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Mechanical Properties of Unsaturated Polyester Composites Reinforced with Acid-Treated Palm Kernel Shell at Various Filler Loadings

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ABSTRACT

An attempt has been successfully made to utilize palm kernel shell waste as a potential filler in unsaturated polyester composites. A problem with the hydrophilic characteristics of palm kernel shell, which restricts its compatibility and interfacial bonding with polyester matrix, was overcome by chemical treatment with acrylic acid. The composites were prepared via the hand lay-up technique with different amounts of filler loadings (0, 15, 30, 45, and 60 php). Esterification of cellulosic palm kernel shell fiber by acrylic acid has increased the surface roughness of the fibers which tended to improve the mechanical interlocking between the fiber and matrix interphase. The effects of acid treatment and filler loading on the mechanical and morphological properties of composites were studied using mechanical and morphological tests. The addition of palm kernel shell showed a decrement in strength compared with pristine samples due to poor interaction and compatibility of constituent materials. The modulus for tensile and flexural was gradually increased from 2.4 GPa to 2.9 GPa and 2.8 GPa to 3.5 GPa, respectively, through an increase in filler loadings. These results reflect increased stiffness of the composites imparted by the stiff filler. Nevertheless, 15 php of palm kernel shell treated with acrylic acid yielded improved tensile and flexural strength at 32 MPa and 49 MPa, respectively. This result is due to the increased interfacial adhesion between filler and matrix, as exhibited in the Scanning Electron

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Microscope micrograph, which showed better interfacial interaction within the composites.

Keywords: Acid Treatment; Palm Kernel Shell; Composite; Mechanical Properties

INTRODUCTION

Agro-industries produce a large amount of waste, with 998 million tonnes produced globally each year, and Malaysia disposing of 1.2 million tonnes into landfills each year [1]. Oil palm is one of the primary agricultural commodities cultivated in Malaysia. Several biomass wastes are abundantly produced from the palm oil industry, e.g., empty fruit bunch, mesocarp fiber, and palm kernel shell. These biomass wastes can be turned into value-added products, e.g., methane gas, bioplastic, organic acids, bio-compost, plywood, activated carbon, and animal feedstock [2]. The utilization of these sustainable sources of materials promotes eco-friendly and green environmental conditions for humans, as well as impart economic and social benefits to society.

These biomass waste materials have the potential to be used as a filler in the development of polymer composites. The attention of researchers has been diverted towards the usage of natural fibers, due to diversification in the usage of raw materials to replace synthetic materials [3]. Environmental pollution generated during the production of synthetic materials and the depletion of petroleum sources that cause a rise of the price of the petroleum based-products contributes to the development of bio-degradable polymer composites [3,4]. Natural fiber composites possess several advantages, e.g., light weight, renewable in nature, small cost, as well as a high modulus and specific strength [4]. Phiri *et al.* [5] presented a comprehensive review of the deployment of biomass from agricultural waste in the development of sustainable biopolymer-based composites.

Palm kernel shell (PKS) is a lignocellulosic material which consists of 20.8% cellulose, 22.7% hemicellulose, 50.7% lignin, 4.8 % extractives, and 1.0 % ash [6]. Baffour-Awuah *et al.* [7] published a comprehensive analysis on the characteristics of PKS and PKS polymer composites. PKS could be used as reinforcement in polymer composites [8]. This material is effectively used when improving its compatibility with a polymer matrix by diminishing its hydrophilic characteristics by chemical treatment. Thus, the interfacial adhesion between the composite materials will be increased, which will result in better mechanical properties of the composite. In addition, chemical modification of the filler surface helps improve compatibility by dispersing filler throughout the matrix more effectively [4]. Chemical treatments of the fibers aim to improve the adhesion between the fiber surface and the polymer matrix by changing the crystalline structure of the cell wall, increasing surface roughness, and eliminating hydrophilic components, e.g., hemicellulose and lignin [9]. Hence, the surface modification of natural filler needs to be performed before it can be used as a filler in polymer matrices.

Multiple studies have used various fibers and polymer matrices to form composites, including the use of a silane coupling agent [4,10,11], acetic anhydride and maleic anhydride [12], sodium hydroxide [13-16]. Recent studies used modified PKS/particulate cassava peel (PCP) hybrid-reinforced epoxy composites, and it was discovered that chemical treatment significantly improves the mechanical properties of these composites compared to untreated samples [3]. Despite these advancements, there remains a need for alternative chemical treatments that can enhance fiber-matrix interaction while maintaining fiber integrity and minimizing environmental concerns.

