of the LLDPE/PVOH/KNF composites were also evaluated.

EXPERIMENTAL

Materials

The KNF was provided by the National Kenaf and Tobacco Board (Kelantan, Malaysia). The lysine was purchased from Sigma-Aldrich (M) Sdn. Bhd. (Petaling Jaya, Malaysia). Linear low-density polyethylene (LLDPE) with a density of 0.92 g/cm³ was supplied by Lotte Chemical Titan Nusantara (Jakarta, Indonesia). Polyvinyl alcohol (PVOH) with a density of 1.269 g/cm³ and a molecular weight of 89,000 g/mol to 98,000 g/mol was purchased from Sigma-Aldrich (M) Sdn. Bhd (Petaling Jaya, Malaysia). A blend of LLDPE and PVOH were used as the polymer matrices at a composition ratio of 60:40. This was done to provide good processability and tensile properties, as reported by Ismail *et al.* (2009).

Treatment of the KNF

The 3% lysine solution was prepared by dissolving the lysine powder in deionized (DI) water. The KNF was immersed in the prepared lysine solution for 24 h at room

temperature. After 24 h, the KNF was filtered, washed thoroughly using DI water, and dried at room temperature.

Preparation of the Composites

The LLDPE/PVOH/KNF composites with the treated and untreated KNF were prepared using the melt-mixing method followed by compression molding. The melt-mixing process was carried out at 150 °C and 50 rpm for 10 min. The compression molding process was carried out using an electrical hydraulic hot press at 150 °C for 11 min.

Material Characterization

The composites were characterized relative to their tensile properties, morphology, spectroscopic analysis, thermal stability, and water absorption. The tensile tests were carried out in accordance with ASTM D638-14 (2014) using an Instrom 3366 Universal Testing Machine (Norwood, MA, USA). The test was performed at a crosshead speed of 5 mm/min with a fixed gauge length of 50 mm. Five specimens were tested for each composite, and the average values for the tensile strength, tensile modulus, and elongation at break were reported. The analysis of the tensile fractured surface morphology of the composites was carried out using a scanning electron microscope (SEM) (Supra-35VP; ZEISS, Oberkochen, Germany). The spectroscopic analysis of the composites was completed using a Fourier-transform infrared (FTIR) spectrometer (2000 Explorer; PerkinElmer, Waltham, USA). Each spectrum was scanned for 16 consecutive scans within the wavenumber range of 4000 cm⁻¹ to 550 cm⁻¹ at a resolution of 4 cm⁻¹. The composite samples of 10 mg \pm 2 mg were used for the characterization using a thermogravimetry analyzer (Pyris 6; PerkinElmer, Waltham, USA) in a temperature scan range of 30 °C to 600 °C under a nitrogen atmosphere at a heating rate of 20 °C/min.

RESULTS AND DISCUSSION

Processing Torque

Figure 1 shows the processing torques of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings. There are three major peaks in Fig. 1, which indicate an increased melt viscosity after the sequential addition of each material into the chamber in the order of LLDPE, PVOH, and then KNF (untreated and lysine-treated). This corresponds to the high shear action generated as the materials provide resistance to the flow in the mixing chamber (Pang *et al.* 2016). Meanwhile, the gradual decrement in the torques and the subsequently stabilized at the end of the mixing time (approximately 560 s to 600 s) were attributed to the melting and homogeneous melt mixture obtained.

The stabilization torques of the LLDPE/PVOH/KNF composites with the lysinetreated and untreated KNF at different KNF loadings are displayed in Fig. 2. The composites with 40 parts-per-hundred resin (phr) KNF (untreated and lysine-treated) exhibited higher stabilization torques (17.8 N/m and 17.9 N/m) than the composites with 10 phr KNF (untreated and lysine-treated), which had stabilization torques of 17.3 N/m and 17.4 N/m, respectively. The results revealed that the incorporation of KNF increased the viscosity of the polymer melt, particularly at higher KNF loading. However, at similar KNF loading, the composites with the lysine-treated KNF showed higher stabilization torques in comparison to the composites with the untreated KNF. This is attributed to the enhancement in the interfacial adhesion between the lysine-treated KNF and the LLDPE/PVOH matrices, which subsequently restricted the flowability of the composites.



Fig. 1. The processing torques of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings



Fig. 2. The stabilization torques of the LLDPE/PVOH/KNF composites with lysine-treated and untreated KNF at different KNF loadings

Tensile Properties

Figure 3 displays the tensile strength of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings. The tensile strength decreased for all the composites as the KNF loading increased up to 40 phr. The results

show that addition of KNF into the LLDPE/PVOH composites decreased the ability of the polymer matrices to transfer the applied stress, especially at higher KNF loading (*i.e.*, 40 phr). This is attributed to the poor wetting of KNF with the polymer matrices of the composites at the higher KNF loading. However, the tensile strength of the composites with 10 phr lysine-treated KNF (9.18 MPa) was similar to the composite with 0 phr KNF (9.26 MPa). This suggests that the addition of 10 phr lysine-treated KNF may reinforce the LLDPE/PVOH composites due to the good interfacial adhesion between them. Moreover, all the composites with the lysine-treated KNF showed higher tensile strength than composites with the untreated KNF (at similar loading). This is credited to the improvement in the adhesion of the lysine-treated KNF and LLDPE/PVOH matrices. This subsequently resulted in a better stress transfer than the untreated KNF. A similar observation was reported by Pang and Ismail (2014) and Datta and Kopczyńska (2015), who found that the tensile strength of composites increased because of the better filler-matrix adhesion resulting from filler treatment.



