

Communication

Influence of Surface Treatment on Tensile Properties of Low-Density Polyethylene/Cellulose Woven Biocomposites: A Preliminary Study

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Abstract: Cellulose woven (CW) was surface treated by means of hexadecyltrimethylammonium bromide surfactant (HTAB) in aqueous solution medium at elevated temperature. The parameters of the surface treatment that have been studied are HTAB concentration (0.2, 0.4, 0.6, 0.8 and 1.0 wt%) and treatment time (1, 2, 3, 4 and 5 h). The untreated and treated CW filled low-density polyethylene (LDPE) biocomposites were prepared via compression molding technique. The tensile testing results of LDPE/CW biocomposites demonstrated that the optimum HTAB concentration for treatment of CW in 1 h was 0.4 wt%, while the optimum treatment time at 0.4 wt% HTAB was 2 h. The SEM (scanning electron microscope) images indicated that there is no significant difference in the morphology of the untreated and treated CW; however the morphology of the LDPE/treated CW biocomposite showed better interfacial adhesion as compared with the untreated ones. The FTIR (Fourier transform infrared spectroscopy) spectra revealed that the presence of HTAB on the surface of treated CW and also revealed the existence of intermolecular interactions between LDPE and treated CW. In summary, HTAB could potentially be used as a treatment agent for modifying the surface of CW and consequently improved the tensile properties of LDPE/CW biocomposites.

Keywords: surface treatment; surfactant; low-density polyethylene; cellulose woven; biocomposites

1. Introduction

The use of polysaccharide-like cellulose as natural filler in various polymer composite systems has been widely investigated by many researchers [1–4]. This is due to the fact that the cellulose is one of the most abundant polymers on the earth, which is also low-cost, non-toxic, renewable, environmentally friendly and biodegradable [5,6]. Cellulose from cotton linters in the form of woven was used in this preliminary study. The utilization of cellulose woven as filler in polymer biocomposites is very promising due to its strength, stiffness and the structural stability of the products. Besides that, the preparation of polyethylene (PE)/cellulose woven biocomposites may also possibly be used in the production of diverse products with biocompatible and biodegradable properties. Nevertheless, the compatibility and adhesion between the natural fillers and the PE in the biocomposite systems is very low [7,8], thus resulting in reductions in their performance, particularly tensile properties and other correlated characteristics of the final natural filler-reinforced composite products.

This is because the polar hydrophilic nature of cellulose caused compatibility problems with the non-polar hydrophobic character of PE, which led to poor physicochemical properties of the biocomposites [9,10]. Therefore, surface treatment was usually applied to resolve this problem. The treatment by using chemicals could modify the natural filler and improve the interfacial adhesion between filler and matrix. Chemical treatments by using alkali and silane were carried out in the previous study to roughen the surface of natural filler and contributed to the improvement of the surface interaction with polymer matrix [11]. On the other hand, another way to improve the compatibility between two polymers that have different polarity and hydrophobicity is by using surface active agent (surfactant) in the surface treatment of natural fillers. The surfactants do not leave any undesired by-product or residue that might be harmful to the human and environment after treatment as compared to acid and alkali.

Surfactants are organic salts that have amphiphilic character, which contain both non-polar hydrophobic and polar hydrophilic groups [12]. Moreover, they can act as interaction links between non-polar hydrophobic and polar hydrophilic polymers [13]. Previous studies have indicated that the anionic and cationic surfactants could successfully be utilized as compatibilizing agents in polymer blend systems especially for polar and non-polar polymers [14,15]. In addition, natural fillers such as starch, sawdust and cellulose powders could also be treated with ionic and non-ionic surfactants for improving the properties of polymer composites [16–18]. Therefore, the surfactants could definitely increase compatibility between the components phases of the biocomposites. In this preliminary study, the cationic surfactant was used as a treatment agent for natural filler to enhance the physicochemical properties of PE/cellulose woven biocomposites system. The tensile, morphological and chemical properties of prepared biocomposites have been characterized and studied to determine the optimum surfactant concentration for treatment of cellulose woven and also the optimum treatment time.

