HYBRIDIZATION AND THERMAL STABILITY EFFECTS ON PHYSICAL PROPERTIES OF HYBRID GLASS/JUTE FIBER REINFORCED EPOXY COMPOSITES

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ABSTRACT: Incorporation of natural fibers with partial replacement of synthetic fibers leads to a new venture in composite tooling industries due to their recyclability limitation as well as potential for weight and cost reduction. This work compares the density and evaluates the effect of thermal cycles of 0, 1, 3, 7, 12, 20, 30 and 50 cycles at two different temperatures; i) 120°C and ii) 200°C to the physical properties of hybrid glass/jute fiber reinforced epoxy laminated composites. The composites were prepared via vacuum infusion working at a pressure of 100 kPa, soaking time of 120 minutes and with the use of flow media. The composite exposed to thermal cycles at 200°C exhibited lower density due to removal of volatile elements which lead to porous structures. It experienced higher weight loss from partial degradation of natural fibers at 200°C. The composite subjected to thermal cycle at 120°C showed stable physical properties even after 50
thermal cycles. The water absorption reflects the level of curing completeness, which varies with exposed temperature and number of thermal cycles. The findings indicate that the hybrid glass/jute fiber reinforced epoxy composite exhibits potential to be used as a tooling material with apparent weight reduction and dimensional stability under thermal effect.

**KEYWORDS**: Hybrid Laminates; Natural Fiber; Glass Fiber; Physical Properties; Thermal Stability

### 1.0 INTRODUCTION

Nowadays, the use of natural fibers in composite applications has received substantial interest due to government policies towards green materials. This type of composites provides several advantages over the existing synthetic fibers especially for their contribution in the sustainability aspect. Natural fibers can be found in fibrous form and can be extracted from plant leaves, stalks, fruits and seeds at very low cost [1]. The most common natural plants used in applications are bast fibers, such as kenaf, hemp, jute, flax and sisal. Some of these materials have higher specific strength than fiber glass and similar specific modulus [2]. They also offer some advantages over synthetic fiber such as low cost, low density and biodegradability. Furthermore, natural fibers are environmentally friendly, less harmful to health and unlike glass and carbon fibers, the energy consumption to produce them is very small [3]. Natural fibers are considered as suitable replacements for synthetic fibers in many sectors such as automobiles, furniture, packaging and construction [4].

Jute fibers possess immense potential as fibrous reinforcement in polymeric composites. The fibers are extracted from jute plant named Corchorus that belongs to the Malvaceae family. Jute fibers extracted from bast or skin of plant are off-white to brown in colour and measured 1-4m long. The fiber thickness varies between 40 and 80 μm which leads to a variation in the tensile strength between 100 and 480 MPa. In addition, jute fibers can withstand the temperature of up to 100°C [5]. The density of jute fibers is in the range of 1.25 to 1.5 g/cm³ compared with 2.54 g/cm³ for E-glass fibers and 1.8 to 2.1g/cm³ for carbon fibers.

Hybridization of two or more different continuous fibers in a matrix is a versatile approach to improve strength or stiffness of structural composites. Performance of a hybrid composite is a combination of
weighted amounts of the individual components which balances between the strength and weakness of the individual components [6]. The selection of suitable components is determined by the desired properties required for the final products. Despite the excellent performance of synthetic fibers such as high strength and stiffness [7] as well as good chemical resistance [8], issues on production cost and disposal of after used products of non-biodegradable materials are crucial. Therefore, the utilization of either solely natural fibers or hybridization with synthetic fibers has attracted the attention of scientists and technologists [9]. It has been observed that natural fiber composites have compatible electrical resistance, good thermal and acoustic insulation properties as well as good resistance to fracture [9]. Furthermore, the use of green composites offers several advantages such as light weight, low production cost and low thermal mass [10].