In this study, acrylic acid (AA) was employed as a chemical treatment to investigate its effect on the mechanical properties of unsaturated polyester (UP)/PKS composites. This modification is expected to enhance the composite's overall mechanical performance while minimizing excessive fiber degradation. This study also evaluates the potential for using PKS waste as a filler in polymer composites, as well as if oil palm biomass waste benefits the composite in addition to lowering its cost.

EXPERIMENTAL METHODOLOGY

Materials

The palm kernel shells (PKS) were obtained from Malpom Industries Sdn. Bhd. (Penang, Malaysia). Unsaturated polyester resin (Reversol P 9509 commercial grade) was obtained Echomo Trading Sdn. Bhd. (Penang, Malaysia). As for glacial grade acrylic acid and methyl ethyl ketone peroxide (MEKP) catalyst, it was supplied by Zarm Scientific & Supplies Sdn. Bhd. (Penang, Malaysia).

Preparation of Palm Kernel Shells (PKS)

Random sized PKS obtained from Malpom Industries were manually cleaned and dried at a temperature of 80 °C for 24 hours. Then, they were crushed and ground into a powder. After that, the PKS powder was sieved by using a sieve machine and the particle size was analyzed using Malvern Particle Size Analyser Instrument. The average particle size of the PKS obtained is 63 μm.

Acid Treatment of Palm Kernel Shells (PKS)

The PKS were esterified using 3% acrylic acid in an ethanol solution. The powdered PKS was poured into a chamber mixer (model IKA RW20 digital) and the solution was added slowly to ensure the uniform distribution of the acrylic acid. After that, the filler was continuously mixed for 1 hour. After filtering and cleaning with distilled water, the PKS was dried for 24 hours at 80 °C.

Composite Preparation

The UP/PKS composite was prepared via the hand lay-up technique. First, the PKS was added into the polymer resin, which was manually stirred at room temperature. After homogeneous dispersion was observed, 2 php of MEKP catalyst was added. Then, the mixture was poured into a mold and allowed to cure for 10 minutes at 90 °C in an oven. The amount of PKS added for both the untreated and treated composites prepared in this study were 0, 15, 30, 45 and 60 parts per hundred polymers (php).

Characterizations

Fourier Transform Infrared Spectroscopy (FTIR) Analysis

FTIR analysis of untreated and treated PKS was carried out in a Perkin Elmer Spectrometer 2000 FT-IR. The PKS filler was dispersed in dry KBr powder and mixed to obtain fine particles. The KBr pellet technique was applied with a scanned range between 400 and 4000 cm^{-1}.

Mechanical Testing

Tensile test was carried out according to ASTM standard D 638 using a universal testing machine (Instron model 5569). Samples from the rectangular bar specimens were cut, using a composite cutter machine, with the following dimensions: 3 mm thickness, 15 mm width, and 150 mm length. The gauge length of the sample was set at 100 mm and the crosshead speed was 5 mm/min, which was conducted at a temperature of 25 °C ± 3 °C (laboratory temperature). The tensile properties for 5 identical samples of each composition were measured and the average values were reported.

The flexural test was carried out according to ASTM standard D 790 using a universal testing machine (Instron model 5569). A sample of the rectangular bar specimen with the same dimensions as the tensile test samples were used. The gauge length was set at 50 mm and the test was performed at a temperature of 25 °C ± 3 °C at a speed of 5 mm/min.

Morphology Study

A morphology study of the tensile fractured surfaces for the 30 php and 60 php PKS UP/PKS composites were carried out using a scanning electron microscopy (JOEL JSM-6460LA). The fractured surface specimens were mounted on an aluminum stabs and sputter coated with a thin layer of palladium to avoid electrostatic charging during examination.

RESULTS AND DISCUSSION

FTIR Analysis

The FTIR spectrum of untreated PKS is depicted in Figure 1(a).

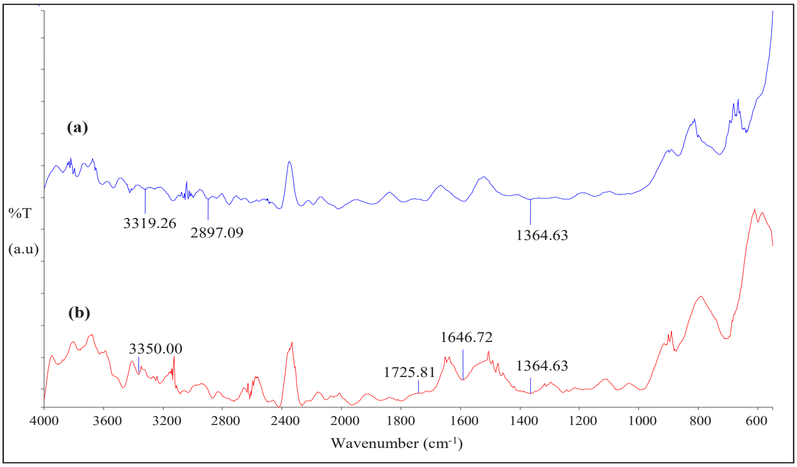

Figure 1: FTIR spectra of (a) untreated PKS and (b) treated PKS.

