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Effects of Kenaf and Rice Husk on Water Absorption and Flexural Properties of Kenaf/$CaCO_3$/HDPE and Rice Husk/$CaCO_3$/HDPE Hybrid Composites

Noor Zuhaira Abd Aziz, Rahmah Mohamed, Mohd Muizz Fahimi M.

***Abstract*—**Rice husk and kenaf filled with calcium carbonate ($CaCO_3$) and high density polyethylene (HDPE) composite were prepared separately using twin-screw extruder at 50rpm. Different filler loading up to 30 parts of rice husk particulate and kenaf fiber were mixed with the fixed 30% amount of $CaCO_3$ mineral filler to produce rice husk/$CaCO_3$/HDPE and kenaf/$CaCO_3$/HDPE hybrid composites. In this study, the effects of natural fiber for both rice husk and kenaf in $CaCO_3$/HDPE composite on physical, mechanical and morphology properties were investigated. Field Emission Scanning Microscope (FeSEM) was used to investigate the impact fracture surfaces of the hybrid composite. The property analyses showed that water absorption increased with the presence of kenaf and rice husk fillers. Natural fibers in composite significantly influence water absorption properties due to natural characters of fibers which contain cellulose, hemicellulose and lignin structures. The result showed that 10% of additional natural fibers into hybrid composite had caused decreased flexural strength, however additional of high natural fiber (>10%) filler loading has proved to increase its flexural strength.

***Keywords*—**Hybrid composites, Water absorption, Mechanical properties.

I. Introduction

NOWADAYS, incorporation of $CaCO_3$ as mineral filler into polymer matrix is extensively being applied in plastic industry mainly to reduce cost.

However, higher loading of $CaCO_3$ in wood plastic composite (WPC) could harm the strength properties of the composite. Therefore, in this research hybridization was applied to $CaCO_3$/HDPE composite to modify and develop the optimum formulation of composite properties according to desired application. Kenaf and rice husk as the natural fiber fillers were added into two different $CaCO_3$/HDPE composites. The main purpose of adding natural fiber filler into $CaCO_3$/HDPE composites is primarily due to its low cost property and at the same time, natural fiber actually could acts as the reinforcing filler to composite. The used of cellulosic material for reinforcing polyolefin matrices is the great interest due to the inherent advantages of cellulosic reinforcements [1]. Kenaf as fibrous filler and rice husk as particulate filler were considered in the study. Hence, in this research the effect on flexural and water absorption of natural fiber fillers in $CaCO_3$/HDPE composites were investigated.

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II. Experimental

A. Material

HDPE used in this study was supplied by TITAN (M) Sdn. Bhd. The 50 mesh kenaf fibers and rice husk particulate were obtained from MARDI. The 2000 mesh of $CaCO_3$ was obtained from ZANTAT Sdn. Bhd.

B. Sample Preparation

The rice husk flour was mixed with HDPE and $CaCO_3$. The compound mixture was melted by extruded using twin screw extruder at temperature profile range between 160^oC and 190^oC from feed to die zone. Compounded pallets were fabricated and compressed by hot press at the temperature of 190^oC. The same process was repeated and rice husk was substituted with kenaf. The compositions prepared are as given in Table I.

TABLE I
COMPOSITION OF THE STUDIED FORMULATIONS

Samples	HDPE (%)	Kenaf (%)	Rice Husk (%)	$CaCO_3$ (%)
C1	70	-	-	30
K1	60	10	-	30
K2	50	20	-	30
K3	40	30	-	30
RH1	60	-	10	30
RH2	50	-	20	30
RH3	40	-	30	30

C. Composite Characterization

Water absorption properties were measured according to ASTM D570-98. This test method determine the amount of water absorb into sample. The samples were undergoing the long term immersion. The weighing process was repeated every 24 hours for first two weeks and every once a weeks until the increase in weight per week shows constant result.

The water absorption was determined as:

$$\%\text{water absorption} = \frac{W_1 - W_0}{W_0} \times 100 \quad (1)$$

where W_1 is the mass of the sample after immersion, and W_0 is the mass of the samples before immersion.

Flexural test was measured according to ASTM D790 specification. The test was carried out using the three-point bending test Instron Machine. The cross-head speed applied was 5mm/min.

Morphological studies of the hybrid composites were carried out using a Field Emission Scanning Microscope (FESEM) SUPRA-40 VP with software INCA Suite versions 4.12 with the magnification, 5000x. The impact fracture surfaces of the specimen were observed by microscope. The fracture surfaces were gold-sputtered to prevent electrostatic charge build-up during evaluation.