The process parameters to fabricate hybrid glass/jute fiber reinforced epoxy laminated composites via vacuum infusion technique were optimized using response surface methodology as reported by Mohamad et al. [11]. The composites exhibited high flexural strength of 194.85 MPa and flexural elastic modulus of 13.41 GPa. Despite the potential of these green composites to be used as tooling material, studies on thermal effect on its properties are crucial. During service, a tooling material is subjected to repeat thermal cycling and is likely to induce distortion on the tooling due to the difference in thermal mass and coefficient of thermal expansion. Thermal cycling is defined as alternate heating and cooling of materials. Low thermal cycle means the time taken for completion of the cycle is large enough to cool the component. On the other hand, high thermal cycle means time involved is in milliseconds and the heating and cooling is influenced by the thermal inertia of the system under consideration [12]. During heating and cooling, the material experiences expansion and contraction which changes dimension of the material when subjected to thermal cycling. In this study, the physical properties of hybrid glass/jute fiber reinforced epoxy laminated composites under thermal effect are evaluated.

2.0 EXPERIMENTAL

2.1 Materials
The unmodified basic liquid epoxy resin, DM15 (F3) A made from bisphenole-A and epichlorohydrin with medium viscosity of 12 – 15 PaS was used as the polymer matrix. A low viscosity (4.7 PaS)
modified liquid amine epoxy hardener, DM 15 (F3) B was used as catalyst and accelerator. Both the epoxy resin and hardener were obtained from Chemrex Corporation Sdn. Bhd. Kuala Lumpur, Malaysia. Woven E-glass of 7 x 6 (warp x weft) as reinforcing material was purchased from ML Fibreglass Sdn. Bhd., Johor Bahru, Malaysia. The jute fiber mat used was 13 warp x 12 weft and manufactured by Sonali Aansh Group, Dhaka, Bangladesh. The density of the jute fiber is 1.28 g/cm³.

2.2 Sample Preparation
Prior to vacuum infusion process, the working surface was applied with release agent to facilitate easy removal of molds. The epoxy resin and hardener were mixed according to 5:1 weight ratio. The hybrid laminated composites of 7 plies were fabricated in the following sequence: Glass/Jute/Glass/Jute/Glass/Jute/Glass. The number of layer was selected since it was the maximum capacity for the utilized vacuum infusion facility to ensure overall coverage of resin on fibers without obvious resin starvation. Moreover, the stacking sequence was chosen to overcome hydrophilicity of jute fibers and ensure minimal moisture absorption into laminated composites once in contact with water [13]. Once soaked for a specific time, the vacuum was cut off and the composites were left to cure at ambient temperature before mold release. Then, the composites were exposed to thermal cycling effect by placing in and out of an oven (Thermotec 2100 from Contherm Scientific Ltd) for 10 minutes for each cycle. The thermal cycling test was conducted at two (2) different temperatures; 120°C and 200°C for 0, 1, 3, 7, 12, 20, 30 and 50 cycles. Then, the physical properties such as density and water absorption were tested, and the results were supported by thermal (TGA) analysis of the materials.

2.3 Testing and Analyses

Density
Density measurement was performed to investigate the contribution of hybridization between glass and jute fibers to the weight reduction of hybrid laminated composites as compared to glass laminated composites. It was measured according to ASTM 792 (Standard Test Methods for Density and Specific Gravity (Relative Density) of Plastic by Displacement) by using an electronic densimeter. This test method involves weighing a one-piece sample of 1 to 50 g in water and using a
sinker with plastics that are lighter than water. The sample’s dimensions for this experiment were 20mm x 20mm. The samples were placed in a beaker and readings were recorded. The weights of the sample were recorded in water, in air and in the total mass/volume g/m³. Finally, the density was measured according to the Archimedes’s Principle using Equation (1). The procedure was repeated for each sample.

\[
\text{Density} = \frac{\text{Mass in Air}}{\text{Mass in Air} - \text{Mass in Water}}
\]  

(1)

