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THE INFLUENCE OF FILLER LOADING AND THE CHEMICALLY-MODIFIED FILLER SURFACE ON THE MECHANICAL PROPERTIES OF PALM KERNEL SHELL REINFORCED POLYESTER COMPOSITES

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ABSTRACT

Composites of unsaturated polyester (UP)/palm kernel shell (PKS) were prepared by hand lay-up technique. The effect of filler loading and modified PKS surface with silane coupling agent on mechanical properties and morphology of UP/PKS composites were studied. Tensile and flexural tests were done to determine the effect of filler loading on mechanical properties of composites. A silane coupling agent, 3-aminopropyltriethoxysilane (3-APE) was used to improve fiber-matrix adhesion. Increased of PKS filler loading in composites have decreased the value of tensile and flexural strength due to poor wettability and dispersion of PKS in matrix, but an increased in tensile and flexural modulus, indicate that stiffness was increased. However, the composites showed further improvement in these properties when treated PKS along with 3-APE was used at the same composition of filler due to better interfacial adhesion between filler and matrix. The study of Scanning Electron Microscope (SEM) showed the filler-matrix interaction was improved with an incorporation of 3-APE. Fourier Transform InfraRed Spectroscopy (FTIR) for chemically-modified filler showed the appearance of new peak indicating the interaction between 3- APE and PKS filler have been obtained.

Keywords: Unsaturated Polyester; Palm Kernel Shell; Silane coupling agent; Composites; Mechanical properties

INTRODUCTION

The use of green natural bio-fillers has captured great interests for researchers recently in development of polymer composite. Open burning of agricultural waste that leads to ozone depletion and global warming as well as depletion of petroleum source which cause a rise of price in the petroleum based-product are the factors that contribute to the

development of bio-degradable polymer composites. Since Malaysia is among the world's largest producer of oil palms, its waste is abundantly available which has the potential to be used as filler in polymer composites to replace synthetic fillers. Natural fiber composites possesses several advantages such as lightweight, renewable in nature, small cost, high modulus and specific strength. Green composites will be easily disposed or recycled, which is not possible with synthetic fiber [1]. However, hydrophilic nature of natural bio-fillers limits its potential as reinforcing agent due to poor interfacial adhesion with the hydrophobic matrix, leading to poor mechanical properties of composites [2]. Hence, surface modification of fillers is very important to overcome this disadvantage. The surface modification on the PKS can be chemically modified by coupling the surface with 3-aminopropyltriethoxysilane [3]. According to Shinsuke & Hiroyuki [4], the mechanical properties of a microfibrillated cellulose composite was increased with silane coupling agent due the enhancement in the interfacial adhesion between fibers and matrix. Composites with silanized sisal fiber with 2wt % of silane at different amount of fiber also shown significant enhancement of mechanical properties as better interfacial adhesion of fiber and matrix obtained [5]. In this study, the mechanical properties of UP/PKS composites obtained with coupling the surface with 3-aminopropyltriethoxysilane were compared with the UP/PKS composites obtained without any surface modification with silane coupling agent.

EXPERIMENTAL

Materials.
The palm kernel shell (PKS) was obtained from Malpom Industries Sdn. Bhd., Penang. Unsaturated Polyester with the grade Reversol P 9509 and Methyl Ethyl Ketone Peroxide (MEKP) catalyst were obtained from Echemo Trading Sdn. Bhd. and Zarm Scientific & Supplies Sdn. Bhd., Penang, respectively. Silane coupling agent, 3-aminopropyltriethoxysilane (3-APE) and ethanol were supplied by Fluka.

Preparation of Palm Kernel Shell (PKS).
The palm kernel shell was cleaned manually and dried at 80°C for 24 hours. The dried PKS was crushed and grinded into powder form. The powder was then sieved with a 63 µm size sieve. The average particle size of PKS obtained was analysed by using Melvern Particle Size Analyser Instrument. The organic and inorganic content of PKS was determined by Loss of Ignition measurement and the result obtained is 98.16% and 1.84% of organic and inorganic content, respectively.

Filler Treatment.
3-APE was dilute in ethanol solution. The amount of 3-APE used is 3% by weight of filler. The filler was charged into a chamber mixer and solution was added slowly to ensure uniform distribution of 3-APE. The filler was then continuously mixed for 1 hour. The treated filler then dried at 80°C for 24 hours to allow a complete evaporation of ethanol.