III. Result and Discussion

A. Water Absorption

Fig. 1 shows the effect of kenaf and rice husk on water absorption of $CaCO_3$/HDPE composite. The water absorption of the composites were significantly increased in the early stage evidently for formulations C1 and K1 and continued until they reached the maximum and constant water uptake, this point is called saturation point and the duration of each formulations to reach the saturation point were different and this apply to all formulation [2]. Composite K1 is the earlier composite that reached saturation point at day 32 which was because K1 contained the lowest filler compared to other filled composites.

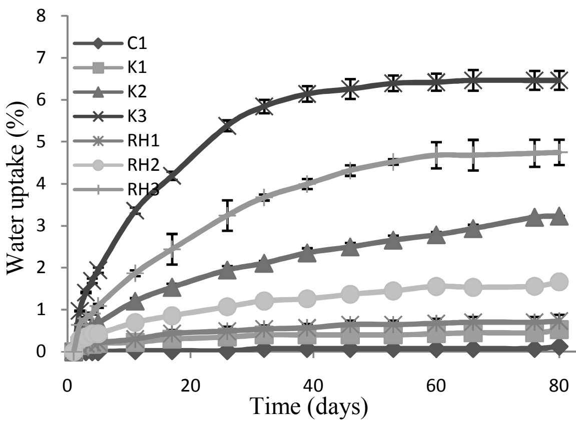


Fig. 1 Water absorption behaviors for hybrid composite

Water absorption of RH1 is higher than K1. Water absorption of RH1 which was filled with 10% rice husk shows higher percentage compared to K1 which was filled with 10% of kenaf filler, regardless their based composite which were made of $CaCO_3$/HDPE hybrid. This is due to the better interfacial adhesion between filler and matrix in K1 than RH1 that create fewer gaps in the interfacial region due to stronger interaction between fiber-matrix in hybrid composites. This was proved from previous study which K1 produce higher tensile strength than RH1 [3]. Poor interaction between fiber-matrix interactions due to incomplete wettability fiber-matrix adhesion in RH1 could cause higher voids entrapped since there are high gap in interfacial region thus lead to high water to accumulate at the interface between fiber-matrix [4]. However from the observation, this condition only occurs at low fiber loading (10%wt). Composites K2 and K3 have produced higher water absorption than RH2 and RH3 when compared with their similar filler loadings. Water absorption of the composite mainly due to the cellulosic material that increases the rates of water uptake by forming the hydrogen bonding between water and the hydroxyl group from cellulosic cell wall fiber [2], [4]. It was well exhibited that kenaf which contains higher cellulose (~45%) tend to form higher hydrogen bonding from its hydroxyl (OH) group and water molecules which indicates that it has higher tendency to absorb high amount of water. In contrast, rice husk contained lower cellulose amount (~35%) exhibited less hydrogen bonding since rice husk produce less free-OH group compare to kenaf.

B. Flexural Strength

Fig. 2 shows the effects of kenaf and rice husk on bending strength in $CaCO_3$/HDPE composite. The result shows that the additions of 10% of either kenaf or rice husk into $CaCO_3$/HDPE composites could decrease the bending strength of the hybrid composite.

