

# Tensile properties and water absorption assessment of linear low-Density Polyethylene/Poly (Vinyl Alcohol)/Kenaf composites: effect of eco-friendly coupling agent

A L Pang<sup>\*1</sup>, H Ismail<sup>1,2</sup> and A Abu Bakar<sup>1</sup>

<sup>1</sup> School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia (USM), Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia

<sup>2</sup> Cluster for Polymer Composites (CPC), Science and Engineering Research Centre (SERC), Universiti Sains Malaysia (USM), Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia

\*E-mail: pangailing87@yahoo.com

**Abstract.** Linear low-density polyethylene (LLDPE)/poly (vinyl alcohol) (PVOH) filled with untreated kenaf (UT-KNF) and eco-friendly coupling agent (ECA)-treated kenaf (ECAT-KNF) were prepared using ThermoHaake internal mixer, respectively. Filler loadings of UT-KNF and ECAT-KNF used in this study are 10 and 40 parts per hundred parts of resin (phr). The effect of ECA on tensile properties and water absorption of LLDPE/PVOH/KNF composites were investigated. Field emission scanning electron microscopy (FESEM) analysis was applied to visualize filler-matrix adhesion. The results indicate LLDPE/PVOH/ECAT-KNF composites possess higher tensile strength and tensile modulus, but lower elongation at break compared to LLDPE/PVOH/UT-KNF composites. The morphological studies of tensile fractured surfaces using FESEM support the increment in tensile properties of LLDPE/PVOH/ECAT-KNF composites. Nevertheless, LLDPE/PVOH/UT-KNF composites reveal higher water absorption compared to LLDPE/PVOH/ECAT-KNF composites.

## 1. Introduction

It is a regular trend at present among researchers, industries and governments focusing on developing new products based on natural resources that minimize the impact to environment [1]. The use of natural resources such as kenaf (KNF) as reinforcement or filler in polymers is being actively investigated [2-4]. Kenaf offers a wide range of merits such as light in weight, environmental friendly, available in abundance and so on [5-7].

Due to the fact that KNF is naturally hydrophilic, it tends to absorb moisture and exhibit poor filler-matrix adhesion in polymer composites [6,8]. Subsequently, the mechanical properties of KNF-polymer composites are affected and to tackle this weakness, chemical treatment of KNF and/or modification of polymers can be applied to enhance filler-matrix adhesion [3]. There are vast studies on chemical treatment of KNF composites conducted in the interest of enhancing adhesion between KNF and matrix materials [9-13].

In this work, surface modification of KNF with eco-friendly coupling agent (ECA) was performed, before using it as filler into LLDPE/PVOH blend. The effect of ECA on tensile properties, morphological properties, and water absorption of LLDPE/PVOH/KNF composites were examined.



## 2. Experimental

### 2.1. Materials

LLDPE was supplied by PT. Lotte Chemical Titan Nusantara, Indonesia with melt flow rate of 1 g/10 min at 190 °C and a density of 0.92 g/cm<sup>3</sup>. PVOH was purchased from Sigma-Aldrich (M) Sdn. Bhd., with molecular weight of 89,000 to 98,000 g/mol and density of 1.269 g/cm<sup>3</sup>. Epichlorohydrin and benzyl-trimethylammonium chloride were products from Sigma-Aldrich (M) Sdn. Bhd., with molecular weight of 92.52 and 185.69 g/mol, respectively. Ethanol was supplied by Merck (M) Sdn. Bhd. with molar mass of 46.07 g/mol. KNF was obtained from National Kenaf and Tobacco Board (LKTN), Kelantan, Malaysia.

### 2.2. Characterizations

Universal Testing Machine (Model: Instron 3366) was used to determine tensile properties (strength, modulus and elongation at break) of composites, according American Society for Testing and Materials (ASTM D638-Type IV). The tensile fractured surfaces morphology of composites was examined under a FESEM (Zeiss Supra-35VP). Water absorption was carried out in accordance to ASTM D570, and the percentage of water absorption was calculated as Equation 1:

$$W_t(\%) = \frac{W_2 - W_1}{W_1} \times 100 \quad (1)$$

where  $W_t$  is the total water absorption by composites,  $W_1$  and  $W_2$  are the weights of composites before and after immersion in distilled water, respectively.

