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Water absorption analysis on impregnated kenaf with nanosilica for epoxy/kenaf composite

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Abstract. Due to the existence of hydroxyl group from cellulose and hemicellulose in natural fibre, it has a high tendency to absorb significant amount of water. Nanosilica has high surface area due to its nanosize, which is capable to seal the fibre’s pores. In this work, impregnation technique was used to fill the pores of kenaf fibre with nanosilica in order to reduce the water uptake of the resulting composite. To impregnate the randomly orientated kenaf fibres with nanosilica, the fibres were initially degassed in a plastic container to enable the inclusion of nanosilica to fill the fibre pores. The nanosilica/epoxy mixture was infused into the container and left under vacuum condition for 24 hours. The hardener was then applied onto the fibres before being hot pressed to aid the curing process. For the water absorption test, the specimens were dried before being placed in a glass container filled distilled water and regularly weighed until approximately 80 days. Like any other kenaf reinforced epoxy, the specimens absorbed water at a rapid rate in the beginning before slowing down and approaching equilibrium. The maximum water absorption rates were 14.4% and 12.9% for specimens with composition of 50 vol% kenaf, 5 vol% nanosilica and 60 vol% kenaf, 5 vol% nanosilica, respectively. Specimens with nanosilica infused but not impregnated had shown a significantly higher water absorption rates at 24.5% and 29.6% respectively. These results show that nanosilica can help reduce the water absorption rate of the natural fibres by blocking the pores of the fibres rendering them unable to absorb water.

1. Introduction
Kenaf, like many other natural plant fibres, has been studied extensively as a susceptible replacement of synthetic fibres such as glass and carbon fibres. Natural fibre is biodegradable, tough, renewable, low cost and low in density, which contribute to its high specific strength and non-abrasive nature, and making natural fibre easy to be processed while allowing high filling levels [1-3]. However, natural fibres tend to absorb moisture either from the air or from rain water, which will affect the properties of the overall composite. The hygroscopic nature of natural plant fibre is contributed by its microscopic like tubes with cell walls surrounding the central lumen [4]. The lumen contributes to water absorption behaviour due to the transport of water molecules via capillary action, which also involves the pores of the fibres [5]. Furthermore, natural plant fibres structure composition contain high amount of cellulose and hemicellulose, which produce the polar hydroxyl group that makes them hydrophilic in nature [6],
As matrix materials have a tendency of being hydrophobic, hydrophilic natural fibres will repel them and making it difficult to achieve good fibre wettability during production [8].

There are three types of mechanism in which moisture will be absorbed into the composite [9-11] as listed below:

a) Diffusion of the water molecules into the micro bubbles between polymeric chains. The micro bubbles are produced during the mixing and fabrication procedure.

b) Via capillary transport into the gaps and flaws in between the interfaces of fibre and matrix which occurs due to improper fibre wettability and poor fibre-matrix interaction.

c) Micro cracks in the matrix produced due to the swelling of fibres. This mostly happens in the case of natural fibre reinforced composites.

In the case of this work, in addition to the kenaf fibre, nanosilica is introduced into the system and the mixing technique needs to be considered. Basri et al. [9] reported that a high speed, low time mixing of nanosilica into epoxy resin system produced specimens with the lowest water absorption rate. When fabricating composite, the idea behind impregnation process is to fill the empty spaces of the natural fibre (lumen cavity and cell wall) with polymer resin [7, 12]. By blocking the water path into the fibre, it is anticipated that the fibre will reduce its water uptake. In this work, nanosilica is considered as the impregnation agent alongside polymer matrix due to its minuscule size contributing to the material’s high interface area of about 1500 m²/g, having active function and high interfacial interaction with resin [9,13]. Nanosilica is expected to be able to impregnate the fibres’ lumen to reduce the fibre water absorption rate.

