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Water Absorption Behaviour of Epoxy/Acrylated Epoxidized Palm Oil (AEPO) Reinforced Hybrid Kenaf/Glass Fiber Montmorillonite (HMT) Composites

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Abstract. The use of fiber-reinforced vegetable oil - polymer composites has increased in various technical fields. However, the long-term operating performance of these materials is still not well understood, limiting the development of these composites. In this study, the water absorption performance of hybrid composites, which consist of kenaf fiber and glass fiber as reinforcement, epoxy resin and acrylated epoxidized palm oil (AEPO) as a matrix, and montmorillonite (MMT) nano clays as a filler was evaluated with the function of different fibers layering order. The hand lay-up method is used to produce the composites with the variable number of kenaf fibers and glass fibers layer sequences. The water absorption kinetics of epoxy/ AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites are described in this paper. It has been observed that the water absorption rate of the composites depends on the fiber layering sequences. The alternative sequence of Glass-Kenaf-Kenaf-Glass and Kenaf-Glass-Kenaf-Glass composites layers exhibited the lowest moisture absorption rates of 7.61% and 7.63%, respectively.

1 Introduction

Increasing environmental awareness has led to increasing demand for bio-based polymer materials from renewable resources for commercial applications. Bio-based polymer composites derived from vegetable oils are the most interesting bio-based polymers because vegetable oils are available in large quantities worldwide. The global production of most vegetable oils is increasing each year. Vegetable oils are widely used in food and non-food products [1]. Recently, however, there has been a great deal of interest in exploring vegetable oils as a starting material in the development of polymeric composites either by direct synthesis of vegetable oils or by mixing vegetable oils with polymer resin.

Blending vegetable oil (VOs) with petroleum-based polymer resins are the most commonly reported in the literature. Indeed, partial replacement of polymeric resins with vegetable oils could result in materials with acceptable properties suitable for the current market. For instance, many studies have reported blending thermoset resins with vegetable oils [2–4]. In their study, they found that incorporating vegetable oils with polymer resins enhanced the toughness properties and consequently overcame the brittleness of thermoset resin.

Vegetable oils composed of various chemical structures and compositions that allow them to be activated for condensation polymerization with hardening agents [5]. However, vegetable oils in their natural form are not reactive; they need to undergo various modification techniques, such as epoxidation, acrylate, or maleinization, to increase the functionality of vegetable oils to improve their reactivity with the polymer resin [6-7]. Acrylation is a commonly used technique to further modify epoxidized vegetable oils by adding more polymerizable functional groups (such as acrylates and hydroxyls) to increase their reactivity. Some research reports claim that polymer resins containing acrylated epoxidized vegetable oils have better mechanical and thermal properties than polymer resins containing epoxidized vegetable oils [8–10].

Despite excellent mechanical toughness and impact strength properties, the addition of vegetable oil, however, has reduced the strength and modulus of the thermoset/vegetable oil polymer resins. To balance the rigidity and toughness properties of vegetable oil/polymer resins, nanofillers are usually incorporated into the polymer resins. According to previous reports, the well-dispersed nanofillers in the polymer matrix can improve mechanical properties and the barrier properties of the resin [11-12].

In recent years, there has been interested in the development of hybrid fiber composites with two or more reinforcing fibers in a single matrix. Hybridization of the fibers offers a good compromise in terms of mechanical and thermal properties [13]. Nevertheless, one of the limitations of hybrid fiber-reinforced composites is their durability when exposed to moisture humidity which may affect the mechanical performance of the composites. Hence, in this study, the effect of varying kenaf fiber and glass fiber mats sequences on water absorption behavior of epoxy/ acrylated epoxidized palm oil

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(AEPO) reinforced hybrid kenaf/glass fiber-filled montmorillonite (MMT) composites will be investigated. To date, the study on the water absorption behavior of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites has not yet been reported in the literature.

2 Experimental

2.1 Materials

The diglycidyl ethers of bisphenol-A, epoxy resin and Jointmine 905-3S, epoxy hardener were provided by Dow Chemical Pacific Singapore ("Dow"), Singapore. The epoxidized palm oil (EPO) was supplied by Budi Oil Sdn. Bhd., Selangor. The montmorillonite nanoclay (Cloisite 30B) was purchased from Southern Clay Products, Inc. USA. The chopped strand mat (CSM) glass fiber and kenaf fiber are provided by Euro-Chemo Pharma Sdn. Bhd. Bhd, Malaysia and Kenaf Bio Solution Sdn. Bhd, respectively.

