Effect of Water Absorption on the Tensile Characteristics of Natural/ Synthetic Fabrics Reinforced Hybrid Composites

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1. INTRODUCTION

Natural fiber strengthened polymer composites have raised great attention and interests among materials scientists due to the considerations of promoting an environmental friendly, high specific strength, low density, cheaply and biodegradable compared to the synthetic fiber-reinforced composites. The various types of natural fibers such as jute, hemp, sisal, bananas, flax, and bamboo can be very cost-effective material for applications such as automotive exterior, packing, building, construction industry, electric and electronic devices, and transportation [1-6]. One of the main major reasons for limitation using natural fibers is a high response of water absorption which distorts its mechanical properties. A lot of works used the combination of synthetic fiber and natural fibers reinforced polymer to reduce the uptake water. Sanjay et al. [7] the water absorption of laminate [G/G/B/G/B/G/B/G/G] was slightly exceeded from that obtained for laminate configuration with glass only. Carbon-E-glass/polyester hybrid composites, prepared by hand lay-up at several stacking sequences, showed the maximum moisture ratio reached to 3.5% at immersion time 264 hour for the combination [G/G/G/G] [8]. Also, Jesthi et al. [9] used the same fibers (carbon and glass) reinforced polyester composites at various stacking sequences to evaluate the tensile strength after immersed into the seawater for 90 days. After seawater aging, the retention of tensile strength in both designations [C/ C/G/G/G] and [C/G/G/C/G] was greater than that of the glass laminate composite. Sivakumar et al. [10] the maximum percentage of water absorption of polymer composites...
based on different weight fractions of the glass/nylon/jute obtained for the composition (glass 11% + Nylon 26% + jute 35%) and reached to 9.52%. The addition of jute fiber with glass fiber in polymer epoxy composite enhanced the tensile strength and impact energy but reduced the resistance for water absorption compared to pure glass composite [11]. The addition of the carbon layer to the flax fabrics reinforced epoxy composite improved the tensile strength and reached 288 MPa compared to the flax composite [12]. In addition, Gupta et al. [13] the highest water absorption ratio obtained for hybrid composite with sisal 20 wt% and glass 20 wt% and reached 6%, compared to other composites. In another study, Gupta and Deep et al. [14] the designation (G/S/G/S/G) provided maximum values of the tensile strength and flexural strength in both dry and wet conditions as compared to other stacking sequences. The hybridization using carbon fiber with natural fibers such as cross-ply flax-based polymer composite showed an enhancement in the tensile properties (284.8 MPa) and reduction in water absorption ratio (5.5%) compared to those without hybridization [15]. Girisha et al. [16] studied the effect of water absorption of composites reinforced by natural fibers such as sisal and coir at different weight fractions (20 %, 30 %, and 40%) on their tensile properties. The maximum tensile strength after hydro aging obtained for the hybrid composite reinforced by 40 wt% sisal-coir fiber reached 48 MPa. The water absorption of hybrid composite reinforced by jute/glass fiber in several environments such as seawater, distilled water, and acidic water has been investigated [17]. The samples immersed in distilled water gives maximum moisture content and higher value of the diffusion coefficient compared to other water conditions. Sathish et al. [18] hybrid composite reinforced 30% flax and 10% bamboo exhibited maximum tensile strength (31.55 MPa) and minimum water absorption ratio (9.3%), compared with the natural bamboo composite. Gopal et al. [19] the composite sample (15% sisal: 15% jute: 5% flax) with comparison with various concentrations (20:10:5 and 25:5:5) gives a minimum ratio of water absorption (14.73%). The effect of carbon fiber hybridization on the water absorption of unidirectional and cross-ply flax specimens has been investigated [15]. Hybridization using synthetic fiber (carbon) with two types of natural fibers (UD and CP flax) gave maximum water absorption of the hybrid carbon/UD flax specimen and carbon/CP specimens of approximately 2 and 8%, respectively. Calabrese et al. [20] used another synthetic fiber (glass fiber) with natural fiber (flax) reinforced epoxy composite to evaluate the water absorption capacity. This hybridization reduced the saturation in water absorption ratio to 6.9%, in addition, Saidane et al. [21] used the same hybridization [20], but at different layers configurations between the flax and carbon fibers, to study the effect of stacking sequence on tensile strength after hydro aging effect. For the combination [G/F/F/F/F], the reduction in tensile strength was about 11%, while it reached to about 21% for the flax laminate.