The spectrum shows a peak at 3319.26 cm^{-1} which corresponds to O-H stretching of moisture or cellulose. In comparison with PKS treated with acrylic acid in Figure 1(b), the O-H peak is shifted to 3350 cm^{-1}. This peak is more intense which shows that there is addition of O-H group from acrylic acid. Both untreated and treated PKS show peaks at 2897.09 cm^{-1} and at 1364.63 cm^{-1}. These two peaks can be assigned to C-H stretching of methyl group of cellulose and the C-H deformation of hemicellulose and cellulose, respectively [4,17]. Peak at 1646.72 cm^{-1} was also observed for both untreated and treated PKS. This peak corresponds to the C=C aromatic lignin for the untreated PKS and overlaps with C=C from acrylic acid for the treated PKS. Additionally, the treated PKS shows a peak at 1725.81 cm^{-1} indicates the C=O stretching vibration of RCOOR crosslinks between cellulose and lignin or cellulose and hemicelluloses [18]. This suggests ester formation in the treated PKS due to the reaction of hydroxyl group in PKS with acrylic acid. Acrylic acid is a type of carboxylic acid that can react with hydroxyl groups present in the cellulose and lignin of

PKS. This esterification process reduced the hydrophilicity of the fiber, thus increased the moisture resistance [19] as well as increased the surface roughness of fiber resulting in better mechanical interlocking which enhances the amount of cellulose exposed on the fiber surface [15]. The mechanism of the reaction of AA with fiber has been reported by Sahoo *et al.* [15] and presented in Eq. (1).

$$\text{Fiber}-\text{OH} + \text{HO}-\text{CO}-\text{CH}=\text{CH}_2 \longrightarrow \text{Fiber}-\text{O}-\text{CO}-\text{CH}=\text{CH}_2 + \text{H}_2\text{O} \qquad (1)$$

Tensile Properties

Figure 2(a) depicts the effects of the filler loading amount on the tensile strength of the untreated and acrylic acid treated UP/PKS composites.

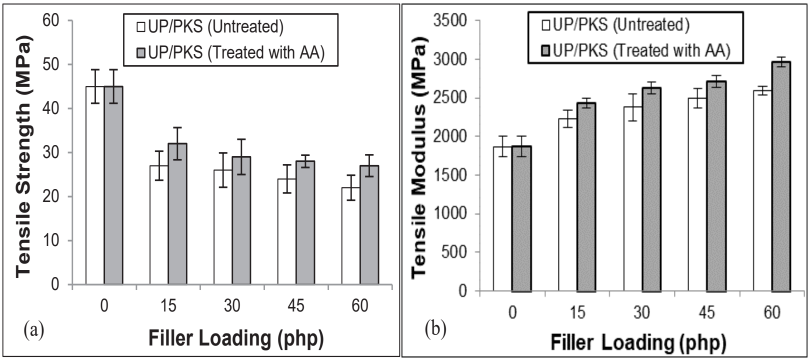

Figure 2: The effects of the acrylic acid treatment and filler loading amount on the (a) tensile strength and (b) tensile modulus of the untreated and treated UP/PKS composites.

The results indicated that the tensile strength of both composites decreased as the filler loading amount increased. This was due to the poor wettability and dispersion of the PKS in the UP matrix. However, at similar filler loading amounts, the treated composites had higher tensile strength than the untreated UP/PKS composites, with values of 32, 29, 28, and 27 MPa for 15, 30, 45, and 60 php of PKS, respectively. This was because there were strong interactions and increased interfacial adhesion between the PKS and UP after chemical modification, as supported by Sahoo *et al.* [20]. Acid treatment has roughened the surface of the PKS, attributed to the removal of hemicellulose and external surface impurities present in the fiber [6].

Hence, the mechanical interlocking between the fiber and matrix interphase increased, which tended to provide better stress transmission during the deformation process. In 2019, Sahoo's study on acrylic acid-treated rattan fibers discovered that esterification increases the surface roughness of the fiber, which leads to improved mechanical interlocking [15]. Moreover, after the acid treatment, there was more available surface area on the PKS that could contact and interact with the UP resin. This agreed with the study by Kommula *et al.* [21], who stated that the acid-treated fiber strands had cleaner and rougher surfaces, which facilitated both mechanical interlocking as well as increased effective surface availability for wetting by the resin.

