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Short Communication

Moisture absorption behavior of sugar palm fiber reinforced epoxy composites

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Abstract

In engineering practice, moisture absorption test is generally used for quality control purposes and to measure the degradation of the quality for the composite materials. The objectives of this study are to investigate the value of Fickian diffusivity constant, moisture equilibrium content and correction factor for the natural fiber composites. Tests were carried out on composite plates, which was a combination of sugar palm fiber and epoxy resins and two different fiber compositions have been chosen which were 10% and 20% by weight. Pure epoxy plates have been used for the control measures. The specimens were oven dried for 60 h in an air-circulating oven operated with 108 °C before being immersed in the constant temperature water bath, which the distilled water was set at 40 °C for the moisture absorption behavior test for 33 days. From this study, plates with 20% fiber loading possessed the highest amount of moisture prior to the moisture absorption behavior test, which is 0.93%. In moisture absorption behavior test, the corrected value of Fickian diffusivity constant for the 20% fiber loading is 3.76×10^{-7} mm^2/s, which is the highest among other composites. It is shown that, for composite plates that contain higher fiber composition, the moisture absorption rate is even higher.

1. Introduction

There are many potential natural resources, which Malaysia has in abundance. Most of it comes from the forest and agriculture such as sugar palm *(Arenga pinnata)* fiber or locally called *ijuk* fiber. As seen in today's market, there are loads of products that have been produced using natural fibers. Apart from that, there are feasible resources which contained bio-fiber materials that can be manufactured into composite based on epoxy resins or other matrices.

As well as having to withstand loading extremes, composite components have also to survive in a range of different environments of moisture and temperature. Relative humidity can vary from 0% to 100%. Temperatures for most uses range from −40 °C to 70 °C, although military aircraft have to withstand temperatures up to 130 °C in flight and even greater around engines. The effect of these temperatures on composite properties needs to be known and ideally predicted [1]. Most polymer matrix composites absorb moisture from humid atmospheres. This is usually confined to the resin matrix, but some fibers also absorb moisture. Epoxy resins absorb between 1% and 10% by weight of moisture, so a typical composite with 60% by volume of fibers will absorb around 0.3–3% moisture, but the systems in widespread use tend to absorb in the range 1–2% by weight [1]. It has been agreed by a number of studies done on moisture absorption of composite that the moisture absorption by fibers is negligible thus in this study, weight gained is considered directly from the moisture absorption by the resins.

Composites offer a quite good long term behaviors in various aggressive environments and high strength – stiffness to weight ratios. However, it appears that these reinforced plastics are more or less sensitive to water and moisture through absorption of water molecules, leading first to physical degradations, like plasticization of the

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matrix with water and the differential swelling between fibers and resin or between plies. At longer times, the uptake of water can also induce chemical degradations like hydrolysis of the matrix and the glass sizing, leading to delamination. Consequently, the residual mechanical properties depend on these degradations and for a mean life time of 20 years, the main effects to these materials are the plasticization of the resin (reversible loss of properties) and the differential swelling (cracks appearing at interfaces fiber-resin). So, it appears to be important to focus on this latter phenomenon [2].

This study focuses on the moisture absorption or desorption behavior of the sugar palm fiber reinforced epoxy composites in the through-the-thickness direction for single-phase Fickian solid materials in flat panel form. In term of application, the results are useful in the marine sector for producing fishing boat components or its construction.

2. Experimental procedure

2.1. Materials

The fiber that used was sugar palm (*Arenga Pinnata*) fiber or *ijuk* fiber. This fiber is black/brown in colour, with a diameter of up to 0.50 mm (0.020″). The fiber is stiff and durable. Sugar palm fibers have some advantages over traditional reinforcement fiber materials in terms of cost, density, being renewable, non-toxic, abrasiveness and biodegradability. The matrix used was epoxy resin based on diglycidyl ether of bisphenol A (DGEBA) purchased from Wah Ma Chemical Sdn. Bhd, Malaysia.

2.2. Mold preparation

Few methods have been devoted to prepare the composite plates for this study and none of them produced expected surface morphology of the composites. Among them were hot press molding and manual preparation using mold from aluminum and treated with heat in oven. Those methods produced bad surface morphology even after being supplied with silicon mold release agent and too many voids in the composites, which were undesirable. Since epoxy is in liquid form, very few methods were identified ideal to be applied in order to fabricate the composite plates.