In the present work, different fiber configurations of flax/sisal/glass/carbon reinforced unsaturated polyester hybrid composites have been prepared using Hand lay-up (HLU) route. The samples were immersed in distilled water to evaluate their resistance to water absorption and swelling thickness. The tensile strength test was carried out before and after hydro aging. In addition, fracture surfaces were examined using SEM to investigate bonding between layers before and after hydro aging.

2. MATERIALS AND EXPERIMENTAL PROCEDURE

2.1 Materials and Manufacturing

Composites with various accumulating sequences and proportional fibers content were manufactured using the hand lay-up technique. The unsaturated polyester and hardener with a proper proportion of 4:1 were mixed in the presence of a catalyst plus1.5% resin using mechanical stirring for 5 min. The mixture was fed into the mold with dimension 250 x250 cm. Then, the fibers were laid according to the designated sequences. Finally, the roller was passed over the laid composite to remove bubbles formed during manufacturing. The prepared composites were kept at room temperature for two days to provide optimal stiffness and retraction. The fibers used to reinforce unsaturated polyester are flax, sisal, E-glass, carbon.

Table 1 shows the physical and mechanical properties of the sisal, flax, E-glass, and carbon fabric reinforcements [22-25]. Non-hybrid and hybrid composite samples consist of six fabric layers. The combinations were named F, S, C, G, L1, L2, L3, L4, L5, L6, L7, and L8 according to the designation listed in Table 2.

| TABLE 1. Mechanical property of the used fabrics |
|-----------------------------------------------|
| Properties                  | Flax | Sisal | E-glass | Carbon |
| Density [g/cm³]              | 1.40 | 1.46  | 2.55    | 1.78   |
| Tensile strength (MPa)       | 1500 | 700   | 3400    | 4200-4800 |
| Elastic modulus (GPa)        | 60-80| 35    | 73      | 260    |
| Shear modulus (GPa)          | 28.2 | 18.2  | 33      | 110    |
| Specific (E/d)               | 30   | 29    | 29      | -      |
| Poisson ratio                | 0.23 | 0.23  | 0.22    | 0.279  |
| Elongation (%)               | 3.2  | 3-7   | 1.8-3.2 | 1.75-1.95 |
| Moisture (%)                 | 8-12 | 10-20 | -       | -      |
| Fiber diameter (µm)          | 40-600| 50-200|17      | 5-7    |
| Lignin Content (%)           | 67/3 | 75/12 | -      | -      |
2.2 Water Absorption and Thickness Swelling Test

The water absorption and swelling thickness behavior of the non-hybrid and hybrid composite were evaluated. In order to study the effect of hydro aging, both weight and thickness of samples were measured before and after immersion for various intervals time in distilled water. The weight of the sample was measured by using an electronic balance with an accuracy of 0.1 mg. This procedure was repeated until the weight of the samples remained constant up to a 4-digit number with increasing time of immersion.

The estimated water absorption ratio and the swelling thickness percentage, is calculated using Equations (1) and (2) below:

\[ WA(\%) = \frac{w_t - w_o}{w_o} \times 100 \]  

\[ TS(\%) = \frac{t_t - t_o}{t_o} \times 100 \]

where \( w_o \), \( w_t \) and \( w_o \) are the water absorption percentage, the weight of the initial and wet samples at a given time, respectively.

2.3 Tensile Strength

Figure 1 show the setup and dimensions of the tensile test sample used to evaluate the tensile characteristics. The tensile strength of composites was measured before and after hydro aging with the universal testing machine (INSTRON 8801) at a crosshead speed of 5 mm/min. According to ASTM D638 standard, the average of five tensile strength measurements for each composite sample with dimension (165 mm x13 mm x4 mm) was recorded.