Figure 2(b) revealed the trend of the tensile modulus for the UP/PKS composites. The tensile modulus for both the untreated and treated UP/PKS composites increased as the filler loading amount increased, which indicated that the stiffness of all samples increased as the filler loading amount increased. This was due to the presence of more rigid fillers in the composite system. Nevertheless, it can be clearly seen that at similar filler loading amounts, the treated composites exhibited a higher tensile modulus than the untreated composites. The presence of acrylic acid (AA) in the polymer composites can enhance the ability of the UP/PKS composite to resist deformation, due to improvements in adhesion across the interface of the material. The surface of the PKS was roughened by the AA due to the removal of hemicellulose, thus improving the wettability of the PKS filler by UP resin, leading to improved compatibility or interaction, as well as better bonding, between the PKS and the UP. Thus, the stiffness of the composite sample was increased. The increased tensile modulus was also attributed to the tendency of the fibers to become tightly packed, which was a result of the removal of the hemicelluloses by the acetic acid treatment [21]. In addition, Nayak & Mohanty [14] asserted that the stiffness of acrylic acid treated fiber is increased due to the esterification of cellulosic fiber. This process leads to an increase in the surface roughness of the fiber which enhances the amount of cellulose exposed on the fiber surface as well as in the reaction sites. A comparable trend of finding has been observed by Jena *et al*. [16] while employing vetiver fiber.

Flexural Properties

Figure 3(a) demonstrated the results of the flexural strength of the untreated and AA treated UP/PKS composites.

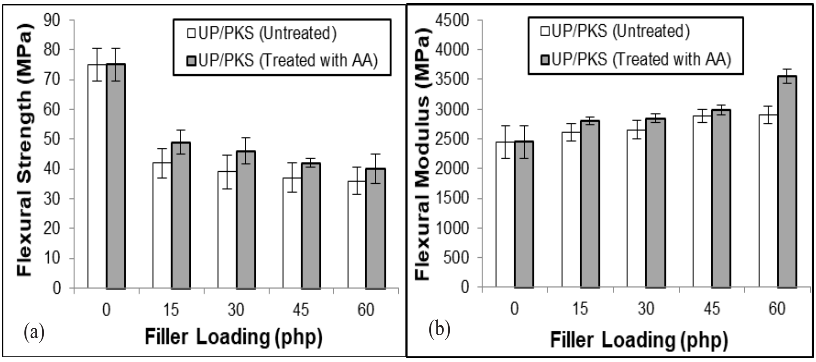

Figure 3: The effect of the acrylic acid treatment and the filler loading amount on the (a) flexural strength and (b) flexural modulus of the untreated and treated UP/PKS composites.

The flexural strength gradually decreased for both the untreated and AA treated composites, which was again due to the poor bonding and interaction between the PKS and the UP matrix. Furthermore, by increasing the PKS loading amount, this meant that there was a reduction in the amount of UP matrix inside the system, which contributed to a decrease in the flexural strength of the UP/PKS composites with data of 49, 46, 42, and 40 MPa at 15, 30, 45, and 60 php of PKS, respectively. However, the results showed that the AA treated composites had a higher flexural strength than the untreated composites. The presence of AA is the primary factor that resulted in the higher flexural strength of the treated UP/PKS composites when compared to the untreated PP/PKS composites. Samanth & Subrahmanya Bhat [22] mentioned that AA was used to modify the fiber surface, which eventually resulted in increased flexural strength due to improved stress propagation under mechanical loading. This observation also agreed with the increased tensile strength as discussed above. The composites can withstand higher loads during the flexural testing bending mode because the stress is better transferred from the fiber to the matrix due to the increased interfacial adhesion [20] of the PKS and UP matrix interphase.

The effects of the acid treatment and the PKS loading amount on the flexural modulus of the untreated and AA treated UP/PKS composites are presented in Figure 3(b). The flexural modulus had a similar trend to the tensile modulus results, which was discussed above. This finding was parallel to and supported by the increase in tensile modulus, as the value of the flexural modulus is equal to the tensile modulus. An increase in the tensile modulus tends to increase the flexural modulus of a material, as it is the measure of the material's stiffness. At similar filler loading amounts, the AA treated UP/PKS composites exhibited a higher flexural modulus compared to the untreated composites. Again, from this result, it can be concluded that there was increased interaction and compatibility between the filler and the matrix. This was due to the effects of the acid treatment that roughened the PKS filler surface, along with the removal of certain hemicelluloses and external surface impurities present in the fiber, which contributed to a higher strength and modulus of the material [6,21]. The treated UP/PKS composite samples had the ability to resist the higher deformation under an applied load due to the good adhesion of the filler-matrix interphase. In addition, there was also greater dispersion of the treated PKS filler throughout the UP matrix, which lead to this observation.