2. Experimental Section

2.1. Materials

The polymer matrix used is a low-density polyethylene, LDPE (coating grade), acquired from Lotte Chemical Titan (M) Sdn. Bhd (Kuala Lumpur, Malaysia). The cationic surfactant used is a hexadecyltrimethylammonium bromide, abbreviated as HTAB (purity 99%) and procured from Sigma Aldrich (St. Louis, MO, USA). The filler of biocomposites is cellulose woven (CW) which was weaved plain, obtained from flat cloth diapers that consists of 100% cotton. The chemicals were used as received without further purification.

2.2. Surface Treatment of CW

HTAB was mixed with 200 mL of distilled water in a beaker and stirred rigorously by using a magnetic stirrer hot-plate apparatus until a transparent solution was obtained. Then, the HTAB solution was heated at temperature of 80 °C. The CW was surface treated by immersing two layers of CW ($150 \times 150 \text{ mm}^2$) into the HTAB solution for one hour with stirring at 350 rpm. The treated CW was dried in an oven at 70 °C for overnight. The HTAB concentration in aqueous solution was varied from 0.2 to 1.0 wt%. The optimum HTAB concentration was determined through tensile testing of the LDPE/CW biocomposites. The immersion time was also varied from 1 to 5 h to identify the optimum treatment time.

2.3. Preparation of LDPE/CW Biocomposites

The biocomposites were prepared through compression molding technique by using a hydraulic hot press machine. The LDPE was sandwiched between two layers of treated CW in the mold with dimension of $150 \times 150 \times 1 \text{ mm}^3$. The molding procedures involved preheating at 150 °C for 7 min, compression at 150 °C for 2 min and subsequent cooling at 20 °C for 5 min [13]. The resultant biocomposites were conditioned in an oven at 70 °C for at least up to 24 h prior to characterizations. The biocomposite containing only LDPE and untreated CW has also been prepared for comparison.

2.4. Characterization of LDPE/CW Biocomposites

The tensile extension, tensile strain, tensile stress and modulus properties were measured according to the ASTM D638 [19] at room temperature by using an Instron universal testing machine (model 5567, Norwood, MA, USA) equipped with a 30 kN load cell. The initial gauge length and the crosshead speed were fixed to 40 mm and $5 \text{ mm} \cdot \text{min}^{-1}$, respectively. The biocomposite samples of 1 mm thickness were cut into a dumbbell shape (Type V) by using a die cutter. Ten samples were used for each HTAB concentration and treatment time and the average data along with corresponding standard deviation were calculated.

The morphology of untreated and treated CW and also their biocomposites was visualized by using Hitachi (model S-3400N, Tokyo, Japan) scanning electron microscope (SEM). The samples for examination were obtained from tensile test samples that have been cross-sectioned at room temperature. The fractured surfaces of the samples were sputter-coated with a thin layer of gold to

avoid electrostatic charge during examination. The morphologies were observed at magnification of 50× and 3000× with accelerating voltages of 5.0 kV.

The functional groups of the untreated and treated CW as well as the biocomposites were determined by using Perkin Elmer Spectrum 100 Series FTIR (Fourier transform infrared spectroscopy) (Waltham, MA, USA). The spectra were obtained by employing a universal attenuated total reflectance (UATR) equipped with a ZnSe-diamond composite crystal accessory. Each sample was scanned 16 times, in the wavenumber ranged from 4000 to 500 cm^{-1} and resolution of 4 cm^{-1} at room temperature [20].

3. Results and Discussion

3.1. Tensile Testing

Figure 1a,b shows the tensile extension and tensile strain at break of LDPE/CW biocomposites with different HTAB concentrations in aqueous solution. From the results, it was discovered that the extension and strain properties of LDPE/untreated CW biocomposite (0 wt% HTAB) are lower than LDPE/treated CW biocomposites (0.2 to 0.6 wt% HTAB). This is due to the poor interfacial adhesion between polar hydrophilic untreated natural fiber woven and non-polar hydrophobic synthetic polymer [21]. On the other hand, an increase of the extension and strain properties of LDPE/treated CW biocomposites are attributed to the ability of HTAB which can interact with both polar hydrophilic CW and non-polar hydrophobic LDPE since it possesses amphiphilic character [15]. Additionally, the extension and strain properties of LDPE/treated CW biocomposites are drastically increased as the HTAB concentration slightly increased. However, the highest extension and strain properties of biocomposites were found at concentration of 0.4 wt% HTAB in one hour of treatment. Unfortunately, CW treated with 0.6 wt% HTAB has not significantly increased the extension and strain properties of the biocomposite compared to the previous ones. It can be seen that a decreasing trend was also showed in LDPE/treated CW biocomposites (0.8 and 1.0 wt% HTAB).

