Effect of Graphene Nanoplatelets on Water Absorption Properties of Coconut Shell Reinforced Unsaturated Polyester Composites

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Abstract: Coconut shell (CS) reinforced unsaturated polyester (UPE) composites have been prepared by using hand lay-up and compression molding techniques. To improve fiber-matrix adhesion, the CS was chemically treated by two chemical treatments, which are alkali (NaOH) and alkali-silane with concentration NaOH (6%) and silane (2%). To enhance the performance of CS-UPE composites, graphene nano platelets (GNP) was also added as nanofiller. The water absorption tests were conducted to characterize the physical properties of the composites. The result shows that water absorption increases with the increasing GNP. The more adding of GNP fillers' weight percentages, the higher the water absorption will be getting.

1. Introduction
A composite is a combination of two or more materials and not soluble in each other. One constituent is called the reinforcing phase in the form of fibers, particles, or flakes [1]. In modern engineering applications, polymeric composites based on organic fibers have been widely used.

A unique combination of mechanical, technological, and service properties was high strength and lightweight [2]. The fact that polymeric composites were developed with plastic production. Fibers are the manufacturing phase responsible for the material's strength and rigidity as a whole [3].

In recent years, much agricultural waste such as coconut shell (CS) has been abundantly unused in our country. It is an excellent opportunity to utilize CS as fibers reinforcement for polymer composite and can be a perfect opportunity for developing new bio-based products [4]. CS also has many properties like lightweight material; existing polymers are mainly blended with different materials to cost production and have specific applications [5].

Next, polyester was chosen in this research due to the ease of processing. It has led to numerous polymers' applications, most notably as matrix materials for fiber reinforcement laminated composites.

Besides that, graphene has been a hot topic in this material and chemistry field. Usually, it has been used with electronic and biomedical applications [6]. The graphene application has been increased day by day because of its properties like high electrical conductivity, stronger than steel, thin and lightweight, high thermal conductivity, and very high transparency [7,8].

Natural fibers work well as reinforcement in polymers. However, the main weakness of the application of natural fibers is their susceptibility to moisture. This problem occurred as they are hydrophilic [9]. Moreover, the fiber and matrix's chemical structure is different and makes it more challenging to produce high-performance composite materials. The mechanical and physical properties...
of natural fiber reinforced polymer can be improved by various chemical treatments, i.e., NaOH or NaOH-silane.

The combination of CS and UPE matrix still low in terms of mechanical and physical properties. It would be attractive to observe the presence of graphene in CS/UPE composites since it was well known in improving the mechanical and physical properties of composite material. Therefore, the effect of graphene nano platelets (GNP) on water absorption properties of CS reinforced UPE composites will be investigated in this study.

2. Experimental Method

2.1. Materials

The coconut shell (CS) was collected from local plantation sources located at Air Lanas Jeli. All measurements according to the size of the CS were prepared. The matrix resin system used in this work included unsaturated polyester (UPE) resin grade Reversol P9565, methyl ethyl ketone peroxide (MEKP) as a catalyst, and cobalt naphthenate as an initiator, which was purchased from Re vertex Sdn. Bhd respectively. GNP as nano reinforcing filler was supplied by Empayar Abadi Resources Sdn. Bhd. This study's chemical treatments are sodium hydroxide (NaOH) and silane treatments provided by Sigma-Aldrich.

2.2. Chemical Treatment of Coconut Shell Fibers

Chemical treatment on CS fibers was conducted by alkali (NaOH) and alkali-silane. For alkali treatment, the CS fibers was immersed for 3 hours in 6% alkali (NaOH) solution at room temperature. For alkali-silane chemical treatment, the CS was soaked in NaOH solution for 2 hours. Then, it was poured with silane solution prepared by 2% methacryloxypropyl trimethoxysilane diluted in 95% ethanol solution. Then, the CS fibers treated by both chemical treatment methods were washed several times using fresh tap water until neutral pH to ensure complete removal of the NaOH. Finally, the treated fibers were dried in an oven at 80°C for 2 h. The composition of chemical treatment is shown in Table 1.

| Sample Design | Treatment (%) |
|---------------|--------------|
|               | CS | NaOH | Silane |
| NaOH          | 30 | 6    | -     |
| NaOH-Silane   | 30 | 6    | 2     |