The best method to produce composite plates was to use the heat resistant transparent plastic called Mylar as the base and the top cover for the mold. The mixture of epoxy and hardener causes a chemical reaction called exotherm and this increases the temperature of the mixture significantly hence the usage of Mylar. The barrier for the mold was double-sided tape and being glued to become a square mold, which was 16 cm in length. The thickness of the mold was 1.5 mm $\pm$ 0.12 mm.

2.3. Specimen fabrication

There were 14 specimens that have been fabricated for this study of which six specimens were done using 10% by weight, six were done using 20% fiber loading by weight and the other two were pure epoxy plates as experimental control measure. Mixing ratio of the epoxy that has been used for this study was 4:1 where 4 parts of epoxy to 1 part of hardener. Calculations for the quantity of the fiber, epoxy and hardener have been made before fabrication process was started. The epoxy and hardener were mixed initially together according to the ratio and being stirred up for about five minutes before the chopped fibers then stirred together with the mixture. The mixture then left to be transformed to its semisolid state before being poured to the designated mold. The mixture was spread equally on the surface of the mold before manual compression took place and it was left for curing using room temperature.

The composite plate then was released from the mold after being cured for more than 36 h. The curing speed depends on the temperature. Autoclave molding employs elevated temperature in curing the composite as it takes less time to be hardened. The composite plates then were cut to a square plate 15 cm in size. This ensures that the edge that was on contact with the barrier does not obstruct any moisture that is absorbed. Specimens were then ready to be tested.

2.4. Test method: moisture absorption

(i) The oven-dry specimen mass is recorded as the baseline mass, W_b, for moisture absorption. The specimen is placed in the conditioning chamber, which has previously reached the specified steady-state environment.

(ii) The specimen is removed from the conditioning chamber at the end of each time interval and the specimen is placed in the specimen bag. The bag is closed or sealed and the specimen is allowed to come to laboratory temperature. The specimen is removed from the bag and the specimen is wiped free of surface moisture with an absorbent lint-free towel. The specimen is weighed immediately to the required precision. Each measurement is recorded as W_i, along with the corresponding total elapsed time and the time interval since the previous weighing and the specimen is returned to either the specimen bag or the conditioning chamber. The specimen is not out of the conditioning chamber for more than 30 min/weighing and is not out of the specimen bag for more than 5 min/weighing.

(iii) The percent mass change at each time interval is calculated and recorded. The absorption of moisture is monitored until effective moisture equilibrium is reached. The last value of percent mass change is recorded as the moisture equilibrium content, M_m [3].

2.5. Test method: moisture desorption following absorption

If the material is known to contain or is suspected to contain, any appreciable amount of moisture-soluble ingredients, the specimen (following moisture absorption) is dried in an oven in to a state of essentially moisture-free equilibrium. This may also be done for evaluation of the moisture desorption characteristics of the material.

If the post-test oven-dry equilibrium mass is lower than the preconditioned mass, the difference is considered soluble matter lost during the testing process. For such materials, the effective moisture equilibrium content is calculated by adding the mass of the moisture-soluble matter (W_b–W_p) to the mass of the coupon at establishment of effective moisture equilibrium. This corrected moisture equilibrium mass is used to calculate the percent mass change and the resulting values are recorded as the moisture equilibrium content.

2.6. Oven-dry specimen conditioning

(i) The specimen is initially weighed to the required precision and this value is recorded as the baseline-mass, W_b, for oven-drying. The specimen is placed in the oven, which is located for the purpose of humidity control, in a facility that has a standard laboratory atmosphere.

(ii) At the end of each time interval, the specimen is removed from the oven and the specimen is placed in a desiccator to cool to laboratory temperature. After the specimen has cooled to laboratory temperature, the specimen is removed from the desiccator and the specimen is wiped free of surface moisture with an absorbent lint-free towel. The specimen is immediately weighed to the required precision. Each measurement is recorded as W_i, along with the corresponding total elapsed time and the time interval since the previous weighing and the specimen is returned to either the oven of the desiccator. The specimen is not out of the conditioning chamber for more than 30 min/weighing and is not out of the specimen bag for more than 5 min/weighing.