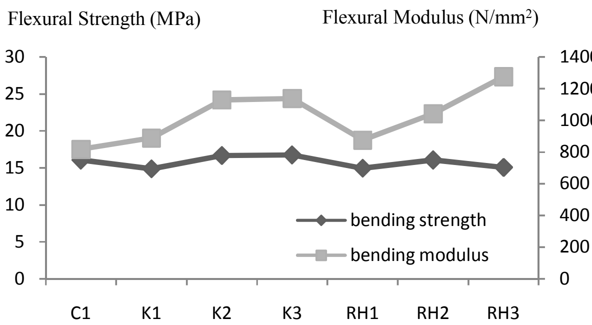
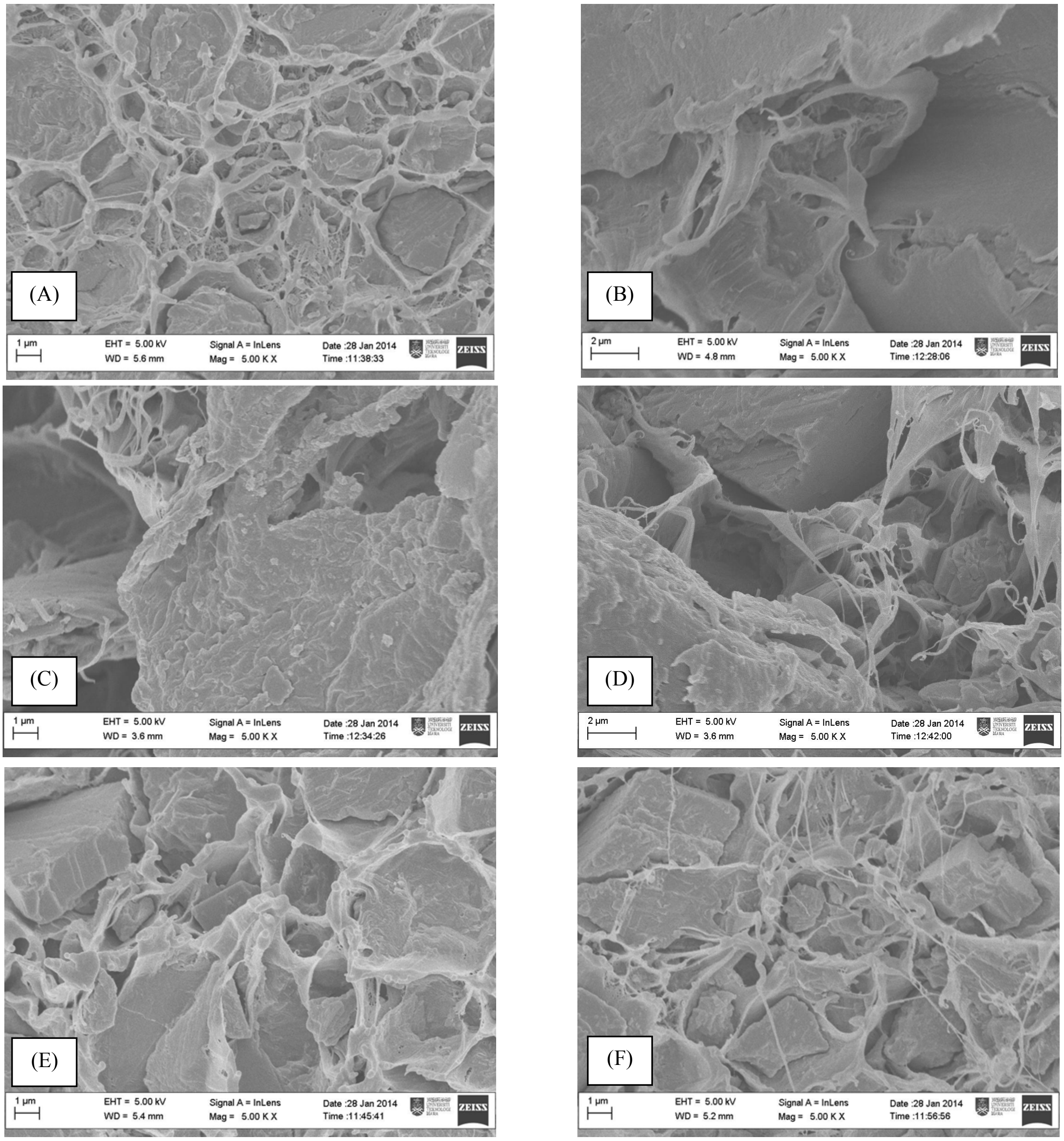


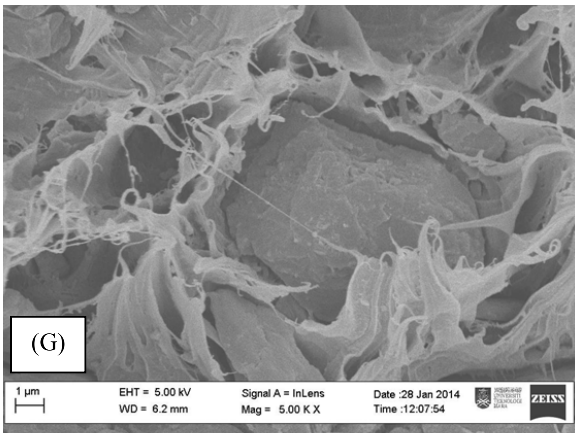
Fig. 2 Flexural strength and flexural modulus of the hybrid composites

However, it is interesting to observe that addition high natural fiber filler loading into hybrid composite had increase bending strength. This is due to better adhesion between kenaf and rice husk, separately with $CaCO_3$/HDPE composites at >10% fiber loading. This circumstance indicated that there were good interactions remaining between natural fibers filler with $CaCO_3$/HDPE composites [5]. However, 30% rice husk filled of $CaCO_3$/HDPE composite was significantly decreased the bending strength. This phenomenon is caused by the behavior of rice husk that tend to form higher agglomerates which could cause poor interfacial adhesion between filler and polymer matrix [6]. Flexural modulus increased with increasing natural fiber filler. The maximum flexural modulus was 1276.14 N/mm^2 in RH3 which contain higher rice husk filler. While K1 which contain 10% kenaf 30% $CaCO_3$ 60% HDPE showed lowest flexural modulus values, 729.331 N/mm^2.