**Water Absorption**

The relative rates of water absorption behavior for hybrid laminated composites were determined according to ASTM D570 (Standard Test Methods for Water Absorption of Plastics). Samples were prepared in the form of a bar with dimension of 50mm x 50mm and dried in a vacuum oven at 50 ± 3°C for 24 hours. The dried weight of composite was measured. Then, they were immersed in distilled water in a water bath machine to get a weight that is to the nearest 0.001 g. The water absorption test was conducted at two (2) different parameters; i) the sample was immersed in water at 40°C temperature and after 48 hours, the sample was taken out, and then the surface was wiped off with a dry cloth and weighed immediately using a balance. The percentage increment in weight for the immersion (water absorption), WA was calculated to the nearest 0.01% using Equation (2) whereas, ii) the sample was immersed in water for 5 days at room temperature and for every 24 hours, the sample was taken out and water was wiped off from surface and then weighed to the nearest 0.001g immediately using balance. The percentage increment in weight for the immersion (water absorption) was calculated to the nearest 0.01% using the Equation (2) for 5 days.

\[
\text{WA} \, (\%) = \frac{\text{Wet weight} - \text{Conditioned weight}}{\text{Conditioned weight}} \times 100\%
\]  

(2)

**Thermogravimetric (TGA) Analysis**

The TGA analysis was performed by using Perkin Elmer TGA 7. The weight of sample used was about 10 mg. The samples were heated
from 35°C to 800°C at a heating rate of 10°C/min in a dynamic nitrogen atmosphere. Prior to that, each sample was held at a temperature of 35°C for one minute to eliminate the moisture content. The weight loss during the heating period was automatically recorded and plotted as a function of temperature.

3.0 RESULTS & DISCUSSION

3.1 Density of Laminated Composites

Table 1 lists the density of hybrid laminated composite samples as compared to the control sample; glass laminated composite samples. From the result, it shows that hybrid laminated composites have lower density and lighter than glass laminated composites. The results indicate that the hybridization between glass and jute fiber shows high potential to reduce the weight, as much as 25%. This observation was an unequivocal evidence for the contribution of low density jute fibers to the total density of the composites. The hybridization of different type of fibers in a matrix allows a closer tailoring of composite properties. These results give a good impression in the selection of materials to be widely utilized as construction materials in composite tooling technology.

<table>
<thead>
<tr>
<th>Laminated Composite</th>
<th>Mass of Sample in air (gram), ( W_a )</th>
<th>Mass of Sample immersed in water (gram), ( W_w )</th>
<th>Density (g/cm(^3))</th>
<th>Average of Density (g/cm(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass Laminated Composite</td>
<td>0.5921</td>
<td>0.2755</td>
<td>1.87</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.7615</td>
<td>0.3677</td>
<td>1.93</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.7700</td>
<td>0.3681</td>
<td>1.92</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.7197</td>
<td>0.3371</td>
<td>1.88</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.8533</td>
<td>0.3969</td>
<td>1.87</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.9643</td>
<td>0.4443</td>
<td>1.85</td>
<td></td>
</tr>
<tr>
<td>Hybrid Laminated Composite</td>
<td>0.7405</td>
<td>0.2110</td>
<td>1.40</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.7291</td>
<td>0.2120</td>
<td>1.41</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.7084</td>
<td>0.2112</td>
<td>1.42</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.8641</td>
<td>0.2422</td>
<td>1.39</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.6844</td>
<td>0.2017</td>
<td>1.42</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.9592</td>
<td>0.2590</td>
<td>1.37</td>
<td></td>
</tr>
</tbody>
</table>
The effect of thermal cycles on density for two different temperatures, 120 and 200°C is depicted in Figure 1. It is apparent that composites exposed to the thermal cycle of 200°C show lower density than those exposed to 120°C. The removal of fume/volatile elements at 200°C occurred at a faster rate resulted in rather porous and opened structures which then decreased the density of the composites. It was confirmed with the morphological analysis conducted on the sample, but, the result is not included in this paper. This is in line with the thermal analysis results observed using TGA. The release of volatile components for the composites at room temperature or at 120°C occurred at a rather slower rate, which allowed the composites to maintain its closed structures and structural integrity. This finding is in good agreement with the water absorption of the composites.