Figure 1c,d demonstrates the tensile stress and modulus at break of LDPE/CW biocomposites with different HTAB concentrations in aqueous solution. It was discovered that the tensile stress of LDPE/untreated CW biocomposite (0 wt%) is also lower than LDPE/treated CW biocomposites (0.2 to 0.6 wt% HTAB). However, the tensile stress decreased as the HTAB concentration increased from 0.8 to 1.0 wt%. This is due to high concentration of the surfactant which has caused the softening effect on strength of LDPE/CW biocomposites [22]. Therefore, based on this result 0.4 wt% HTAB was considered as the optimum concentration for treatment of CW in one hour. In addition, it can be seen that the tensile stress of the biocomposites displayed a similar behavior as the tensile extension and tensile strain. In most cases, when the extension and strain properties increased, the tensile stress would radically decrease [23], this is because decrease of stiffness of the samples which caused by increase of pliability. Nevertheless, the obtained result showed an opposite trend, this is probably due to the treated cellulose filler was in the form of woven which could provide the tautness to the biocomposites. On the other hand, the modulus of LDPE/treated CW biocomposites is lower than LDPE/untreated CW biocomposite. This is because of the improvement of extension and strain properties of the biocomposites which could dramatically deteriorate their rigidity character.

Figure 1. Influences of hexadecyltrimethylammonium bromide surfactant (HTAB) concentration on (a) tensile extension; (b) tensile strain; (c) tensile stress; and (d) modulus at break of low-density polyethylene (LDPE)/cellulose woven (CW) biocomposites.