2.4 Morphological Characterizations

The morphology of composites was investigated at fractured surface of samples using (SEM, Quanta FEJ20) at 20 kV.

| Laminate | Fiber configurations | Weight fraction (wt. %) | Volume fraction (vt. %) |
|----------|----------------------|-------------------------|-------------------------|
|          | Matrix  | Carbon | Glass | Flax | Sisal | Matrix  | Carbon | Glass | Flax | Sisal |
| F        | [F/F/F/F/F/F]       | 40 0 0 60 0 40.6        | 0 0 0 59.4 0          |
| S        | [S/S/S/S/S/S]       | 40 0 0 0 60 41.46       | 0 0 0 58.54 0         |
| C        | [C/C/C/C/C]         | 40 60 0 0 0 46.65       | 0 53.35 0 0 0         |
| G        | [G/G/G/G/G/G]       | 40 0 60 0 0 53.42       | 0 46.58 0 0 0         |
| L1       | [C/C/F/C/F/C]       | 40 40 0 20 0 44.46      | 0 33.88 0 21.66 0     |
| L2       | [F/F/C/C/F/F]       | 40 20 0 40 0 42.45      | 0 16.18 0 41.37 0     |
| L3       | [C/C/S/C/C]         | 40 40 0 0 20 44.78      | 0 34.14 0 0 21.08     |
| L4       | [S/S/C/C/S/S]       | 40 20 0 0 40 43.06      | 0 16.41 0 0 40.53     |
| L5       | [G/G/S/F/G/C]       | 40 20 20 10 10 47.24    | 18 12.12 11.52 11.12  |
| L6       | [G/C/C/F/G/G]       | 40 20 20 10 10 47.24    | 18 12.12 11.52 11.12  |
| L7       | [S/F/G/C/F/S]       | 40 10 10 20 20 43.92    | 8.37 5.63 21.41 20.67 |
| L8       | [F/S/G/C/S/F]       | 40 10 10 20 20 43.92    | 8.37 5.63 21.41 20.67 |

The cross-section of investigated samples is coated with 100 Å thick platinum in the JEOL sputter ion coater.

3. RESULTS AND DISCUSSIONS

3.1 Water Absorption and Swelling Thickness Behavior

Figure 2 shows the dependency of water absorption ratio and swelling thickness on the immersion time for the hybrid and non-hybrid composites with various fiber configurations. Each data point was calculated from an average value of five samples for each laminate. These figures indicate that the
water absorption ratio for all laminates was increased with increasing immersion time until saturation point. These results coincide well with previous work for natural and synthetic fiber [1, 7]. From Figure 2 can be claimed that the non-hybrid composites with sisal or flax have the highest water absorption saturation up to 14% and 12%, and swelling thickness (20%), (18%), respectively. However, the minimum value of water absorption ratio was obtained for carbon composites with fiber configuration [C/C/C/C/C/C] and reached 1.45% as shown in Figure 2a. The small value of water absorption ratio, obtained for fiber configuration [C/C/C/C/C/C], can be attributed to low sensitivity of carbon to water absorption. Therefore, the swelling thickness diminished due to the smallest value of the water absorption ratio. It can be seen from Figure 2(b) that the trend of swelling thickness is similar to the water absorption behavior with immersion time until it reached the equilibrium conditions. In general, the constituents of flax and sisal fiber are cellulose and hemicellulose. The flow of water into the capillaries (pore absorption) and diffusion into amorphous body of cellulose and hemicellulose (fiber absorption) are the main sources for the uptake of water in natural fiber (flax and sisal). The probability of pores creation in natural fiber is high as compared to synthetic fiber. Therefore, the uptake of water in natural fiber is higher than that of synthetic fiber. Density and size of formed pores depended on the size, shape, and arrangement of natural fiber. The diffusion of water through an inter-molecular distance of chains leads to fiber swelling. The lower water uptake by flax fiber compared to sisal fiber reflects the fact that the number and size of pores in flax fiber are less than that in sisal fiber. High moisture susceptibility in natural fiber is considered one of the major issues in the usage of these fibers. Hence, we tend to use combination of natural and synthetic fibers. The water uptake ratio of hybrid composites laminate (L1) with fiber configuration [C/C/F/F/C/C] was around 37.6% of that obtained for non-hybrid composites with fiber configuration [F/F/F/F/F/F]. In addition, water uptake for laminate (L3) reached 50% of that obtained for sample (S). The drop in water uptake for laminate (L1) as compared to laminate F can be attributed to the existence of carbon fibers layers at the top and the bottom of the stack which works as a barrier for contact between the water molecules and flax fibers. On the other hand, the water uptake percentage of laminates L2 and L4 reached 62.5% and 71.4% as compared to F and S laminates, respectively due to the existence of natural fabrics at outer layers. In comparison with hybrid-composites, the laminate L5 with fiber configuration [C/G/S/F/G/C] is in second place in resistance for water uptake after laminate L1 due to the hybridization by the hydrophobic carbon fibers and adhesion between resin matrix and fiber.