Fig. 3. The tensile strength of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings

The tensile modulus of the LLDPE/PVOH/KNF composites with the lysinetreated and untreated KNF at different KNF loadings is presented in Fig. 4. The tensile modulus of all the composites increased as the KNF loading increased up to 40 phr. This was due to an increase in the stiffness of the composites, particularly at higher filler loading (Pang *et al.* 2015). However, at similar KNF loading, the tensile modulus of the composites with the lysine-treated KNF was higher than that of the composites with the untreated KNF. For instance, at 10 phr KNF loading, the tensile moduli were 796.5 MPa and 914.6 MPa for the composites with the untreated KFN and the lysine-treated KNF, respectively. This shows that the lysine treatment improved the stiffness of the KNF and enhanced the interfacial adhesion between the KNF and the LLDPE/PVOH matrices.



Fig. 4. The tensile modulus of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings

Figure 5 illustrates the elongation at break of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at different KNF loadings. As seen in Fig. 5, all the composites exhibited a decreasing trend in the elongation at break as KNF loading increased. This is due to the lower deformability of the LLDPE/PVOH matrices at the high KNF loading. Furthermore, the composites with the lysine-treated KNF showed a lower elongation at break compared to the untreated KNF composites with a similar KNF loading. This is correlated to the higher stiffness obtained earlier, due to the enhanced adhesion between the lysine-treated KNF and LLDPE/PVOH matrices.



Fig. 5. The elongation at break of the LLDPE/PVOH/KNF composites with the lysine-treated and untreated KNF at the different KNF loadings

FTIR Analysis

Figure 6 shows the FTIR spectra of the LLDPE/PVOH/KNF composites at the 40 phr lysine-treated KNF and untreated KNF loadings. The differences observed in Fig. 6a and 6b are at the characteristic peaks of 3400 cm⁻¹ to 3200 cm⁻¹ (O-H stretching), 1709 cm⁻¹ (C=O stretching), 1645 cm⁻¹ (O-H stretching of absorbed water), 1559 cm⁻¹ (NH₂ stretching), 1596 cm⁻¹ (carboxylate (COO⁻) asymmetric stretching and NH₃⁺ asymmetric deformation), and 1100 cm⁻¹ to 900 cm⁻¹ (C-O and C-O-C stretching) (Kitadai *et al.* 2009; Liuyun *et al.* 2016; Pang *et al.* 2018). The peaks corresponding to O-H stretching (3400 cm⁻¹ to 3200 cm⁻¹) in the composites with the untreated KNF (Fig. 6a) became broadened in the composites with the lysine-treated KNF (Fig. 6b). This suggests that the reaction between lysine and hydroxyl group of KNF had occurred, thereby leading to small shifts in wavenumbers of O-H stretching peaks. Moreover, the peak at 1709 cm⁻¹ (C=O stretching of COOH) became apparent in the composites with the lysine-treated KNF, as illustrated in Fig. 6b. This shows that the lysine was successfully adsorbed on the KNF surface during the treatment.

Furthermore, a peak at 1559 cm⁻¹ was observed in the lysine-treated composites (Fig. 6b), corresponded to the NH₂ group of lysine (Kitadai *et al.* 2009). Another peak at 1596 cm⁻¹, which was assigned to the carboxylate (COO⁻) asymmetric stretching and the NH₃⁺ asymmetric deformation, intensified in the lysine-treated KNF composites (Kitadai *et al.* 2009; Liuyun *et al.* 2016). This finding implies the presence of lysine in LLDPE/PVOH/KNF composites. Meanwhile, the existence of the peak at 1645 cm⁻¹ in the lysine-treated KNF composites suggested that the lysine treatment had a minimal influence on the removal of water. As seen in Fig. 6b, the peaks in the region of 1160 cm⁻¹ to 900 cm⁻¹ were more apparent compared to the peaks in Fig. 6a. This may be due to the additional contribution of the C-O stretching from the lysine structure.



Fig. 6. The FTIR spectra of the LLDPE/PVOH/KNF composites with (a) 40 phr untreated and (b) 40 phr lysine-treated KNF

Morphological Study

Figure 7 illustrates the SEM micrographs of the LLDPE/PVOH/KNF composite tensile fractured surfaces with the lysine-treated and untreated KNF at 10, 20, and 40 phr KNF loadings.