(iii) The percent mass change at each time interval is calculated and recorded. The specimen mass is monitored until effective moisture equilibrium is reached. The original moisture content of the material is calculated and recorded [3].

3. Results and discussion

3.1. Oven-dry test results and analysis

In order to make sure that the composite plate reaches its moisture equilibrium content, the plates first of all were dried up to its moisture-free state. This has been done using air-circulating oven type MERMERT model TV30U and the test temperature that has been used was 108 °C as moisture in the composite plates only can be freed if the temperature was higher than 100 °C. This is because that the matrix cured at lower temperature contains a higher concentration of hydroxyl group so it can bind with the water molecules easily through hydrogen bonding. The bound water molecules on the surface of the matrix can also be removed easily from the matrix when heated above 100 °C. In contrast, the sorbed water in the network of the matrix is hard to remove and remains after heating for 1h at 110 °C.

Six specimens have been tested and the weight of each specimen was measured every 2 h time interval for 60 h. The composite plates were said to be moisture-free if there was no significant decrease in the weight of the plates after a numbers of measures. It is crucial to collect the laboratory environmental data such as temperature and humidity as changes in temperature and humidity result in experimental anomalies. At time when the tests were on run, the laboratory temperature and humidity level both were recorded as 27.5 °C and 64.2%, respectively.

As these composites plates were made using short fibers that have been chopped and orientated randomly inside the resin, it was suspected that some of the fibers were placed near and almost on the same level as the surface of the composites. As the fiber composition in 20% fiber loading composites were higher, the number of short fibers that deposited at the surface of the plates were higher, hence the highest percentage. Epoxy plates acted as the control specimens that have nothing impregnated inside them have the lowest moisture content among others, as there were no fibers deposited at the surface of the composites. As such, the moisture uptake was lower compared to the other specimens. Fig. 1 shows the post-desorption moisture content by the representation of bar chart and it is clear that the highest moisture content comes from 20% fiber loading composites.

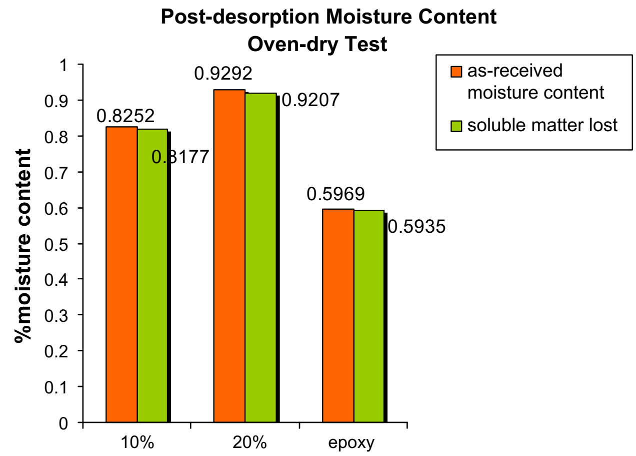

Fig. 1. Post-desorption moisture content.

3.2. Moisture absorption property determination

All specimens had undergone procedure A in determining the moisture effective equilibrium content thus led to the determination of the Fickian diffusivity constant. All specimens were placed inside the temperature control water bath located at the Soil Engineering and Natural Resources Laboratory, Department of Biological and Agricultural Engineering, Universiti Putra Malaysia. The moisture conditioning process included immersing the specimens in distilled water bath which has 100% relative humidity level with a constant temperature of 40 °C as this humidity level and temperature were used to simulate the water characteristics of sea water though the water used for this test was only distilled water. A time interval of 24 h per weighing has been set up for the purpose of the operator's convenient.

The moisture absorption behavior of the sugar palm fiber reinforced composites can be characterized by two quantities; the equilibrium moisture content, which characterizes the composite affinity for water (hydrophilicity) and the duration of the transient which is directly related to the sample thickness and to a parameter called diffusion coefficient or diffusivity which is the characteristic of the rate of transport of water molecules in the composite. The saturation water mass concentration is a constant that can be achieved only if the interaction between composite and water is reversible. If equilibrium of mass in the composite were not reached, it was likely that the mass would increase or decrease after that maximum point and this could indicate the existence of an irreversible process due to either hydrolysis or damage.