Preparation of Composites.
The hand lay-up technique has been used for preparation of UP/PKS composites. PKS was first added into polyester and stirred manually at room temperature and then 2php of MEKP catalyst was added. The composites were poured into a mold and cured in an oven at 90°C for 10 minutes. PKS composition in preparation of untreated and treated UP/PKS composites are 0, 15, 30, 45, 60 php, respectively.

Mechanical Testing
Tensile Test.
Tensile test was carried out according to ASTM D 638 using a Universal Testing Machine, Instron model 5569. A sample of rectangular bar specimens with 3 mm thickness, 15 mm width and 150 mm length were cut by using a cutter machine. The gauge length was set at 100 mm and the test was performed at 25 ± 3 °C with the cross head speed of 5 mm/min. Tensile properties for 5 identical samples of each composition were measured and the average values were reported.

Flexural Test.
Flexural test was carried out according to ASTM D790 test method using a Universal Testing Machine, Instron model 5569. Rectangular bar specimens, with the same dimension with tensile test specimens were used. The gauge length was set at 50 mm and the test was performed at 25 ± 3 °C with the cross head speed of 2 mm/min.

Morphology Study
Scanning Electron Microscopy (SEM).
Morphology studies of the tensile fracture surfaces of all compositions of UP/PKS composites were carried out using a scanning electron microscopy model JOEL JSM-6460LA (SEM). The fracture surface specimens were mounted on aluminium stabs and sputter coated with a thin layer of palladium to avoid electrostatic charging during examination.

Characterization
Fourier-Transform Infrared Spectroscopy (FT-IR).
The FTIR analysis of the untreated and treated PKS was performed by a Perkin Elmer Spectrometer 2000 FT-IR. The PKS filler was dispersed in dry KBr powder and mixed to obtain fine particle. KBr pellet technique was applied with scanned range between 400-4000 $cm^{-1.}$

RESULTS AND DISCUSSION

Tensile Properties.
Figure 1a and 1b reveal the trends of tensile strength and tensile modulus for untreated and treated UP/PKS composites, respectively. It can be seen that the increase of filler loadings led to decrease of tensile strength of composites, which is due to poor wettability and dispersion of PKS in UP matrix. For all composites added with filler, the tensile strength for

chemically modified UP/PKS composites is higher than the composites obtained without the chemical modification (Figure 1a). This is due to better interfacial adhesion between the filler and the matrix after chemical modification. Strong adhesion between filler and matrix interface can cause better stress transfer from the matrix to the filler leads to a higher tensile strength [6]. The hydroxyl group on the cellulose fibers could react better with the silane coupling agent. The increment in mechanical properties of natural fiber composites modified with silane coupling agent has been reported [4,7-9] which is due to the enhancement of fiber matrix adhesion

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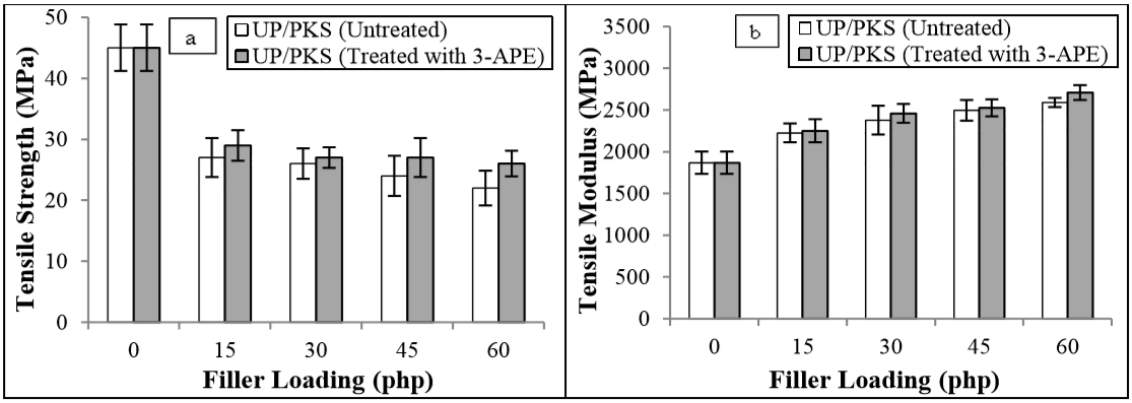

Figure 1: (a) Tensile strength (b) Tensile modulus of untreated and treated UP/PKS composites as a function of filler loading

It can be seen that the tensile modulus of both composites in Figure 1b gradually increased as the filler loading increased from 15 phr to 60 phr which showed addition of PKS increases the stiffness of composites. However, tensile modulus for chemically-modified UP/PKS composites is higher than the composites obtained without the chemical modification. This indicates that the compatibility and fiber-matrix adhesion between the PKS and UP is enhanced by addition of 3- APE. Girones *et. al.*, [10] stated that silane coupling agent can improve the compatibility between the components of the composites. The result also provides evident that the stiffness of the composites increased with the introduction of coupling agent. This result is in agreement with Shinsuke & Hiroyuki [4].