3.2. Tensile Characteristics

Figures 3a and b show the load-displacement curves of the laminate (L1) with fiber configuration [C/C/F/F/C/C] before and after water absorption. These figures reflect the condition that the stress of a laminate (L1) sample before hydro aging gradually increased up to (73 MPa) at displacement 3.82 mm. However, the stress of this hybrid sample after the hydro aging enhanced up to 61 MPa at 4.1 mm displacement, and reduced abruptly down to 38 MPa at a maximum 4.40 mm displacement, and then, finally the sample was broken. The drop in the breaking stress for laminate (L1) sample after hydro aging can be attributed to the swelling of fibers which enhances the inter-molecular distance of chains as discussed in previous section. Both tensile strength and maximum strain of the hybrid and non-hybrid composites at various fiber configurations are listed in Table 3. Such a table reflects that the tensile strength of all samples before hydro aging is higher than those obtained after hydro aging.
Non-hybrid composites with flax laminate or sisal laminate have small tensile strength with considerable shifting to lower value after hydro aging. Additionally, it was found that the laminate (S) exhibited lower tensile strength compared to the laminate (F). In contrast, non-hybrid composite with carbon has the highest value of tensile strength with slight shifting to lower value after hydro aging effect. The hybrid-composites have moderate tensile strength. In general, mechanical properties of composites reinforced with fabric rely on the nature of each component in composites, distribution, and orientation of fiber through a matrix and of the interphase region between different components. The spread of water into pores as well as the inter-molecular distance of chains after hydro aging effect causes a reduction in the connection between the components of the sample as a whole. However, maximum strain of all laminates was enhanced after hydro aging effect due to an increase in the plasticization effect.

The small value of tensile strength for laminates S and F can be attributed to the lower strength of sisal and flax fiber as well as poor interfacial adhesion between the matrix and fiber. The huge penetrating water through laminates S and F cause an adverse effect on the interfacial region, created between base matrix and fibers, and structural solidity. Therefore, the tensile strength of laminate S and F was extremely reduced after hydro aging while maximum strain was enhanced. When the flax fibers was placed at the core in the laminate (L1), the tensile strength before and after hydro aging was closest to the laminate (C). The response of water uptake for laminate L1 is weak because of the presence of carbon at outer layers. Hence, the change in tensile strength and maximum strain after hydro aging is slight. In contrast, the small value of tensile strength for laminates S and F can be attributed to the lower strength of sisal and flax fiber as well as poor interfacial adhesion between the matrix and fiber. The huge penetrating water through laminates S and F cause an adverse effect on the interfacial region, created between base matrix and fibers, and structural solidity. Therefore, the tensile strength of laminate S and F was extremely reduced after hydro aging while maximum strain was enhanced. When the flax fibers was placed at the core in the laminate (L1), the tensile strength before and after hydro aging was closest to the laminate (C). The response of water uptake for laminate L1 is weak because of the presence of carbon at outer layers. Hence, the change in tensile strength and maximum strain after hydro aging is slight. In contrast,