2. Methodology

2.1. Materials

Epoxamite 100 with 103 SLOW Hardener that was manufactured by Smooth-On is used as the matrix. Hydrophilic type of nanosilica derived from rice husk ash was provided by Maero Tech Sdn. Bhd. (MTSB). The reinforcing fibre used was randomly orientated kenaf mat provided by ZKK Sdn Bhd. The kenaf mat was cut into sections of 150 mm × 150 mm and weighed to be utilized in the fabrication process. For every specimen, weighted kenaf mats were combined to achieve the desired weight. It should be noted that nanosilica is an addition to the kenaf and epoxy’s loading. Table 1 lists all of the specimens fabricated. In the specimen name, "NI" after the number indicates that the specimen is not impregnated whereas "I" is designated for impregnated specimens.

| Specimen Name | Kenaf Loading [vol%] | Epoxy Loading [vol%] | Nanosilica Loading [vol%] | Impregnated or not |
|---------------|----------------------|----------------------|--------------------------|-------------------|
| 400NI         | 40                   | 60                   | 0                        | No                |
| 500NI         | 50                   | 50                   | 0                        | No                |
| 600NI         | 60                   | 40                   | 0                        | No                |
| 405NI         | 40                   | 60                   | 5                        | No                |
| 505NI         | 50                   | 50                   | 5                        | No                |
| 605NI         | 60                   | 40                   | 5                        | No                |
| 405I          | 40                   | 60                   | 5                        | Yes               |
| 505I          | 50                   | 50                   | 5                        | Yes               |
2.2. Fabrication of non-impregnated specimens
The nanosilica is homogenized with epoxy without the hardener at 3000 rpm for 10 minutes using a homogenizer. For the specimens without nanosilica, this homogenization procedure was skipped. The hardener was then mixed into the solution and applied evenly using a brush onto the cut kenaf mats. The wet kenaf mats were stacked inside a 150 mm × 150 mm × 3 mm mould. The mould was then compressed using a 40 tonne hot press at pre-heated at 85°C for 20 minutes and then subsequently cold pressed for 5 minutes. The specimens were left overnight for curing. All specimens were post cured at 80°C for 2 hours.

2.3. Fabrication of impregnated specimens
The nanosilica is homogenized into approximately 2.5 litres of epoxy without the hardener using a homogenizer at 5000 rpm for 20 minutes. The speed and time of the homogenizing process were faster and longer than non-impregnated specimens due to the amount of matrix and nanosilica used. Inside a bell jar, a container with kenaf mats was placed. The air inside the glass jar was evacuated to an impregnation pressure of 600 mmHg (80 kPa) for 5 minutes using a hydraulic vacuum pump before the epoxy and nanosilica were infused until the kenaf mats were fully submerged. The specimens were left under impregnation pressure of 600 mmHg for another 2 hours before the pump was switched off. The specimens were left overnight before releasing the air into the vessel slowly within 30 seconds. The impregnated kenaf mats were drained off by compressing the fibres. After that, the hardener was applied using a brush and the kenaf mats were placed inside a 150 mm × 150 mm × 3 mm waxed mould before being hot pressed at 85°C for 5 minutes and subsequently cold pressed for 5 minutes. The specimens were left overnight for curing. All specimens were post cured at 80°C for 2 hours.

2.4. Water absorption test
The water absorption test was conducted according to ASTM D570-98. The samples were conditioned into approximately 57 mm × 10 mm. For each loading, 3 replications were made. The samples were dried in an oven at 40°C for 24 hours before being weighed. Then the samples were immersed into distilled water with controlled temperature of 30 ± 1°C. Samples were then weighed subsequently after 3, 6, 24, 48, 72 and 96 hours for the first week. Further weighing was conducted every one to two weeks for approximately two and a half months. The weighing is done by removing the sample from water and wiping excess water with dry cotton cloth before placing the sample onto a measuring device with the accuracy to the nearest 0.001 g. The sample was then replaced back into the water immediately. The distilled water was changed every one to two weeks.