Flexural Properties.

Figure 2a and 2b show the flexural strength and flexural modulus of untreated and treated UP/PKS composites with 3-APE. For all composites added with filler, the flexural strength for chemically modified UP/PKS composites is higher than the composites obtained without the chemical modification (Figure 2a). At lower loading, rich area of UP matrix contributes to higher flexural strength. Higher flexural strength observed in treated composites is due to better dispersion and interfacial adhesion. After treatment, impurities such as oil and wax in PKS were removed. Thus, more reaction sites available for bridging to take place between PKS filler and UP matrix. Silane

coupling agent promotes the compatibility between filler and matrix resulting higher strengths of composites [4, 11] and also improves dispersibility of fibers [4].

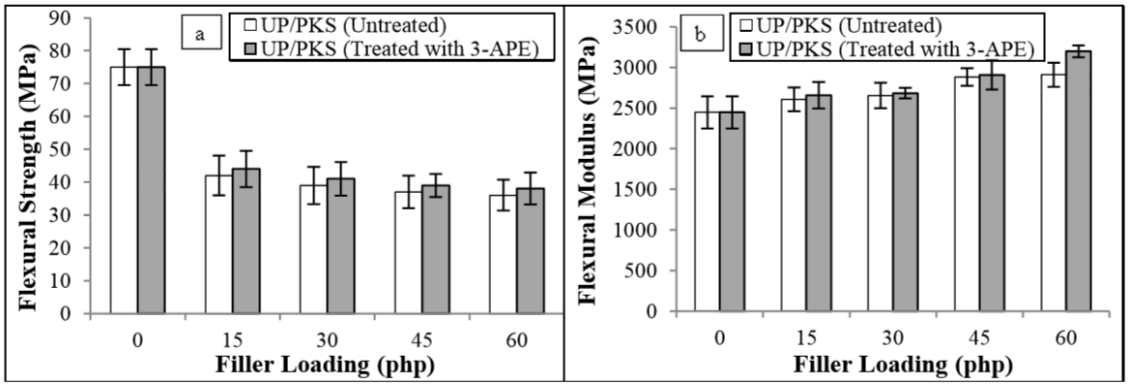

Figure 2: (a) Flexural strength (b) Flexural modulus of untreated and treated UP/PKS composites as a function of filler loading

All of the composites in Figure 2b show increase flexural modulus as increase filler loading. It can be seen that the flexural modulus for chemically-modified UP/PKS composites is higher than the composites obtained without the chemical modification. This result indicates that the presence of 3- APE has shown a significant improvement in the filler-matrix interfacial bonding and better dispersion of filler in polymer matrix. At higher filler loading, the sharing of stress transfer between the filler and matrix is good as well. Therefore, PKS impart stiffness to composites system, thus the flexural modulus is increased by increasing the filler loading.

SEM.

Figure 3 shows SEM micrographs of tensile fractured surface of untreated and treated UP/PKS composites at different PKS loading. Figure 3a indicates the homogeneous surface of unsaturated polyester. Figure 3b shows a quite rough surface of UP/PKS composite at 30 php filler loading indicate the insufficient bonding and compatibility between UP and PKS filler and proposed less adhesion occurred between them. The incorporation of filler loading at 60 php (Figure 3c) indicate that there are voids and bubbles exist, which properties are inferior due to poor interfacial adhesion between the filler and the matrix. The potential of agglomeration to exist at the higher filler loading is also high. This agglomeration cannot withstand high stress and therefore, as the applied stress increased at higher filler loading, the efficiency of stress transfer from the matrix to the filler is reduce, leading to the failure. On the other hand, the SEM micrograph of treated UP/PKS composites at 30 and 60 php (Figure 3d and 3e) show that the surface of PKS in the composites with 3-APE were coated with UP matrix and smooth fracture seems to be obtained. Additionally, voids and cavities did not exist. These contribute to higher mechanical properties after silane treatment which could be attributed to the improvement in the interfacial adhesion between the fibers and matrix.