All specimens were immersed in the water bath for 768 h and data were collected at specific time interval. Graph plotted shows that all specimens are following the Fick's curve and this has been identified as Fick's curve as an initial portion of the graph made a linear line. If there is no discernible initial portion of the moisture absorption versus $\sqrt{time}$ plot, then the assumption of single-phase Fickian moisture absorption and the calculated moisture diffusivity constant may likely to be invalid.

All specimens were likely to follow the same trend and most of them reached effective moisture equilibrium content at day 30 afterwards. Epoxy plates were the only specimens that still having an increment in their weight up to the time when this report is being written. Lee et al. [4], Apicela et al. [5] and Barrie et al. [6] have discussed the method of water transport in epoxy resins thoroughly and most of the tests that have been done using fiber reinforced epoxy resins (eight ply laminates carbon-epoxy) took over three months to reach its effective moisture equilibrium content. Since the epoxy plates still showing a significant increase in weight, the determination of the diffusivity constant are neglected.

Diffusion process in organic polymer matrices usually described as two steps mechanism. The Fick's second law of diffusion used to describe the second step in the diffusion process where solvent diffuses in the direction of the concentration gradient. However most of the specimens that have been tested successfully showed that they were following the Fick's second law of diffusion.

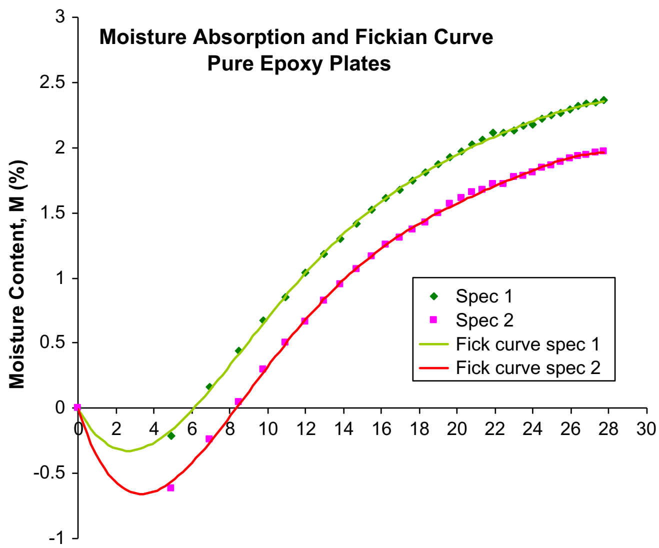

Fig. 2. Moisture absorption and Fickian curve for pure epoxy plates.

Using the equation for the correction factor, the corrected values for D_z can be calculated. Rationale of this correction factors is accounting for moisture ingress through the edges of a rectangular specimen. The corrected diffusivity values still show that the plates with 20% fiber loading have the highest number compared to 10% fiber loading specimens and the difference is relatively small when being represented in percentage which is only 6.24%.

Since the fibers were not woven, it is suspected that the short fibers were not aligned to the direction of the plate's surface (horizontally) but some of it was aligned vertically or even diagonally. This in a way causes a slight deposition of the fibers to the floor surface of the plates and it is has been assumed that the deposition of the fibers into the plates' floor surface were happened during the curing process using manual compaction as the plates were cured using room temperature, the prolonged period could lead to the fibers deposition in epoxy liquid form. This hypothesis can be supported evidently by the existence of brown stains on the absorbent cloth in the process of wiping the specimens prior to its obligatory weighing period. Since epoxy is in its glassy form, there is no way that the stains were coming from the epoxy itself so it is postulated that the stains came from the outer layer of the fibers.