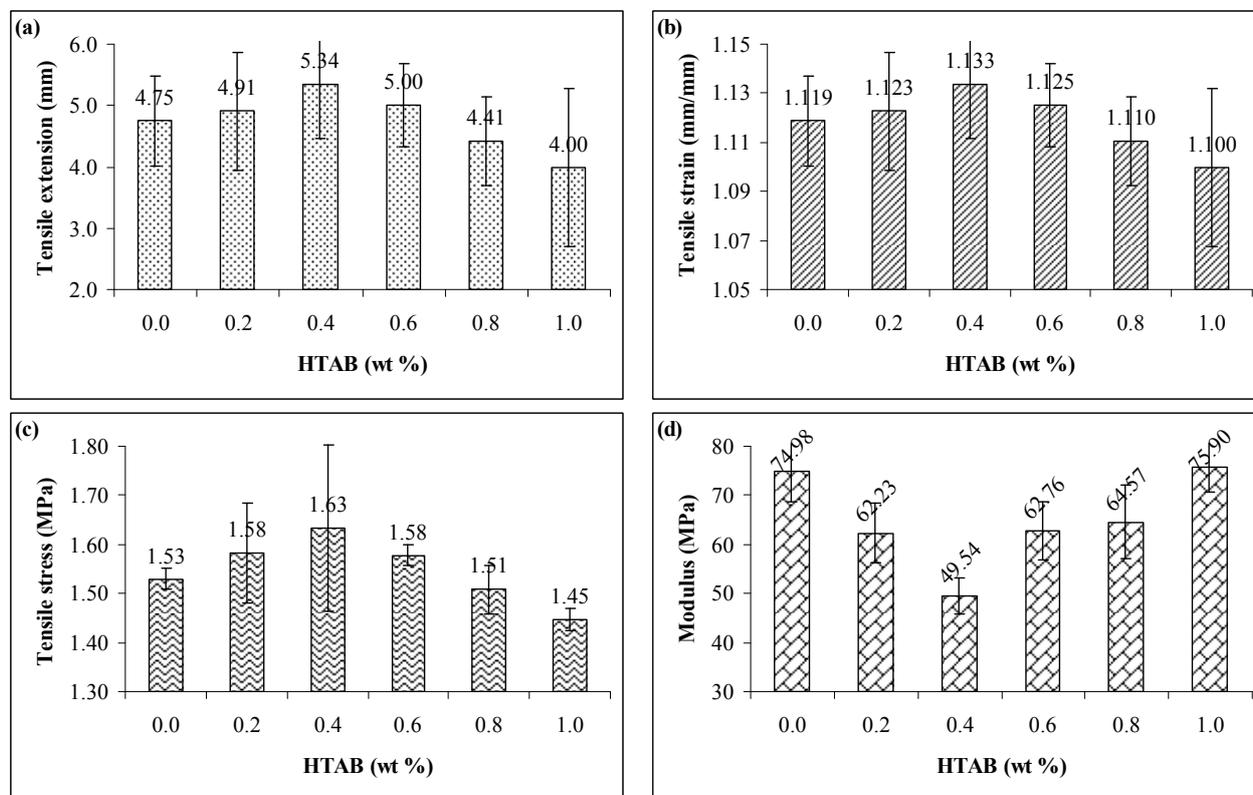
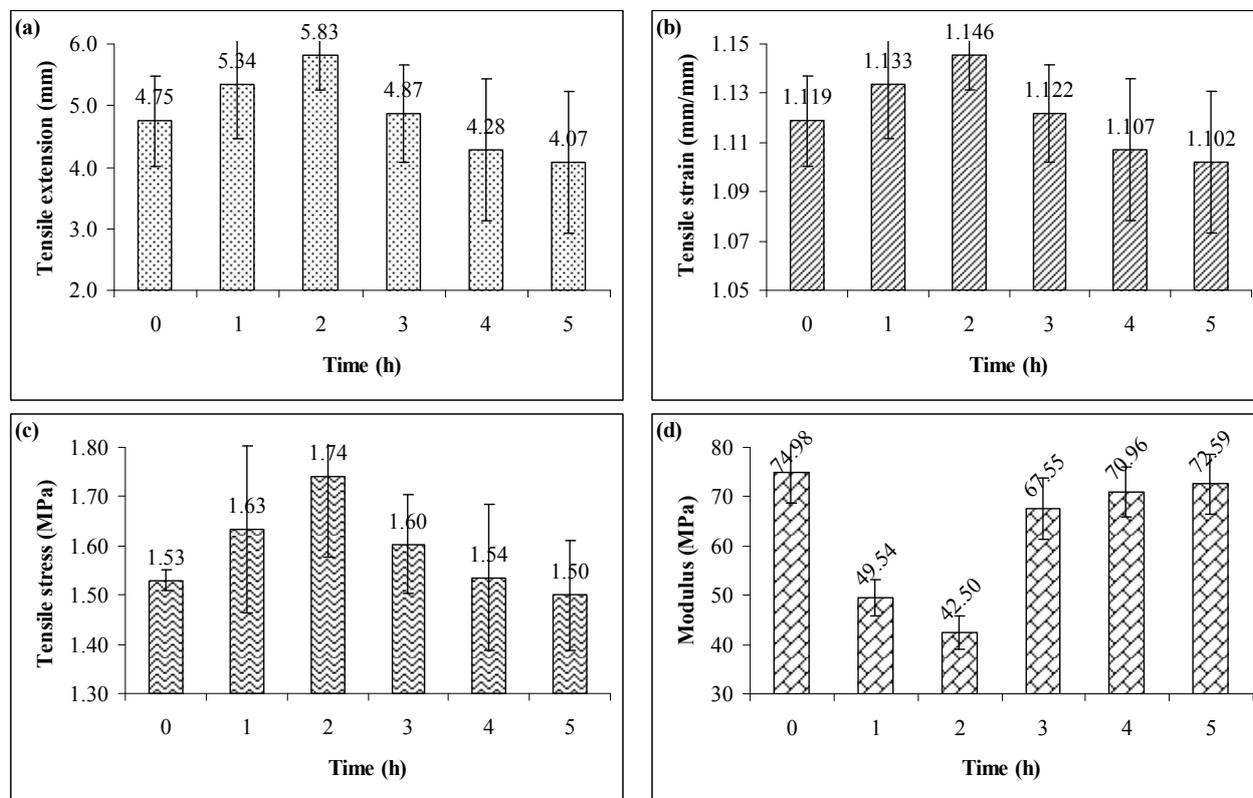