Water intake of each sample was calculated using percentage weight gain of the specimens at the interval measured as shown by Eqn. 1, where $M_i$ is the percentage weight gain, $M_d$ is the initial oven dried mass and $M_t$ is the mass at the interval measured.

$$M_i = \frac{M_t - M_d}{M_d} \times 100$$  \hspace{1cm} (1)

3. Results and discussion
Figure 1 shows water absorption behaviour of nanosilica/kenaf reinforced epoxy (SKRE) and kenaf reinforced epoxy (KRE). For the first day, the KRE absorbed water rapidly at an almost linear rate. This was true for all specimens. Then, the water absorption rate (weight percentage increase rate) slowed down gradually with time. This could be seen until day 9 (15 root hour) for the impregnated specimens and day 25 (24 root hour) for the not impregnated specimens. Further immersions afterward only showed a slight weight increase as the specimens were nearing the equilibrium. Water molecules penetrated the cell walls and occupy the space in fibre lumen, causing the fibre density to increase [7]. In other words, the fibre will swell due to moisture intake. Fibre swelling results in the production of voids in between fibre and matrix interface, furthering penetrability of water and inducing capillary action [14]. Therefore, it can be concluded that rapid water uptake into the composite in the beginning.
is governed by multiple mechanisms: water absorbed by the fibres, voids induced by fibre swelling and existence of micro porosity in the matrix system. When the weight of the specimens are almost constant, it shows that the specimens have less interaction sites with water molecules and is getting saturated with water [9]. All specimens show similar absorption trend like that of Fickian diffusion trend where the water is absorbed sharply and then the absorption rate decreased until the specimens’ weight percentage increase reaches equilibrium.

Table 2 shows the maximum water weight percentage increase for every specimen. From the table, it can be observed that with the increment of kenaf, the maximum water intake is increased too. This finding is in tandem with the knowledge where, the higher content of hydrophilic fibre in a composite, the higher the amount of water will be absorbed. Furthermore, with the exception of 605NI specimen, inclusion of 5 vol% nanosilica has slightly increased the water absorption rate. During homogenizing process, air bubbles may be produced too. The air bubbles consequently are excess spaces for water to occupy inside the composite. In another words, inclusion of nanosilica produces more micro bubbles where water will be diffused into, hence increasing the water absorption rate of a composite [9-11]. Furthermore, improper homogenizing may cause nanosilica to agglomerate, thereafter producing even more voids as the spaces in between nanoparticles are occupied with air instead of the matrix.
However, with the impregnation conditioning, the specimens’ maximum weight increase after the immersion in distilled water was considerably lower than the not impregnated specimens, especially for 605I specimen. There are multiple reasons on the impregnated specimen low water absorption rate such as listed as follow:

a) The absence of micro bubbles. Previously, it is mentioned that homogenizing of nanosilica into epoxy produces more air bubbles. During the impregnation process, the matrix and fibres are subjected to vacuum pressure overnight. Apart from filling the lumen of fibres with nanosilica, this process helps in removing micro bubbles too.

b) The impregnation process succeeds in filling the lumen cavity and cell wall with epoxy and nanosilica. The matrix and nanoparticles that impregnated the fibres’ cell wall and cell lumen acted as a wall that blocked water molecules from diffusing into the fibres or at least reduced the permeability of behaviour of the fibre as the micro pores were blocked due to the matrix existence [7].

c) Good interfacial adhesion between kenaf fibres and epoxy. Poor adhesion between matrix and the lignocellulosic fibres will cause more gaps to appear in between the interfacial region making more hydrogen bonding occur between water and higher abundance of hydroxyl group from the fibres [15]. Epoxy resin used in this work takes 1 hour before the gelling process. This means that when applying epoxy, there is only approximately 1 hour for the matrix to properly wet the fibres. After 1 hour, due to the gelling process, it will be harder for the matrix to create adhesion with the fibres due to the increased viscosity. With impregnation, as the fibres are submerged for overnight inside the resin, there are more time for the matrix to create interfacial adhesion with the fibres.

4. Conclusion
Higher fibre volume content will increase the KRE absorption of more moisteres and the inclusion of nanosilica too will increase the water absorption rate. Impregnation process considerably reduces the water absorption rate. 605I specimen has the lowest water uptake at 13.44% weight increase from the initial submersion. The low water uptake indicates that the nanosilica/epoxy mixture has succeeded in penetrating the cell walls and lumen of the fibre, subsequently blocking water from being absorbed.

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