![Figure 3](image_url)

**Figure 3.** Load-displacement curves of hybrid composite samples with fiber configurations [C/C/F/F/C/C], a) Dry condition b) after hydro aging effect

| Laminate | Fiber configurations | Dry samples Tensile strength (MPa) | Dry samples Maximum strain (%) | Immersed samples Tensile strength (MPa) | Immersed samples Maximum strain (%) |
|----------|----------------------|-----------------------------------|-------------------------------|----------------------------------------|-----------------------------------|
| F        | [F/F/F/F/F/F]        | 14 ± 5.35                         | 0.70                          | 4.32 ± 5.45                            | 1.30                              |
| S        | [S/S/S/S/S/S]        | 10.25 ± 5.46                      | 0.60                          | 2.19 ± 5.22                            | 1                                 |
| C        | [C/C/C/C/C/C]        | 86 ± 3.16                         | 3.03                          | 83 ± 3.22                              | 3.12                              |
| G        | [G/G/G/G/G/G]        | 25 ± 4.78                         | 1.80                          | 20 ± 4.80                              | 2                                 |
| L1       | [C/C/F/F/C/C]        | 73 ± 3.16                         | 2                             | 61 ± 3.22                              | 2.6                               |
| L2       | [F/F/C/C/F/F]        | 31 ± 4.26                         | 1.20                          | 18 ± 4.37                              | 1.80                              |
| L3       | [C/C/S/S/C/C]        | 63 ± 3.35                         | 2.5                           | 51 ± 3.48                              | 2.70                              |
| L4       | [S/S/C/C/S/S]        | 21 ± 5.05                         | 1.10                          | 11 ± 5.20                              | 1.60                              |
| L5       | [C/G/S/F/G/C]        | 57 ± 3.57                         | 2.30                          | 48 ± 3.65                              | 2.45                              |
| L6       | [G/C/S/F/C/G]        | 40 ± 4.06                         | 1.50                          | 35 ± 4.11                              | 1.70                              |
| L7       | [S/F/G/C/F/S]        | 20 ± 4.56                         | 1.35                          | 9.43 ± 4.61                            | 1.73                              |
| L8       | [F/S/C/G/S/F]        | 27 ± 5.17                         | 1.40                          | 16 ± 5.23                              | 1.80                              |
the response of water uptake for laminate L2 is high because of the presence of flax at outer layers so the change in tensile strength and maximum strain after hydro aging is evident. The tensile strength of laminate L3 and L4 has low value as compared to laminate L1 and L2, respectively. This may be attributed to the difference in tensile strength between flax and sisal fibers. The tensile strength of laminate L5 reaches 66% of laminate (C). This is because of the arrangement between the natural and synthetic fibers, where the hybridization by an adding sisal and flax fabrics at the middle portion (core) gives lower modulus and toughness of the laminate. However, the enhancement in the tensile strength of laminate L8 reaches 93% and 163% as compared to laminate F and laminate S, respectively. This is due to the positioning of the carbon and glass fabrics in the core which gives high strength and stiffness of this laminate.

The non-hybrid composite with laminate (C) before hydro aging gives the maximum tensile strength as compared to all other samples. The enhancement in the tensile strength can be attributed to the good arrangement as well as bonding between the resin matrix and fiber at the interface which leads to an increase in the tensile strength. The response of water uptake for laminate C can be neglected because of the hydrophobic nature of carbon; so, the change in tensile strength and maximum strain do not exceed 3.5 and 6.5%, respectively. Nevertheless, the small decreasing in tensile strength can be attributed to the slight degradation at the interface between the resin matrix and fiber by the small water absorption effects, where confirms with the water absorption and swelling thickness results.