## 3. Result and discussion

### 3.1. Tensile properties

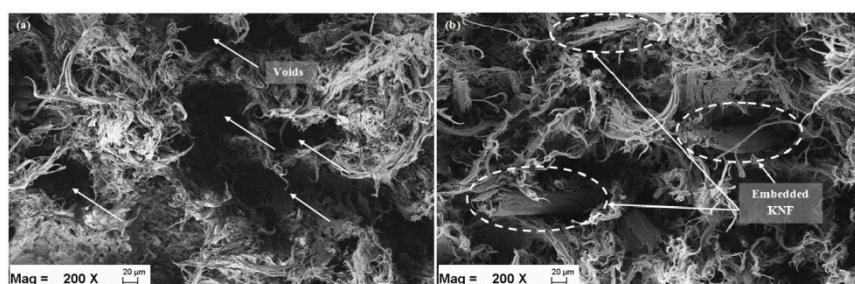
Table 1 displayed the tensile properties of LLDPE/PVOH/UT-KNF and LLDPE/PVOH/ECAT-KNF composites. The tensile strength and elongation at break was found to decrease, while the tensile modulus increases with respect to KNF loading (from 10 phr to 40 phr). This is because of the inability to transfer the load effectively resulting from weak interfacial adhesion between filler and matrix, as well as poor wettability of filler by the matrix [2,4]. Furthermore, it is visible that composites with ECAT-KNF exhibit higher tensile strength and tensile modulus, but lower elongation at break when compared to composites with UT-KNF. The increase in tensile strength and tensile modulus implied that interfacial adhesion between KNF and LLDPE/PVOH was improved in the presence of ECA, and hence permitting efficient stress transfer to occur. Similar findings were observed by Nurfatimah et al. [9] and Majid et al. [12]. Study by Majid et al. [9] shows that benzoyl treatment of kenaf core powder (KCP) improved the adhesion of KCP and polyvinyl chloride/epoxidized natural rubber (PVC/ENR), thereby increase the composites' tensile properties. Nevertheless, the drop in elongation at break is expected, mainly due to the decrement in the deformability of composites after ECA treatment. The possible reactions between ECA, KNF and LLDPE/PVOH matrices are as follow: (i) The interaction of hydroxyl groups of KNF with the oxirane group of ECA [14] (ii) The hydrophobic chain in ECAT-KNF is likely to interact with the hydrocarbon chains of LLDPE/PVOH matrices by intermolecular attraction.

**Table 1.** Tensile properties of LLDPE/PVOH/UT-KNF and LLDPE/PVOH/ECAT-KNF composites.

KNF loadings (phr)	LLDPE/PVOH/UT-KNF composites			LLDPE/PVOH/ECAT-KNF composites		
	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)
10	$8.6 \pm 0.17$	$796.5 \pm 23.71$	$6.5 \pm 0.56$	$9.2 \pm 0.06$	$921.3 \pm 9.25$	$5.5 \pm 0.13$
40	$7.1 \pm 0.15$	$1169.0 \pm 25.54$	$3.0 \pm 0.13$	$7.8 \pm 0.27$	$1397.0 \pm 25.17$	$2.2 \pm 0.34$

### 3.2. Morphological study

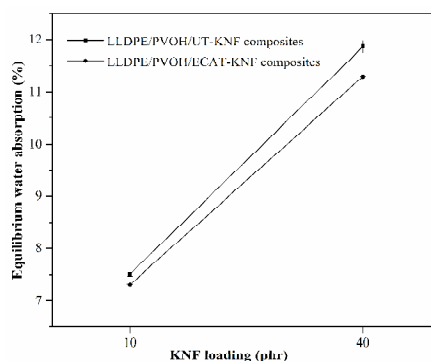
Figure 1 illustrates tensile fracture surfaces morphology of LLDPE/PVOH/UT-KNF and LLDPE/PVOH/ECAT-KNF composites at 10 phr KNF loading. As seen in Figure 1a, there are many voids presence on composites fractured surfaces owing to the detachment of UT-KNF from LLDPE/PVOH matrices after stress application. This observation confirms the poor compatibility between UT-KNF and LLDPE/PVOH caused by their different polarity. Nevertheless, it was found that interfacial adhesion between ECAT-KNF and LLDPE/PVOH matrices was improved, as shown by less voids presence on fractured surfaces. ECAT-KNF was seen well dispersed and embedded firmly within LLDPE/PVOH matrices after application of tensile stress.