2.3. Preparation of CS-UPE Composites

The composite sample was prepared by mixing the untreated and treated CS with UPE as polymer resin and MEKP/cobalt naphthenate as catalyst and initiator. The fabrication process was conducted by compression molding at 120°C for 10 minutes and 5 minutes for cooling at room temperature. The formulations used in this study are shown in Table 2.
Table 2. Preparation of untreated and treated CS-UPE composites with chemical treatment adding by graphene.

| Sample Design | Composition (%) |
|---------------|-----------------|
|               | CS  | UPE  | NaOH | Silane | GNP  |
| UPE           | -   | 100  | -    | -      | -    |
| CS-UPE        | 30  | 70   | -    | -      | -    |
| UCN*          | 30  | 70   | 6    | -      | -    |
| UCNS          | 30  | 70   | 6    | 2      | -    |

-ADDED GNP-

|           | CS  | UPE  | NaOH | Silane | GNP |
|-----------|-----|------|------|--------|-----|
| UCNG1     | 30  | 69.5 | 6    | -      | 0.5 |
| UCNG2     | 30  | 69   | 6    | -      | 1.0 |
| UCNG3     | 30  | 68.5 | 6    | -      | 1.5 |

*UPE+MEKP 2%+Cobalt Naphthanate 1%*

Next, the samples were tested for physical properties. The samples of composites were dried in the oven for 24 hours at 70°C. After that, the composites were immersed in distilled water at room temperature. Then the water absorption of samples determined by weighing at regular intervals. For determining the percentage of water content $(W_t)$, the following equation was used.

$$\% W_t = \frac{W_f - W_d}{W_d} \times 100$$

$%W_t$ = Water absorption, $W_f$ = Weight of sample after, $W_d$ = Initial weight of sample

3. Results and Discussion

Figure 1 shows the water absorption rate on a daily basis for 7 days. All composites have the same readings or pattern of water absorption. The initial water uptake is rapidly increased, followed by a gradual increase until the water balance has been reached. Even the potential for water absorption appears to be controlled as well as the developed composites, but the observation has been made that the composites absorb less water than expected.

From Figure 1, it can be seen that the highest water absorption was shown by CS reinforced UPE composites (CS-UPE). This can be expected since CS fibers and their hydroxyl group (OH-) are strongly hydrophilic as their structure, responsible for high absorption of water than other composites [10,11]. Absorption of the composite material’s water behavior depends on the series of interfacial bonding between the interaction of the fiber and the matrix. The capacity is relatively high whether it is bare or wrapped in composite matrix resin due to the use of fibers.

According to Thaker et al. [12], CS fibers’ hydrophilic nature leads to the formation of hydrogen bonds between water molecules and fillers of (OH-) groups. Due to treatment reasons, the number of OH-groups was reduced, and the treated filler’s treatment showed a high moisturizing absorption compared to the untreated one.

The absorption capacity of the water has been calculated. Figure 1 shows that the percentage of UCN composites increased by more than UCNS. After alkali treatment of CS fibers, water absorption increases from 0.09 % to 0.18 %. This is because the alkali solution is affected by hemicellulosic, which results in the breakdown and destruction of hydrogen bonding.

Besides, hemicellulose of ester-linked substances can be broken down by alkali. Polymer solubility and hydrophobicity can be increased [13]. The results show strongly that the surface modification of
CS-UPE composites by alkali treatment has a significant adverse effect on the absorption of water composites and has a good filler matrix interaction and less agglomeration than untreated fibers. For GNP nanofillers incorporated into CS-UPE, the highest water absorption was shown by UCNG3 with graphene fillers at 1.5wt %, (followed by a drop of 1wt % and 0.5wt %). The more graphene fillers added, the higher the absorption of water. According to Zurutuza et al. [14], the interplanar forces can be reduced due to functional groups' presence, and graphene with a hydrophilic character can be imparted. It also enhanced the interfacial interaction between polymers and graphene. However, graphene dispersion will occur in polymer matrices.

![Graph showing water absorption of untreated and treated CS-UPE composites in 7 days.](image)

**Figure. 1.** Water absorption of untreated and treated CS-UPE composites in 7 days.

4. Conclusion

In summary, the best of chemical treatment had set out with choose alkali treatment, UCN composites rather than UCNS composites. For the physical test, the graph shows that UPE has a low absorption of water than any specimen as the presence of a hydrophilic group of CS fibres. The water absorption of the composites was increased by the addition of the filler content of the CS fibres and the time of immersion. For the graphene, the graph shows that the increase in the wt% of graphene fillers can influence the high absorption of water. The more weight percentages of graphene fillers are added, the higher the absorption of water will be.

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