Figure 2a,b indicates the tensile extension and tensile strain at break of LDPE/CW biocomposites with different treatment times. From these results, it is clearly seen that the prolonged treatment time for another hour significantly improved the extension and strain properties of LDPE/treated CW biocomposites. This is possibly because more time has been given for the HTAB to interact with polar hydrophilic CW. This has increased the interfacial adhesion between CW and LDPE. Nevertheless, 3 to 5 h of treatment CW exhibited the decrease in extension and strain properties. Therefore, it was found that after the CW was treated with 0.4 wt% HTAB for more than 2 h, their extension and strain properties are inversely proportional with an increase in the treatment time. This is because of longer duration of treatment which has weakened the physicochemical properties of CW especially at elevated temperature. On top of that, Figure 2c,d exhibits the tensile stress and modulus at break of the LDPE/CW biocomposites with different treatment times. Based on these results, it can be seen that the optimum treatment time at 0.4 wt% HTAB was 2 h, since it gives the highest tensile stress in comparison with other treatment times. Moreover, it was discovered that the modulus of LDPE/treated CW biocomposites has an identical pattern as the modulus result shown in Figure 1d; therefore the reason for this phenomenon was also same as above. Furthermore, the SEM examination has been performed to confirm the difference in the morphology between LDPE/untreated CW and LDPE/treated CW biocomposites.

Figure 2. Influences of treatment time on (a) tensile extension; (b) tensile strain; (c) tensile stress; and (d) modulus at break of LDPE/CW biocomposites.



3.2. Morphology Examination

Figure 3 demonstrates SEM micrographs of the surfaces of untreated and treated (0.4 wt% HTAB, 2 h) CW samples. The SEM images at magnification of 50 \times for the untreated and treated CW samples (Figure 3a,b) indicated that there are no significant differences between both samples. This observation also suggested that the treated CW remains intact after treatment in hot aqueous solution of 0.4 wt% HTAB for a couple of hours. Moreover, the changes of phase morphology between the untreated and treated CW samples also cannot clearly be seen even at a magnification of 3000 \times (Figure 3c,d). Additionally, there are no erosions perceived on the surface of treated CW. This is most likely caused by the nature of HTAB solution which was not able to erode the surface of CW as acid or alkaline solution [24]. From these results, it was found that the physical properties of treated CW were not affected by surface treatment of HTAB.

Figure 4 displays SEM micrographs of the fracture surfaces (cross-section) of LDPE/untreated CW and LDPE/treated CW (0.4 wt% HTAB, 2 h) biocomposite samples. The SEM images indicated that the LDPE/untreated CW biocomposite sample (Figure 4a) at magnification of 50 \times has many fibers pulled-out compared to LDPE/treated CW biocomposite sample (Figure 4b). This kind of pulled-out is obtained due to the poor interfacial adhesion between the CW filler and LDPE matrix. Furthermore, the more obvious morphological difference was observed at a magnification of 3000 \times for the LDPE/untreated CW biocomposite sample (Figure 4c), which indicates some voids. The formation of such voids towards the fracture force indicated that the weak interactions between the LDPE and CW. In addition, numerous voids are located in the fractured surface also caused by slips of fibers [25].

On the contrary, the LDPE/treated CW biocomposite sample (Figure 4d) exhibited opposite phase morphology, which implies that the sample has better interfacial adhesion. Generally, the tensile and morphological properties of the biocomposites depend strongly on the intermolecular interactions between two components (natural filler and polymer matrix). Thus, FTIR analysis was conducted to determine the types of interactions between them.

Figure 3. SEM (scanning electron microscope) micrographs of the surfaces of the (a) untreated CW; (b) treated CW (at magnification of 50×); (c) untreated CW and (d) treated CW (at magnification of 3000×).