Fig. 2 shows the experimental data and the Fickian curve for the epoxy plates and the phenomenon was progressively

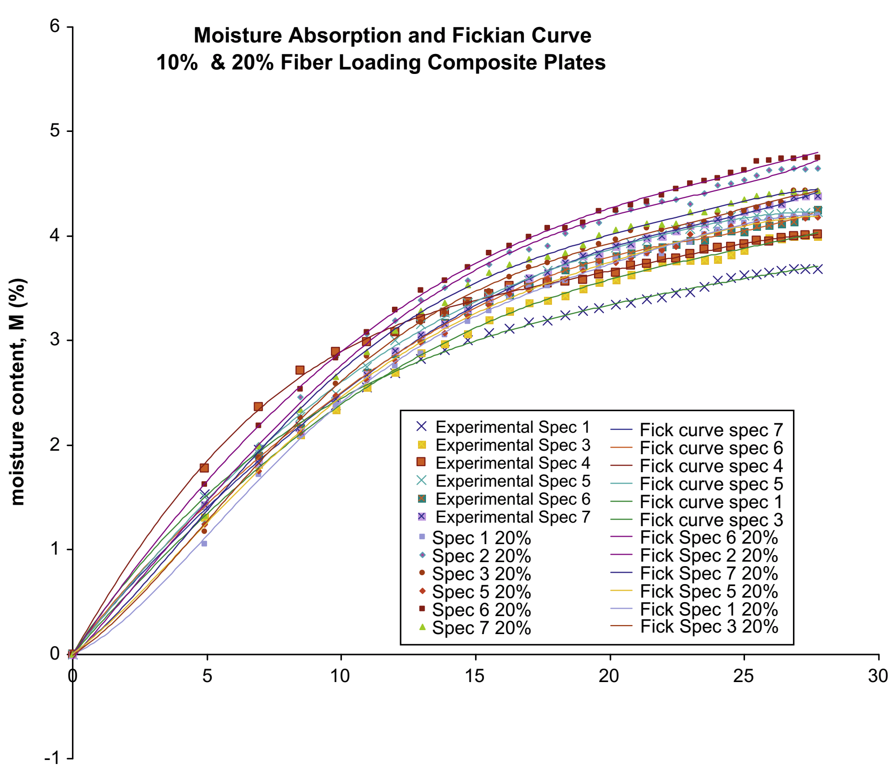

Fig. 3. Moisture absorption and Fickian curve for 10% and 20% fiber loading composite plates.

less evident as the plates were immersed in the water. After an equivalent ageing time of 86,400 s, the samples were dessicated and weighed in the dry state. Weight losses ranging from 0.21% to 0.62% for the plates were investigated. This weight loss, which is in opposing of weight gain could be possibly attributed to micro-cavitational damage; the size and nature of localized crazing apparently affects water uptake of epoxy resins. Weight losses after a fixed time are also strongly influenced by the test temperature. The higher weight losses observed for the plates may indicate that segregation or incomplete curing of some pre-polymer components may occur around inclusions, and that unreacted components or partially bonded groups may be desorbed easily and preferentially to the debonded and water-coated fibers [5].

It is observed that from the graphs of 10% and 20% (Fig. 3) fiber loading, the specimens were behaving in the same trend as the curves progressed. Both specimens produced smooth curves when plotted against square root of time. Micro-bubbles that may entrap inside the composite may become micro-voids that will enhance the moisture uptake of the composites.

4. Conclusions

Experimental and analytical studies have been performed on the moisture absorption behavior of sugar palm fiber reinforced composites plates, whose environmental properties have been fully determined. Composite specimens and pure epoxy plates that acted as control have been subjected to full immersion in water at a specific temperature. The moisture absorption process was determined to be reversible with a small saturation moisture uptake and with a slightly lower absorption through-the-thickness direction. The water transport in this material follows a typical dual sorption-diffusion process and the diffusion process obeys the Fick's law. The behavior through-the-thickness follows a parabolic behavior. The slopes of moisture absorption plot in the initial linear portion of the curve seem to be similar for almost all specimens. The values led to the determination of the Fickian diffusivity constant, which showed that the 20% fiber composite plates have the highest value. After being corrected using the correction factor, a difference of 6.24% in value of the diffusivity constant for both 10% and 20% composite plates were obtained. This shows that 20% composite plates absorb more water compared to other specimens tested. This may due to the deposition of short fibers to the floor surface of the composites, which led to the absorption of moisture by the fiber and epoxy resins too.

Acknowledgments

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