2.2 Preparation of Samples

Nanoclay was first dried in a vacuum oven at 120°C for 4 hours [14]. Then, the dried OMMT was sonicated in acetone with a solution concentration of 50 L acetone to 1 kg clay for 30 minutes. The amount of MMT was set at 1.5 phr for all blend formulations. After that, epoxy resin was added, and the blended solution was mixed at a temperature of 80° C for 2 hours using a mechanical stirrer. The epoxy resin blend solution was then placed in a vacuum oven at 70°C for 1 hour to remove acetone. Then 10 wt% of AEPO was added, and the mixture was continuously stirred for 1 hour at 55 ºC on a hot plate. After that, the hardener was added to the epoxy resin mixture. To prepare the composite, chopped mat glass fibers and kenaf fibers were placed on an aluminum mold, and a roller was used to impregnate the resin mixture. The configuration of the laminated hybrid composites are KKKK, KGGK, GKKG and KGKG (K is kenaf fiber, G is glass fiber). The samples were cured in an oven at 25 ºC for 3 hours and then post-cured at 80 ºC for 6 hours [15].

2.3 Testing and characterization

The water absorption test was performed according to ASTM D57081. The sample for the water absorption test was 75 mm in length, 12 mm in width, and 5 mm in thickness. Each edge of the sample was covered with a layer of waterproof paint. Three samples of each composition were used. The samples were first dried in an oven at 70 ° C for 24 hours and then cooled in a desiccator. Percentage absorbability of composites was calculated from the equation (1):

$$Water\ absorption\ (\%) = \frac{W_w - W_d}{W_d}\ x\ 100 \tag{1}$$

where w_w (g) and w_d (g) are the weight of the sample after and before soaking in water, respectively.

2.3.1 Kinetics of Water Absorption

Sample diffusivity was calculated using Equation (2) as given by [16].

$$Diffusivity\ (Dx) = \pi\left(\frac{h}{4Mm}\right)\left(\frac{M2-M1}{\sqrt{t2}-\sqrt{t1}}\right) \tag{2}$$

where h is the thickness of the sample, Mm is the moisture absorption percentage (%) at equilibrium content, and $\left(\frac{M2-M1}{\sqrt{t2}-\sqrt{t1}}\right)$ is the slope of moisture absorption plot for the initial linear portion of the curve $\sqrt{second}$.

In order to study the composite water absorption, the experimental data was compared with the theoretical Fickian water absorption. Equation 3 was used to calculate the theoretical Fickian moisture absorption percentage.

$$M(T,t) = (Mm - Mi)\left(1 - \exp\left[-7.3\left(\frac{Dx.t}{h^2}\right)^{0.75}\right]\right) + Mi \tag{3}$$

3 Results and Discussion

Fig. 1. shows the water absorption curves of epoxy/ acrylated epoxidized palm oil (AEPO) reinforced hybrid kenaf/glass fiber-filled montmorillonite (MMT) composites in distilled water. Each data point represents the average of the four samples. The water absorption of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites steadily increased with increased immersion times until it reached a saturation point, indicating a Fickian mode of diffusion. The equilibrium content at saturation for all composites is depicted in Table 1.

From the graph in Fig. 1., the 4K composite had the highest water absorption of 13.02% compared to KGGK, GKKG, and KGKG hybrid composites. This result was expected due to the hydrophilic nature of kenaf fiber, which caused composites to absorb more water. The hydroxyl groups found in lignin, hemicelluloses, and cellulose formed hydrogen bonds with water molecules, increasing the water absorption process. Similar observations were reported by Ramamoorthy et al. [17], who studied lyocell fiber-reinforced soybean oil thermosets. Their results showed that water absorption was increased by increased lyocell fiber content.

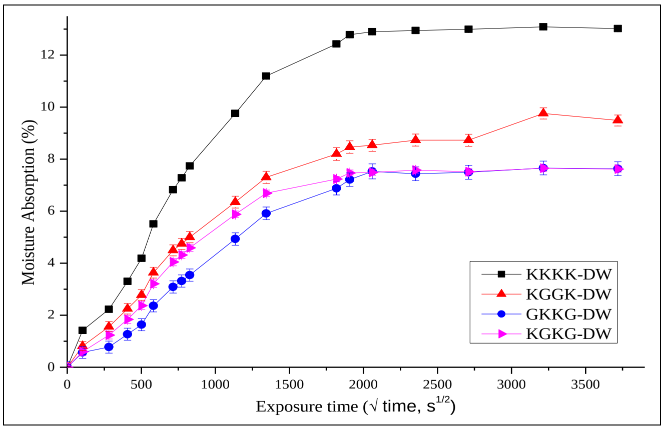

Fig. 1. Water absorption of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites with different layering sequences

The findings also showed that water absorption was reduced in hybrid composites with kenaf and glass fibers. The water absorption of 4K composites decreased from 13.02% to about 9.49%, 7.63%, and 7.61% for KGGK, KGKG, and GKKG composites, respectively. The results show that replacing two hydrophilic kenaf fiber layers with two layers of glass fibers reduced water absorption. The GKKG and KGKG composites showed the lowest water rates of 7.61% and 7.63%, respectively. This shows the efficiency of glass fibers in reducing water absorption. The addition of glass fibers as skin layers (GKKG) acted as a barrier that protected internal fiber layers from direct water contact, drastically reducing water absorption.