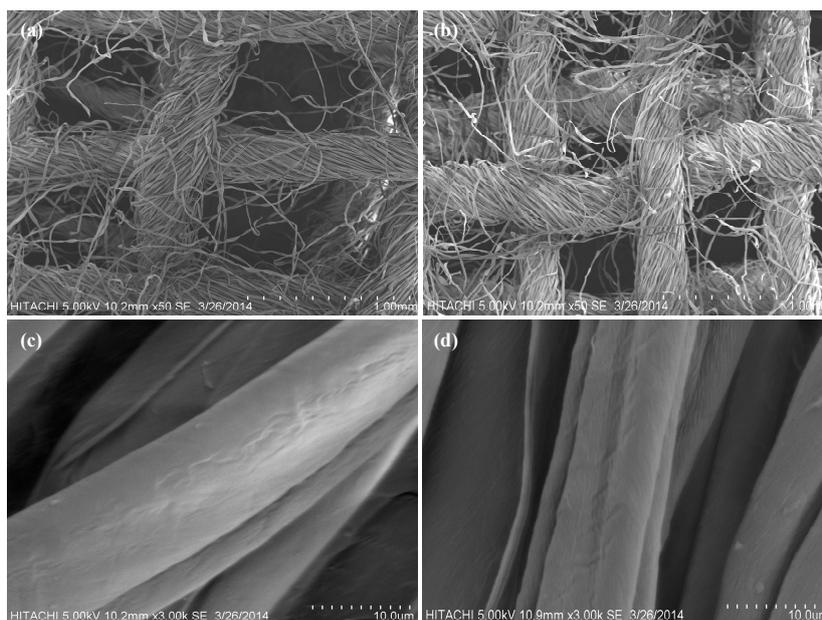
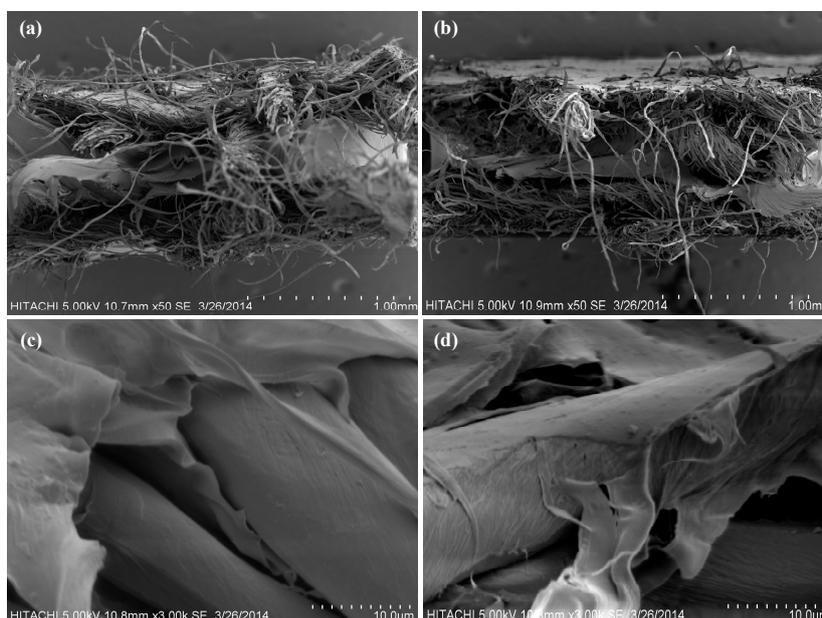


Figure 4. SEM micrographs of the fractured surfaces of the (a) LDPE/untreated CW biocomposite; (b) LDPE/treated CW biocomposite (at magnification of 50×); (c) LDPE/untreated CW biocomposite; and (d) LDPE/treated CW biocomposite (at magnification of 3000×).



3.3. FTIR Study

Figure 5 displays the FTIR spectra of the untreated CW, treated CW, LDPE, LDPE/untreated CW and LDPE/treated CW biocomposites. The treated CW and its biocomposite samples that were used in FTIR characterization are acquired from the optimum parameter of treatment specifically 0.4 wt% HTAB for 2 h as SEM samples. The FTIR spectra of all samples except for LDPE revealed strong intensity broad bands in the range of 3336 to 3330 cm^{-1} that could be attributed to the O–H stretching of the alcohol group. On top of that, the obvious bands with strong intensity that is responsible for the CH_2 asymmetric stretching and CH_2 symmetric stretching of the methylene group discovered at the bands of 2913 and 2849 cm^{-1} , respectively [13]. The apparent bands with medium intensity in the region of 2902 to 2897 cm^{-1} that present in untreated CW and treated CW are associated with the C–H stretching of the alkane group [26]. The noticeable bands with strong intensity ranged from 1464 to 1461 cm^{-1} are due to the C–H bending deformation. The bands with weak intensity at around 1430 to 1428 cm^{-1} are caused by the O–H bending of the alcohol group. Besides that, the bands with strong intensity at around 1029 to 1025 cm^{-1} are could be ascribed to the C–O stretching of the alcohol group while the observable bands with medium intensity at nearly 720 to 715 cm^{-1} are correspond to the CH_2 rocking deformation [13,27]. On the other hand, the bands with weak intensity at around 438 to 431 cm^{-1} are assigned to the CH_3 wagging mode [28]. The appreciable bands with medium intensity in the range of 343 to 333 cm^{-1} that revealed the C–C out of plane bending are present in all samples except for LDPE. It can be seen that the FTIR spectrum of untreated CW is same as cellulose powder spectrum as indicated in the previous study [5]. This result has confirmed that the woven fibers were made from pure cellulose.