Morphology Study

Figure 4 displays the scanning electron micrographs (SEM) of the tensile fractured surfaces of the untreated and AA treated UP/PKS composites at different PKS loading amounts. These micrographs clearly illustrate the distinct differences in surface morphology of the UP/PKS composites before and after acid treatment. Figure 4(a) reveals a uniform surface of the unsaturated polyester. In the pristine sample, which is without the presence of filler, the stress is evenly transferred as there are no interruptions and barriers during the deformation process. The SEM micrograph illustrated in Figure 4(b) shows a rough surface of the UP/PKS composite after PKS incorporation, indicating inadequate bonding and compatibility between the UP and PKS filler. Increased PKS filler to 60 php, as seen in Figure 4(c) causes voids and bubbles in the composite sample, indicating poorer mechanical properties due to weak interfacial adhesion between the filler and matrix. Abdul Wahab & Husseinshah's [4] previous studies align with this finding. The risk for agglomeration and inadequate filler dispersion increases with increasing PKS loading due to the diminished quantity of UP matrix in the composite system, which impairs the effectiveness of stress transfer from the matrix to the filler, leading to failure.

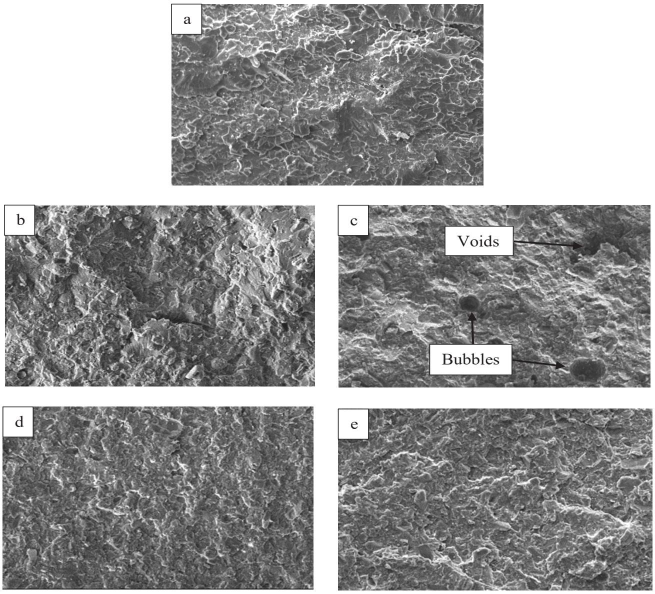

Figure 4: SEM micrographs of the tensile fractured surface of the (a) UP (b) untreated UP/PKS composites (30 php of filler) (c) untreated UP/PKS composite (60 php of filler) (d) AA treated UP/PKS composites (30 php of filler) (e) AA treated UP/PKS of composites (60 php of filler) at 200X magnification.

The SEM micrographs of the treated UP/PKS composites at 30 php and 60 php filler loading amounts, as illustrated in Figures 4(d) and 4(e), revealed a smooth and homogenous surface. This is attributed to improved bonding at the filler-matrix interface and increased wettability of the PKS by the UP matrix because of the filler's roughened surface after chemical treatment that removed hemicelluloses and impurities. The matrix penetrated the porous filler surface, leading to stronger adhesion between the filler and the matrix compared to the untreated UP/PKS filler of the same composition. Hence, the matrix can efficiently transfer applied stress to the filler, resulting in improved mechanical properties. The observed morphologies corresponded with the mechanical properties findings that were obtained in this study.

CONCLUSION

The tensile strength and flexural strength of the composites were reduced as the amount of PKS filler increased in the UP matrix because of poor interaction and compatibility. However, the tensile modulus and flexural modulus increased as the amount of PKS in the UP matrix increased, which indicated that PKS could increase the stiffness of the composites. After chemical treatment, the mechanical properties of the composites were enhanced, which was attributed to better wettability and interfacial adhesion between the composite materials along with increased dispersion of the PKS filler throughout the UP matrix. The SEM micrographs clearly show better interfacial interaction between the PKS and UP matrix after chemical treatment. Thus, the treated PKS have the potential to be used as a filler for the development of polymer composites.

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