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Influence of hydrophobic and hydrophilic mineral fillers on processing, tensile and impact properties of LDPE/KCF biocomposites



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ABSTRACT

In this research, low-cost thermoplastic such as low-density polyethylene (LDPE) was used as a matrix for preparation of the biocomposites. On the other hand, kenaf core fibre (KCF) has been chosen as natural filler due to its availability and renewability. The hydrophobic (talc) and hydrophilic (calcium carbonate) minerals with different loading (0 to 15 wt%) were utilised as secondary fillers. The biocomposites were prepared via melt mixing technique. The processing, tensile and impact properties of the prepared biocomposites were recorded, measured and compared. From the processing recorder results, it was found that the stabilization torque of the LDPE/KCF biocomposites containing talc is lower than calcium carbonate. For the tensile test results, it can be observed that the tensile strength and tensile modulus of the biocomposites with talc are greater than calcium carbonate. It could be concluded that the LDPE/KCF biocomposites with the incorporation of hydropholic mineral possessed stiff character, whereas the incorporation of hydrophilic mineral provided tough behaviour to the biocomposites.

1. Introduction

Kenaf core fibre (KCF) is a by-product in kenaf fibre industry, it was used as a primary filler in this research due to its price is lower than kenaf bast fibre. In our previous research, we also have produced biocomposites by using KCF together with low-density polyethylene (LDPE) [1]. It is one of the ways for overcoming environmental problems since natural fibres were used in production of biocomposites. However, the performances of prepared biocomposites are usually unsatisfactory although the natural fibres have minimised the production cost of the biocomposite products [2]. This is because of the poor compatibility between synthetic polymer especially polyethylene and natural fibre [3]. Therefore, to settle this problem the use of chemicals has been proposed as the surface treatments of the natural fibres [4]. Nevertheless, since the chemicals are expensive and required more energy and time to apply, hence there are some limitations of using them [5].

Talc is a hydrous magnesium silicate with the chemical formula of $Mg_3Si_4O_{10}(OH)_2$ which is one of the hydrophobic minerals [6]. The basal surfaces of talc are hydrophobic and its edge surfaces are hydrophilic [7]. The hydrophobic nature of the basal surfaces is due to the atoms exposed on the surface are linked together by siloxane (Si-O-Si)

bonds, thus they could not form strong hydrogen bonding with water [8]. On the other hand, talc is also important as reinforcing filler for polyolefin.

Calcium carbonate with the chemical formula of $CaCO_3$ is a hydrophilic mineral [9], low-cost and non-toxic substance that has been widely used as functional filler in polymer composites for improving their physicochemical properties. Calcium carbonate mostly used for polyethylene and PP [10]. Various thermoplastic composites have achieved outstanding improvements in mechanical and thermal properties, dimensional stability, gas permeability and biodegradability with the incorporation of calcium carbonate as fillers.

The aim of this research is to study the influence of talc and calcium carbonate on the processing, tensile and impact properties of the LDPE/KCF biocomposites. As far as we are concerned, there are little published articles pertaining to the utilisation of the hydrophobic and hydrophilic minerals as secondary fillers for the biocomposites. Thus, these minerals were also compared with each other for perceiving their effects on stiffness and toughness characteristics of the biocomposites.

2. Materials and methods

LDPE (coating grade) was purchased from the Lotte Chemical Titan

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(M) Sdn. Bhd., Malaysia. KCF (420 μ m) was attained from the National Kenaf and Tobacco Board, Malaysia [11]. Talc (10 μ m) was procured from the Sigma-Aldrich (M) Sdn. Bhd., Malaysia. Calcium carbonate (10 μ m) was acquired from Merck (M) Sdn. Bhd., Malaysia. All materials were used as received without further modification.

Brabender internal mixer machine was used to prepare the biocomposites. The machine was equipped with a real-time processing recorder. The mixing was done at a temperature of 150 °C, and the rotor speed was fixed at 60 rpm. First of all, 24 g of LDPE was inserted into the mixing chamber, and allowed to melt for 3 min. After that, 16 g of KCF was added into the chamber, and permitted to mix for 6 min. Finally, talc or calcium carbonate was incorporated into the composite, and allowed to blend for 6 min. The period of the whole process was $15 \min [1]$. The contents of the talc and calcium carbonate were varied as 0, 3, 9 and 15 wt%.