Figure 5. FTIR spectra of the untreated CW, treated CW, LDPE, LDPE/untreated CW and LDPE/treated CW biocomposites.

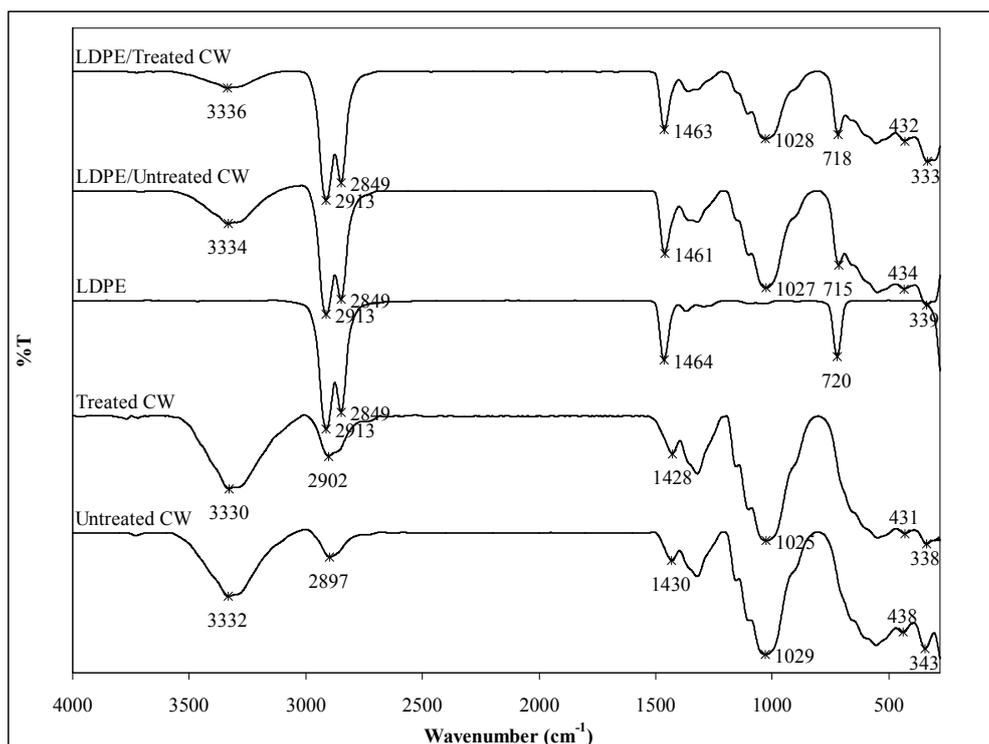
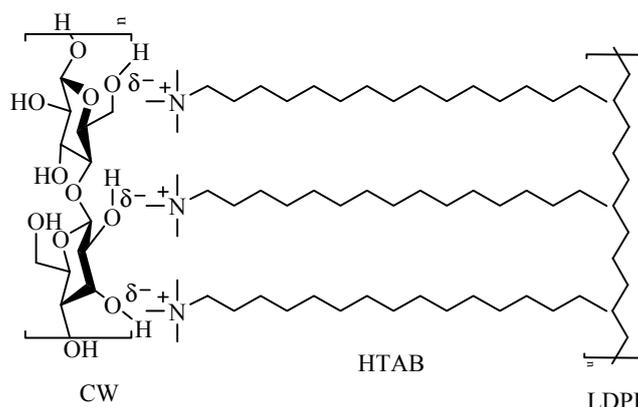