**Figure 1.** FESEM micrographs on tensile fractured surfaces of LLDPE/PVOH/10 phr KNF composites with (a) UT-KNF (b) ECAT-KNF, respectively.

### 3.3. Water absorption

Figure 2 shows the equilibrium water absorption of LLDPE/PVOH/KNF composites under the effect of ECA, respectively. It shows ECAT-KNF composites exhibit lower equilibrium water absorption than that of UT-KNF composites, at both 10 phr and 40 phr KNF loading, respectively. This indicates that ECA treatment has enhanced the adhesion between KNF and LLDPE/PVOH matrix, thus decreasing the formation of micro-voids between them. Subsequently, it limits water from diffuse into ECAT-KNF composites. Based on Figure 2 also, equilibrium water absorption of UT-KNF and ECAT-KNF composites was increased with increasing KNF loading. This is attributed to more hydroxyl groups from KNF to form hydrogen bonding with water, and also presence of micro-voids between KNF and LLDPE/PVOH matrices at higher filler loading.



**Figure 2.** Effect of ECA on equilibrium water absorption of LLDPE/PVOH/KNF composites.

#### 4. Conclusion

The effect of ECA on the tensile properties and water absorption of LLDPE/PVOH/KNF composites were studied, and it is apparent that LLDPE/PVOH/ECAT-KNF composites have more optimal results than that of LLDPE/PVOH/UT-KNF composites. The tensile strength and tensile modulus show increment with the ECA treatment. ECA was found effective in enhancing KNF-LLDPE/PVOH interfacial adhesion, as proven in the FESEM micrographs. Furthermore, LLDPE/PVOH/ECAT-KNF composites exhibit lower equilibrium water absorption, mainly attributable to the good adhesion between KNF and LLDPE/PVOH.

#### Acknowledgement

The authors are grateful to Malaysia Higher Education, RUC research grant (No: 1001/PKT/8640014) from Universiti Sains Malaysia and National Kenaf and Tobacco Board (LKTN), Kelantan, Malaysia for sponsoring and supporting this research.

#### References

- [1] Mariotti N, Wang X M, Rodrigue D and Stevanovic T 2014 *J. Mater. Sci. Res.* **3** 8
- [2] Sarifuddin N, Ismail H and Ahmad Z 2013 *J. Phys. Sci.* **24** 97
- [3] Saba N, Paridah M T and Jawaid M 2015 *Const. Build. Mater.* **76** 87
- [4] Pang A L, Ismail H and Abu Bakar A 2015 *Biores.* **10** 7302
- [5] Akil H M, Omar M F, Mazuki A A M, Safiee S, Ishak Z A M and Abu Bakar A 2011 *Mater. Des.* **32** 4107
- [6] Pang A L and Ismail H 2013 *J. Thermoplast. Compos. Mater.* **27** 1607
- [7] Kamal I, Thirmizir M Z, Beyer G, Saad M J, Abdul Rashid N A and Abdul Kadir Y 2014 *J. Sci. Technol.* **6** 41
- [8] Tan B K, Ching Y C, Gan S N, Ramesh S and Rahman M R 2014 *Mater. Res. Inno.* **18** 144
- [9] Nurfatimah B, Chee C Y, Abdullah L C, Ratnam C T and Azowa N 2014 *Compos. Part A. Appl. Sci. Manuf.* **63** 45
- [10] Tawakkal I S M A, Marlene J C and Stephen W B 2014 *Ind. Crops. Prod.* **61** 74
- [11] Datta J and Kopczynska P 2015 *Ind. Crop. Prod.* **74** 566
- [12] Majid R A, Ismail H and Taib R M 2016 *Procedia. Chem.* **19** 803
- [13] Pang A L, Ismail H and Abu Bakar A 2016 *Biores.* **11** 5889
- [14] Muresan E I, Piroi C, Creanga D, Stelea L, Oprica L and Sandu I 2016 *Rev. Chim.* **67** 871