Fig. 7. The SEM micrographs of the LLDPE/PVOH/KNF composite tensile fractured surfaces with (a) 10 phr untreated KNF; (b) 10 phr lysine-treated KNF; (c) 20 phr untreated KNF; (d) 20 phr lysine-treated KNF; (e) 40 phr untreated KNF; and (f) 40 phr lysine-treated KNF at magnification of 200×

The SEM micrographs revealed that the detachment of the KNF from the matrices was more severe for the composites with the untreated KNF (Figs. 7a, c, and e). This indicates poor interfacial adhesion between the untreated KNF and the LLDPE/PVOH matrices. Meanwhile, at similar KNF loading, the lysine-treated KNF composites exhibited better fiber-matrix interfacial adhesion in comparison to the untreated KNF composites. As can be seen in Figs. 7b, d, and f, less detachment was observed and the lysine-treated KNF was embedded within the LLDPE/PVOH matrices. The result is in agreement with the higher tensile strength obtained in the lysine-treated KNF composites.

Thermogravimetric Analysis (TGA)

The representative TG-derivative thermogravimetry (DTG) thermograms of the LLDPE/PVOH/KNF lysine-treated and untreated KNF composites at 10 phr and 40 phr KNF loadings are shown in Figs. 8 and 9, respectively. Based on Figs. 8 and 9, both the lysine-treated and untreated KNF composites underwent three thermal degradation stages which occurred in the temperature range of 100 °C to 200 °C (evaporation of the surface absorbed water), 250 °C to 400 °C (decomposition of the PVOH and KNF), and 450 °C to 550 °C (decomposition of the LLDPE) (Ismail *et al.* 2009; Pang *et al.* 2018; Taghizadeh and Sabouri 2013). The parameters extracted from the TG-DTG thermograms, including the maximum degradation temperatures (T_{maxI} and T_{maxII}) and the percentage of char residue at 590 °C, are listed in Table 1.

From Table 1, the T_{maxI} and T_{maxII} of the composites with 10 phr untreated KNF were 348 °C and 351 °C, respectively. However, the T_{maxI} and T_{maxII} were shifted to higher temperatures (355 °C and 476 °C) in the composites with 10 phr lysine-treated KNF. Likewise, the composites with 40 phr lysine-treated KNF also exhibited higher T_{maxI} and T_{maxII} than the composites with 40 phr untreated KNF, as can be seen in Table 1.

The increment in the T_{maxI} and T_{maxII} revealed that the thermal stability of the composites was enhanced with the use of the lysine-treated KNF in the LLDPE/PVOH matrices. Additionally, the char residue of the composites with 10 phr and 40 phr lysine-treated KNF were 2.60% and 8.69%, while it was 1.31% and 8.56% in the composites with 10 phr and 40 phr untreated KNF, respectively. The char formation acts as a thermal protective layer, which inhibited the emission of the gaseous degradation products during the heating process (Dikobe and Luyt 2010; Pang *et al.* 2018). The higher char residue percentage observed in the composites with the lysine-treated KNF indicated a higher thermal stability.



Fig. 8. The TG thermograms of the LLDPE/PVOH/KNF composites with the untreated and lysinetreated KNF at 10 phr and 40 phr KNF loadings



Fig. 9. DTG thermograms of the LLDPE/PVOH/KNF composites with untreated and lysinetreated KNF at 10 phr and 40 phr KNF loadings

Table 1. TGA Parameters of the LLDPE/PVOH/KNF Composites with the

 Untreated and Lysine-treated KNF at 10 phr and 40 phr KNF Loadings

| LLDPE/PVOH/KNF Composites | T _{maxl} (°C) | T _{maxII} (°C) | Char Residue at 590 °C (%) |
|------------------------------|---------------------------|----------------------------|-------------------------------|
| 10 phr Untreated KNF | 348 | 471 | 1.31 |
| 10 phr Lysine-treated KNF | 355 | 476 | 2.60 |
| 40 phr Untreated KNF | 351 | 480 | 8.56 |
| 40 phr Lysine-treated KNF | 364 | 487 | 8.69 |

CONCLUSIONS

- 1. The stabilization torque of the LLDPE/PVOH/KNF composites increased with the use of lysine-treated KNF.
- 2. The lysine-treated KNF composites exhibited a noticeable enhancement in the tensile strength and tensile modulus.
- 3. The FTIR analysis confirmed the presence of protonated amino (NH_3^+) and carboxylate (COO⁻) groups in the LLDPE/PVOH/KNF composites after the lysine treatment.
- 4. Good interfacial adhesion between the lysine-treated KNF and the LLDPE/PVOH matrices was revealed by the SEM micrographs.
- 5. The lysine-treated KNF composites exhibited a higher thermal stability than the untreated KNF composites.

ACKNOWLEDGMENTS

The authors would like to extend their appreciation to the Universiti Sains Malaysia, the National Kenaf and Tobacco (Kelantan, Malaysia), and the Malaysian Higher Education (RUC: 1001/ PKT/8640014), for providing materials and funding in this study.

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Article submitted: April 22, 2019; Peer review completed: August 4, 2019; Revised version received: December 16, 2019; Accepted: December 17, 2019; Published: January 29, 2020.

DOI: 10.15376/biores.15.1.1915-1926