The prepared biocomposite samples were converted into a 1 mm sheet through the compression moulding technique by using a hydraulic hot press machine. The moulding processes involved are preheating of mould containing the sample at 150 °C for 7 min, compression of the sample at the same temperature for 2 min, and then cooling of the sample at 20 °C for 5 min [12].

The sheet-shaped biocomposite samples were cut into dumbbell (types V) and rectangular ($60 \times 13 \text{ mm}^2$) shapes by using die cutter and scroll saw, respectively. The dumbbell- and rectangular-shaped samples were dried in an oven at a temperature of 70 °C for at least 24 h prior to characterizations [13].

The maximum tensile strength, tensile modulus, and tensile strain properties of the biocomposite samples were measured according to the ASTM D638-10 standard test method at room temperature (25 °C) by using an Instron universal testing machine (model 5567) equipped with a 30 kN load cell. The crosshead speed and gauge length were 5 mm min⁻¹ and 40 mm, respectively. 10 replicates were done for each sample to determine the average values, and the standard deviation ranges were also reported to show the error range [14].

The impact strength of the biocomposite samples was determined in accordance with the ASTM D256-10 standard test method at room temperature (25 °C) by using an Instron impactor machine (CEAST 9050) equipped with a 0.5 J pendulum. The samples were notched up to 1 mm depth by using a V-notch machine (CEAST Notchvis). The average values from 10 replicates of each sample were calculated, and the ranges of standard deviation were indicated as well [14].

3. Results and discussion

3.1. Processing characteristics

Figs. 1 and 2 demonstrated the processing torque-time curves of the LDPE/KCF biocomposites with different contents of talc and calcium carbonate minerals, respectively. Processing torque is one of the



Fig. 1. Processing torque versus time for talc mineral.



Fig. 2. Processing torque versus time for calcium carbonate mineral.

processing behaviours which was recorded during the processing of the biocomposite samples. At the first minute period interval, the sharp increase peaks in the processing torque curves around 10 to 15 Nm were obtained for all samples during the mixing process. This happened because of the unmelted LDPE has increased the resistance on the internal mixer rotors. However, the peaks started to decrease around 6 to 9 Nm with the increasing of the mixing time as the melting of LDPE took place. At the fourth minute, the processing torques began to rise again to the values from 21 to 24 Nm immediately after the incorporation of KCF to all samples. This is due to the fact that the KCF filler needed more forces for dispersing in the molten LDPE. Again the torques were started to decrease as the KCF became very well dispersed in the LDPE matrix.

For the sample with 0 wt% of mineral, the processing torque has decreased and remained almost unchanged at a particular level until the end of the total mixing time, this is due to the completed KCF dispersion. For the samples with 3 wt% of talc and calcium carbonate, the slight increases of the processing torques at around tenth minute showed that there was a small amount of friction between the minerals and molten biocomposites. However, for the samples with 9 and 15 wt % of talc and calcium carbonate, there were large amounts of frictions between the minerals and molten biocomposites and it displayed by the significant increases in the processing torques. As the dispersion of the minerals was accomplished, the torques slowly started to decrease. This is because of the decrease in the melt viscosity of the LDPE/KCF biocomposites. After the twelfth minute, the processing torques of all samples remained stable until the end of melt processing.

Figs. 3 and 4 illustrated the stabilization torque-mineral graphs of the LDPE/KCF biocomposites with different loadings of talc and calcium carbonate, respectively. The torque values at fifteenth minute of mixing process were considered as the stabilization torque values [1,11]. From the graphs in Figs. 3 and 4, it can be seen that for the biocomposites incorporated with minerals, they have higher stabilization torques compared to the biocomposite without mineral at a similar



Fig. 3. Stabilization torque versus talc loading.

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