Moreover, alternating layers of glass and kenaf fibers (KGKG) made the water penetration and saturation process for each layer become longer than in non-hybrid composites (4K). Overall, the water absorption of hybrid composites was found to be lower than laminated non-hybrid composites (4K). A similar observation was reported by Sanjay and Yogesha [18], who studied the effect of hybridization on the physical and water absorption properties of epoxy resin composites reinforced with jute/kenaf/glass fibers. They found that the incorporation of glass fiber reduced the water absorption of jute/kenaf composites.

The diffusion coefficient rate (D_x) represented the water molecules' ability to penetrate into composites. In this study, the diffusion coefficient rate and the maximum moisture uptake of hybrid and non-hybrid kenaf/glass fiber reinforced epoxy/AEPO filled MMT composites in distilled water were determined. These values are summarized in Table 1. The table shows that non-hybrid kenaf composites (4K) had the highest diffusion rate and greatest moisture uptake of 3.91 x10-6 mm2/s and 13.02%, respectively. The lowest diffusion rate and water absorption were possessed by the GKKG hybrid composite (2.08 x10-6 mm2/s and 7.61%, respectively), followed by the KGKG and KGGK hybrid composites. The decrease in composite diffusivity and water uptake was due to the addition of glass fiber layers. These results reveal that composite diffusivity and water uptake differ significantly depending on fiber type.

Table 1. Moisture absorption properties of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites with different layering sequences

Samples	Maximum Moisture Uptake, Mm (%)	Time to Reach Mm (h)	Diffusion Coefficient, D_x (x10^{-6})
4K	13.0 ± 0.03	10100	3.91
KGGK	9.49 ± 0.21	1537	2.65
GKKG	7.61 ± 0.27	1178	2.08
KGKG	7.63 ± 0.12	1010	2.23

Fig. 2. shows the initial linear slope for the water absorption of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites in distilled water. The diffusion rate (D_x) of 4K, KGGK, GKKG, and KGKG composites were calculated from the linear part of the absorption slope using Equation (2).

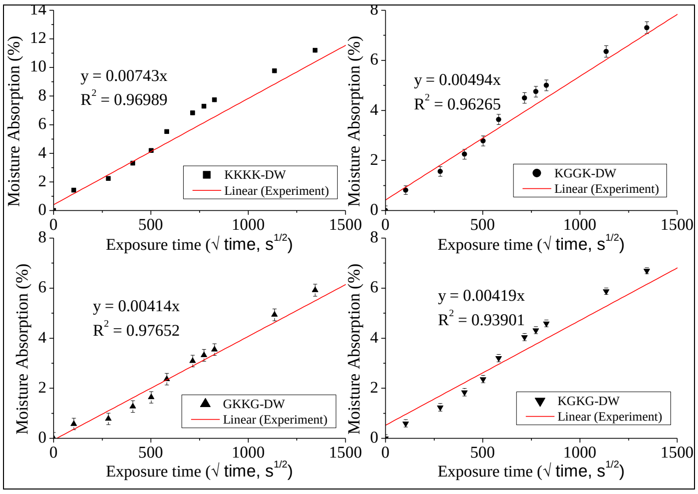

Fig. 2. Initial stage slope for the water absorption of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites with different layering sequences

Figure 3 compares experimental and Fickian theoretical water absorption for the 4K, KGGK, GKKG, and KGKG composites. For all samples, water absorption linearly increased at the initial absorption stage until reaching saturation stage, which no more water can be absorbed by the composites. This proves that all samples followed typical Fickian diffusion behaviors.

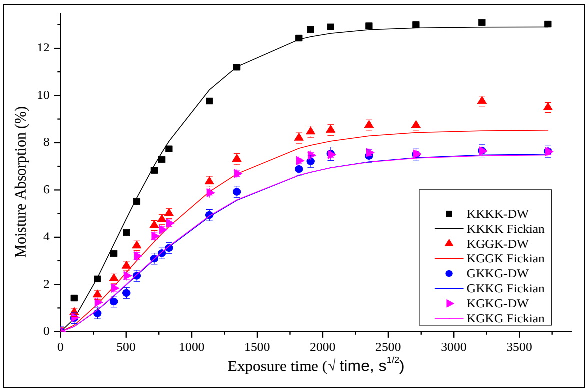

Fig. 3. Comparison between theoretical Fickian and experimental water absorption data for epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites with different layering sequences

4 Conclusion

The water absorption characteristic of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites has been studied. The effect of different fiber layering orders was investigated. The glass fiber and kenaf fiber were used as reinforcement in acrylated epoxidized palm oil (AEPO)/ montmorillonite (MMT)/ epoxy composites. Based on the results from this research work, the water absorption of epoxy/AEPO reinforced hybrid kenaf/glass fiber-filled MMT composites follow the typical Fickian diffusion behavior. The different arrangements of kenaf fiber and glass fiber layering sequences affect the water uptake of the composites. The GKKG and KGKG composites showed the lowest water absorption rates than other layering patterns.

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