3.2 Water absorption

Figure 2 shows the percentage of water absorption for each hybrid laminated composites at accelerated diffusion. The result shows the importance of curing completeness in a composite which determines the susceptibility of a composite towards water absorption. Although the curing process was performed as suggested by the manufacturer, there were still curing level differences between outer and inner layer of the composites. As in our case, the sample’s cutting process exposed the inner layer of the composites to water. These samples were directly immersed in water after a day conditioning process where the entire testing sample was placed in a humidity chamber at 23 ± 2°C and a relative humidity of 50 ± 5% for 24 hours without further exposure to higher temperature. The highest percentage of water absorption was clearly observed in samples with 0 thermal cycle. The hydroxyl groups presence on natural fibers make the composites susceptible to water absorption. In overall, the water absorption decreases with the increase of thermal cycles. A nearly stable water absorption curve suggests that a complete curing occurs at around 12 cycles.
As for the temperature effect, samples exposed to 200°C demonstrated higher water diffusion than those exposed to 120°C during the earlier stage of the testing. This was the results of the faster rate of volatile/fume removal from the surface of the composites which attracts the water molecules penetrated the materials for replacement. So, the highest water diffusion was exhibited by composites exposed to 200 °C at the early stage of the thermal cycle of 0 to 7 cycles. The rate reduced as the curing process was completed at around 12 cycles when 3D networks of crosslinking formed between the epoxy chains. This is attributed to the natural capillaries presence in the jute natural fiber at the cut surfaces of the composites. The amount of absorbed water in fibers depends on density and water diffusivity of the fibers [14-15].
Figure 3 shows the water absorption rate at room temperature for 5 days. The rate increases almost linearly with time and no saturation was observed even after 5 days. This is attributed to the level of curing as well as thermal oxidation of fibers and matrices experienced by the composites. The highest water absorption was observed for the samples without thermal cycles. No exposure to higher temperature during fabrication resulted in slow manner for full curing to be achieved for the hybrid composites. Partial curing attracts higher amount of water molecules to penetrate the samples. The rate reduces at higher temperature. Samples exposed to 200°C manifested slower water absorption rate than those exposed to 120°C. This was due to the level of curing in the samples.

![Graph](image)

Figure 3: Water absorption of hybrid glass/jute laminated composites exposed to (a) 120°C and (b) 200°C for various numbers of cycles
The amount of the absorbed water notably decreases with the increase in thermal cycles. This is also in good agreement with the curing completeness. Uncured composites condition after fabrication was evident from the thermal analysis conducted via thermal gravimetric (TGA), where samples exposed to 200°C at 0 cycle showed lower weight loss for the temperature range between 100 to 300°C compared to one exposed to 120°C.

3.3 Thermal Analysis

TGA thermograms for the hybrid glass/jute laminated composite are shown in Figures 4 and 5. TGA analysis can provide an early indication of the impact of thermal cycling on the hybrid laminated composite based on the characteristics of weight loss versus temperature curve.