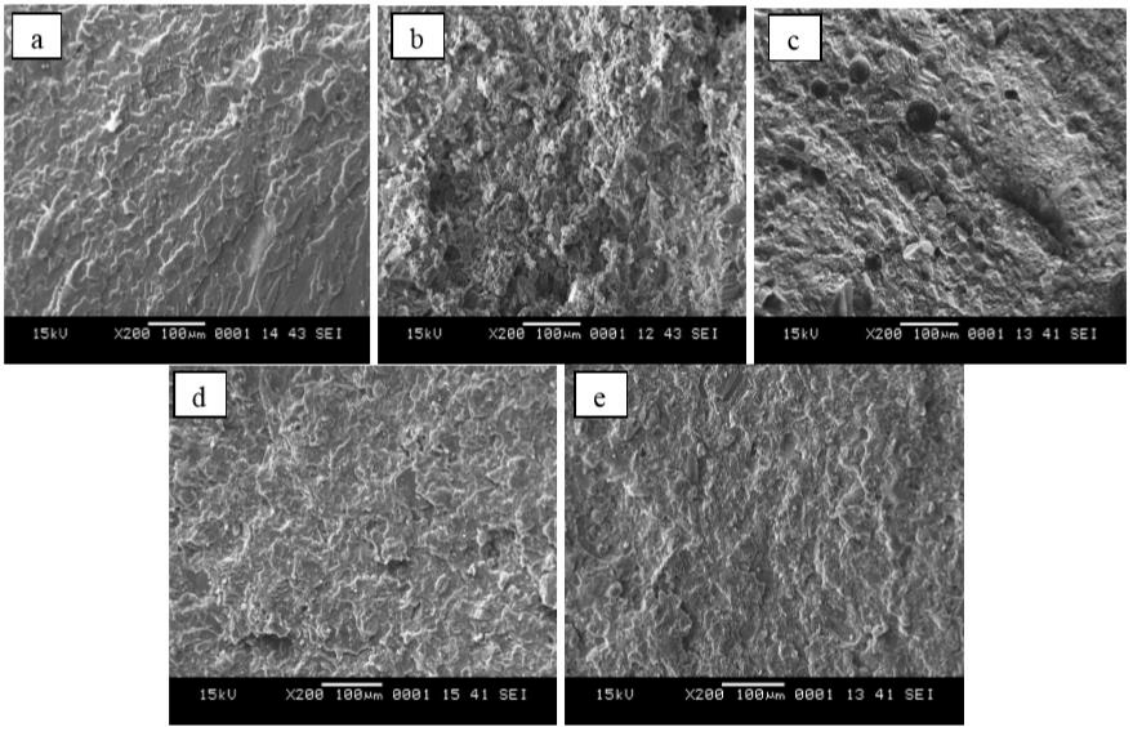

Figure 3: SEM micrograph of tensile fracture surface of (a) UP (b) untreated UP/PKS composites (30 php of filler) (c) untreated UP/PKS composites (60 php of filler) (d) treated UP/PKS composites with 3-APE (30 php of filler) (e) treated UP/PKS composites with 3-APE (60 php of filler)

FT-IR.

Figure 4 show the FTIR comparison of untreated and treated of PKS with 3-APE. For untreated PKS, the peak at 2897.09 cm^{-1} is attributed to methyl group of PKS filler. Palm kernel shell is natural filler mostly composed of cellulose, hemicelluloses, lignins and some pectins. The absorption bands appear at 1045.63 cm^{-1} show the presence of C-OH of the cellulose in PKS. According to Bessadok *et al.*, [12] the C-OH of the cellulose backbone (C-O secondary and C-O primary alcohols) corresponded to the 1056 cm^{-1} and 1030 cm^{-1} peak, respectively.

The treated PKS with 3-APE has lead to the presence of two new absorption bands at 1073.42 cm^{-1} and 1088.61 cm^{-1}. These bands correspond to the C-O stretching which exist due to silane reaction with hydroxyl group of PKS. The peak located at 918.99cm^{-1} indicates the Si-O-C deformation of the silane group. Si-O-C bonds facilitate an enhancement of interfacial adhesion of treated fibers and polymer matrices and of the properties of the resulting composites [13]. The schematic diagram for interaction between filler surface of PKS and silane coupling agent is shown in Figure 5.