3.3. Microstructure

Figures 4 to 6 illustrate the SEM images of the hybrid composites L1, L2, and L6, respectively before and after hydro aging. It can be clear from Figure 4a that the laminate L1 with fiber configuration (C/C/F/F/C/C) has good adhesion between the resin matrix and fiber for dry specimens. After hydro aging effect, the carbon fibers act a barrier between the water molecules and the flax fibers so the water molecules between them are very small causing lower interfacial deboning, breakdown and small swelling of fibers as shown in Figure 4b.

It is clearly seen in Figure 5a that the carbon fiber pull-outs and high void content can be noticed for the dry specimens after tensile test. As a result, the high void content in the hybrid composites with fiber configuration (F/F/C/C/F/F) causes the degradation in laminate and also gives poor interfacial adhesion bonding between the fiber and matrix. Therefore, the tensile strength of this laminate was reduced. In addition, loss contact between the resin matrix and fiber are detected after hydro aging as shown in Figure 5(b). As seen in

Figure 4. SEM image of hybrid composite with fiber configuration [C/C/F/F/C/C]: (a) dry specimen and (b) after hydro aging effect

Figure 5. SEM image of hybrid composite with fiber configuration [F/F/C/C/F/F]: (a) dry specimen and (b) after hydro aging effect

Figure 6. SEM image of hybrid composite with fiber configuration laminate L6 [G/C/F/S/C/G]: (a) dry specimen and (b) after hydro aging effect

Figure 6a, there are many voids at the interface between the matrix and fibers in laminate L6 with fiber configuration (G/C/F/S/C/G) for the dry specimens which cause a weak interfacial adhesion between the matrix resin and fibers. After hydro aging, the fiber was bundled and swelled as shown in Figure 6b due to the water absorption effects.

4. CONCLUSIONS

In this paper, the effect of various fiber configurations of sisal, flax, glass, and carbon fibers reinforced polyester resin hybrid composites on tensile properties before and after the hydro aging were investigated. From the current study, the following conclusions are summarized:
- Laminate (S) with fiber configuration [S/S/S/S/S] absorbs more amount of water up to 14 % while the laminate (C) with fiber configuration [C/C/C/C/C] absorbs minimum amount of water up to 1.45%.

- The laminate (C) with fiber configuration [C/C/C/C/C/C] exhibited maximum tensile strength (86 MPa) compared to all laminates and slight deterioration after hydro aging.

- The tensile strength of laminate L1 with fiber configuration [C/C/C/C/C] is closest to the obtained value for laminate (C).

- The deterioration in tensile strength after hydro aging is observed for hybrid-composites with natural fiber at outer layers, but slightly observed for hybrid-composites with synthetic fiber at outer layers.

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چکیده
تأثیر چرب آب و ضخامت پروری بر خواص کششی پایه‌های آبی کریستال دار، سیال / کرین / سیال / کرین / کرین / سیال / کریستال دار، شده توسط کامپوزیت‌های ترکیبی متین بر پایه آب در اثر بررسی آزمون‌های مختلف، با استفاده از روش‌های مختلف شده است. آزمون جذب آب با غوطه وری [C / C / C / C / C / C / C / C / C / C / C / C / C / C / C / C] آزمون‌ها در آب مقطر برای مدت زمان‌های مختلف تا 144 ساعت انجام شد. نتایج آزمون‌ها بیانگر تأثیر این در جذب آب برای پیکریزی‌های [C / C / C / C / C / C / C / C / C / C / C / C / C / C / C / C] غوطه ور شدن در آب مقطر بررسی شد. این نتایج نشان داد که استحکام کششی پیکریزی‌های [C / C / C / F / F / F / C / C / C / C / C / C / C / C / C / C] نرخ شیب مورفولوژی سطح مقفل نوع کامپوزیت با استفاده از میکروسکوپ الکترونی روبنی (SEM) برابر از بازیابی و استحکام ماتریس پیش و پس از پر شدن در آب بررسی شد. کاهش مقاومت کششی به دلیل نفوذ آب برای کامپوزیت هیریدی با الاف مصنوعی در لوله‌های زیرین است، اما برای کامپوزیت ترکیبی با الاف مصنوعی در لوله‌های زیرین رشد است.