C. Morphology

FeSEM micrographs of the fracture impact surface of hybrid composites are shown in Fig. 3. The impact fracture surface show a significant interfacial adhesion between two different phases: hydrophilic fillers of kenaf or rice husk with calcium carbonate and hydrophobic nature of HDPE polymer matrix. All the fillers used have higher hydrophilicity than the matrix. Hence, delamination and debonding were found to almost all system investigated for different composition. If actual incompatibility exists, the bonding between fillers and matrix tends to be weak due to the different phases. However, there is some composition which showed well dispersed fillers in polymer matrix.





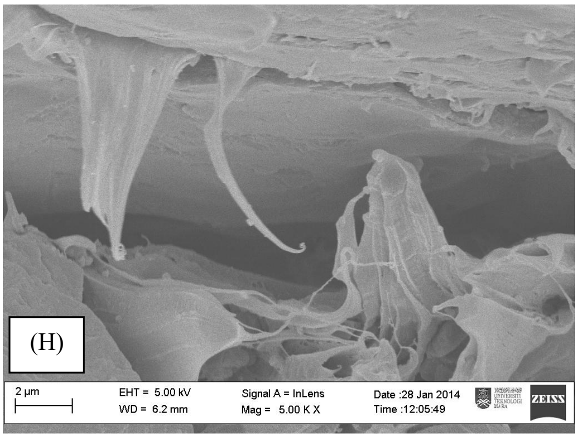


Fig. 3 FeSEM micrograph of impact fracture surface of natural fiber filler filled $CaCO_3$/HDPE composite at magnification x5000: (A) C1, (B) K1, (C) K2, (D) K3, (E) RH1, (F) RH2, (G) RH3 and (H) RH3

FeSEM micrographs of the impact fracture surface of 30% $CaCO_3$ mineral filler filled 70% HDPE polymer matrix is shown in Fig. 3 A. It can be observed that $CaCO_3$ mineral filler tend to produce higher agglomeration due to the smaller size particles (1500mesh). However, with addition of natural fiber smoother fractured surface can be observed in Fig. 3 B and C as compared to other hybrid composites. A well dispersion and strong interfacial adhesion also can be observed due to better distribution and complete wettability between natural fiber and mineral filler with HDPE polymer matrix. It is interested to observe that micrograph in Fig. 3 B shows better dispersion of kenaf fiber in $CaCO_3$/HDPE composite as compared to rice husk particulate in $CaCO_3$/HDPE (Fig. 3 E). The better performance of fiber reinforcement effect had also been investigated by Han Seung Yang et al. and G. R. Liu which also postulated that the fiber did produce higher strength due to better interaction between filler and polymer matrix [7], [8]. The forms of fillers did show differences in the fracture morphology of the fiber and particulate interaction adhesion between fillers-matrix. No fiber pull out were exhibited in lower filler content of 10% kenaf or rice husk in $CaCO_3$/HDPE composite.

The strong interlocking bonding between filler and polymer matrix start to reduce as the filler were increase from 20% which can be observed in Figs. 3 C and F. The arrangement of kenaf and rice husk will result in less compaction of the fillers as fillers are more random and disoriented. The different in the shape and forms between fillers tend to produce more cavities and voids. It can be observed that, increase filler especially rice husk could cause deterioration in the hybrid composites whereas with kenaf, the reduced in strength is much less.

Figs. 3 D and G which contain 30% kenaf and 30% rice husk respectively, show the inhomogeneous dispersion and the wetting of filler by polymer matrix was reduced. Hence, increase the tendency of filler to agglomerate and cause higher void is more significant as compare to lower filled composites. From Figs. 3 D and G, the presence of filler was more visible as more filler were present and more defect site of filler-matrix interfacial was observed. From micrographs of Figs. 3 D and G, poor interfacial adhesion could be observed from the high amount and large size of agglomerate in the micrographs as the content of hybrid fillers composed of kenaf fiber/$CaCO_3$ and rice husk/$CaCO_3$ are about 60%. Higher filler content will result in more and larger agglomerate due to stronger filler-filler interaction which is shown in Fig. 3 H, where the phase of delamination and non-adherence predominates of rice husk particulate in $CaCO_3$/HDPE composite could be observed.

IV. Conclusion

As a conclusion, the addition of either kenaf or rice husk into $CaCO_3$/HDPE composites tends to increase the water absorption of the hybrid composites due to the forming hydrogen bonding between water and the hydroxyl group from natural fibers. On the other hand, high loading (>10%) of kenaf and rice husk were proved to increase the flexural strength and flexural modulus. However, addition 30% rice husk has caused rapid decrease of flexural strength. FeSEM studies of impact fracture surface of the hybrid composite indicate the well dispersed at lower fillers loading either kenaf or rice husk with $CaCO_3$ in HDPE polymer matrix. However, poorer interfacial adhesion could be found in high filler loading (>50%) due to incompatibility between fillers and polymer matrix.

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