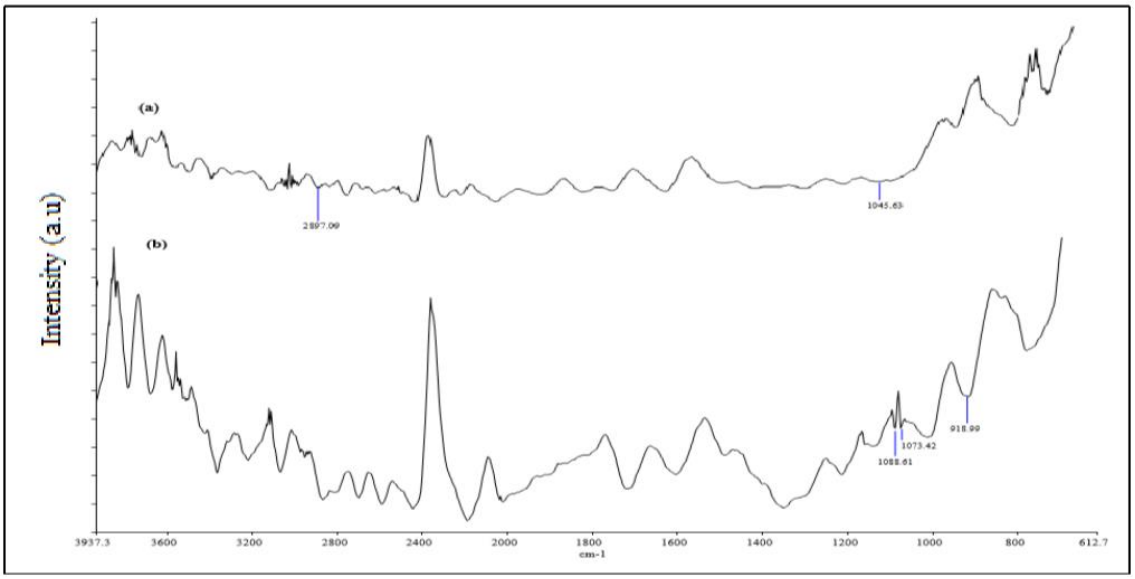

Figure 4: IR spectra for (a) untreated PKS and (b) treated PKS with 3-APE

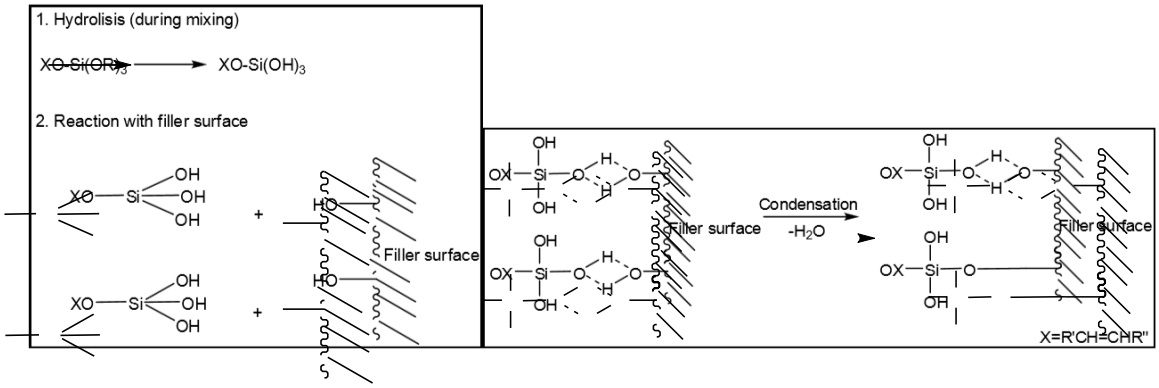

Figure 5: Schematic diagram for interaction between filler surface of PKS and silane coupling agent

CONCLUSION

The incorporation of PKS has increased the tensile modulus and flexural modulus, but a decreased the tensile strength and flexural strength of the composites with increasing filler loading. The presence of a coupling agent, 3-APE has improved the filler-matrix adhesion and consequently improved the tensile modulus and flexural modulus, but lower the tensile strength and flexural strength of the composites as filler loading increased. At the same filler loading, the tensile and flexural properties were higher for composites treated with 3-APE. This was due to the better wettability, dispersion and adhesion between the PKS filler and UP matrix. Micrograph SEM clearly shows that better interfacial interaction between PKS and UP matrix after treatment. The appearance of new peak in FTIR spectra when PKS was treated with 3-APE showed that the silane creating chemical bonding at the PKS/matrix interface.

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