Table 1 presents the FTIR bands of the untreated CW, treated CW, LDPE, LDPE/untreated CW and LDPE/treated CW biocomposites. It was found that the untreated CW and treated CW have slightly different wavenumber in their spectra. This is due to the fact that when CW was treated with HTAB, changes in characteristic bands occurred as result of intermolecular interactions. The bands of the O–H stretching, O–H bending and the C–O stretching of the alcohol group of treated CW significantly shifted to the lower wavenumber. Moreover, an identical property has been observed for the bands of the CH₃ wagging and the C–C bending when CW treated with HTAB. This is because the increase in the formation of the ion-dipole force between the polar hydroxyl groups that weakly negatively charged (δ^-) on the surfaces of CW [29,30] and the polar quaternary ammonium cation groups (δ^+) of the HTAB molecules [15]. In contrast, it can be seen that in Table 1, the bands of the C–H stretching of the alkane group of treated CW considerably shifted to higher wavenumber. This is also due to the presence of the non-polar alkyl group of the HTAB on the surface of treated CW.

On top of that, the LDPE/untreated CW and LDPE/treated CW biocomposites also indicated a different wavenumber in their FTIR spectra. The bands of the O–H stretching and C–O stretching of the alcohol group as well as the C–H bending and CH₂ rocking of the biocomposite that filled with treated CW significantly shifted toward higher wavenumber as compared with the untreated ones. These changes were caused by the decrease in the intermolecular hydrogen bonding of the treated CW and also decrease in the intermolecular bonding of the LDPE. Meanwhile, the bands of the CH₃ wagging and C–C bending of LDPE/treated CW biocomposite shifted toward lower wavenumber compared to the LDPE/untreated CW biocomposite. This is due to the non-polar alkyl groups of HTAB have more affinity to form hydrophobic-hydrophobic interaction with the non-polar groups of LDPE. Hence, HTAB has acted as an intermediary for providing intermolecular interactions (ion-dipole force and hydrophobic-hydrophobic interaction) between LDPE matrix and CW filler. All these interactions have eventually led to good interfacial adhesion and good stress transfer from polymer matrix to the natural filler as previously demonstrated in the tensile testing results. Therefore, an interactional structure for the LDPE/treated CW biocomposites was suggested as displayed in Figure 6.

Table 1. FTIR bands of the untreated CW, treated CW, LDPE, LDPE/untreated CW and LDPE/treated CW biocomposites.

Sample	Wavenumber (cm ⁻¹)									
	O–H stretching	CH ₂ stretching	C–H stretching	CH ₂ stretching	C–H bending	O–H bending	C–O stretching	CH ₂ rocking	CH ₃ wagging	C–C bending
Untreated CW	3332	-	2897	-	-	1430	1029	-	438	343
Treated CW	3330	-	2902	-	-	1428	1025	-	431	338
LDPE	-	2913	-	2849	1464	-	-	720	-	-
LDPE/untreated CW	3334	2913	-	2849	1461	-	1027	715	434	339
LDPE/treated CW	3336	2913	-	2849	1463	-	1028	718	432	333

Figure 6. Suggested interactional structure for the LDPE/treated CW biocomposites.

4. Conclusions

The optimum HTAB concentration for treatment of CW in one hour was 0.4 wt%, while the optimum treatment time at 0.4 wt% HTAB was 2 h, based on the tensile testing results of LDPE/CW biocomposites. There is no significant difference in the morphology of the untreated and treated CW as indicated in the SEM images, nevertheless the morphology of the LDPE/treated CW biocomposite showed better interfacial adhesion when compared with the one without treatment. The presence of HTAB on the surface of treated CW and the existence of intermolecular interactions between LDPE and treated CW were confirmed by the FTIR spectra. It can be summarized that the surface of CW and tensile properties of LDPE/CW biocomposites could be modified and improved, respectively, by using HTAB as a treatment agent.

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Author Contributions

The preliminary study presented here was conducted in cooperation between all authors. Azmah Hanim Mohamed Ariff supervised and conceived the research project. Ahmad Adlie Shamsuri designed and constructed the research methods. Muhammad Kamarul Azroy Azid performed most of the experiments. Ahmad Khuzairi Sudari collected and analyzed the data. All authors have contributed to the writing of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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