(a)
3.3.1 Effect of Thermal Cycles at Constant Temperature

In Figures 4 and 5, all samples manifested four steps of thermal degradation profile via weight loss with increasing temperature. Samples that underwent thermal cycling at 0 cycle showed lower onset temperature than the samples that underwent more than 30 cycles. This could be due to the curing completeness and phenomena of oxidation occurred in functional groups containing oxygen and humidity or water vapor. As observed in the TGA thermogram of hybrid glass/jute laminated composite subjected to 120°C/0 cycle (Figure 4), there was downfall at about 100°C. This was due to vaporization of water molecules and the trapped moistures inside the samples especially in natural jute fibers. Water loss in the initial phase was about 5% in all hybrid laminated composites. This observation indicated the sample at 0 thermal cycle prone to absorb water due to incomplete curing. This is in good agreement with the water absorption results. The sample was the first to experience weight loss if compared to the other samples.
It was found that there were more than three levels of weight loss which represents the thermal degradation profile of the hybrid laminated composites. The first stage involved the removal of water molecules followed by the second stage for the weight loss in the matrix phase from curing process due to removal of solvent in matrices at around 200 to 280°C [16]. Under inert conditions, most thermoset degrades in a temperature range between 400 and 600 °C [16]. The third stage of around 320°C was the thermal degradation of jute fibers and finally followed by the final stage at around 400°C due to the weight loss from decomposition of the epoxy matrices. The heavy downfall during weight loss was observed in the temperature range of 200 to 500°C with approximately 40%-50% of weight loss. Basically, all samples showed two steps slide of thermal degradation profile weight loss with increasing temperature. Results demonstrated that samples exposed to 120°C had lower onset temperature than samples exposed to 200°C (Figure 5). It was due to the pyrolysis phenomena which involved functional groups containing oxygen-labile liberating CO, CO₂ and moisture or water vapor [17]. This supported the argument on faster removal of fume/volatile components in samples exposed to 200°C compared to 120°C which resulted in higher weight loss observed in the samples under TGA (Table 2).

### 3.3.2 Effect of Different Temperatures at Constant Thermal Cycle

Table 2 summarizes the results of TGA thermogram analysis for weight loss and residual percentages at 800°C for hybrid glass/jute laminated composites at 0 cycle. At this early stage, once the samples were exposed to the temperature for the first 10 minutes, the...
temperature assisted further crosslinking in the composites [18]. In this regard, it was found that samples exposed to 200°C had higher temperature level profile for each slide heat measured for both weight loss and maximum weight loss, % indicating higher crosslinking density achieved by the samples. In contrast, samples exposed to 120°C had a lower temperature for each stage of the slide profile. As both samples experienced thermal aging during TGA analysis, the higher oxidation effect led to higher weight reduction of the composites. In this case, higher residual percentage of around 45% at 800°C was observed for samples exposed to 120°C. Meanwhile, lower residual in sample exposed to 200°C showed higher materials degradation level had taken place. These findings are in good correlation with the thermal behaviors for all composites either exposed to thermal cycles at 120 or 200°C as shown. From the results obtained, it can be concluded that the sample exposed to 200°C experiences higher thermal degradation and has slightly lower thermal stability than those exposed to 120°C.

Table 2: Summary for thermal degradation profile weight loss and residue analysis on the temperature stabilization from TGA thermogram

<table>
<thead>
<tr>
<th>Sample</th>
<th>Temperature (°C)</th>
<th>First step weight loss</th>
<th>Second step weight loss</th>
<th>Residue at 800°C (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>120°C</td>
<td>322.76</td>
<td>400.21</td>
<td></td>
<td>44.46</td>
</tr>
<tr>
<td>200°C</td>
<td>325.77</td>
<td>405.68</td>
<td></td>
<td>42.11</td>
</tr>
</tbody>
</table>

4.0 CONCLUSION

The hybridization between glass and jute fiber showed a great contribution in reducing the density of the composite by almost 25%. The number and temperature of thermal cycle were proven to significantly affect the physical stability of the hybrid glass/jute laminated composites. The water absorption of the hybrid glass/jute fiber reinforced epoxy laminated composite was found to be less than 10% absorbed weight and the value decreased with the increment in the number of thermal cycles. The composites exposed to higher temperature of 200°C manifested slightly higher water absorption of 14% compared to those exposed to 120°C. The composites that exposed to thermal cycles at 120°C were proven to maintain physical properties even at the highest cycles. This is a good indicator for a significant thermal stability and structural integrity of the composites. Moreover, the study shows the relation between physical
properties and level of curing completeness of the composites. This finding is significant as it indicates the presence of various fibers (natural and synthetic) in epoxy matrices [19] alters the curing duration recommended by the manufacturers.

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