

Thermomechanical Behavior of Methylene Diphenyl Diisocyanate-Bonded Flax/Glass Woven Fabric Reinforced Laminated Composites

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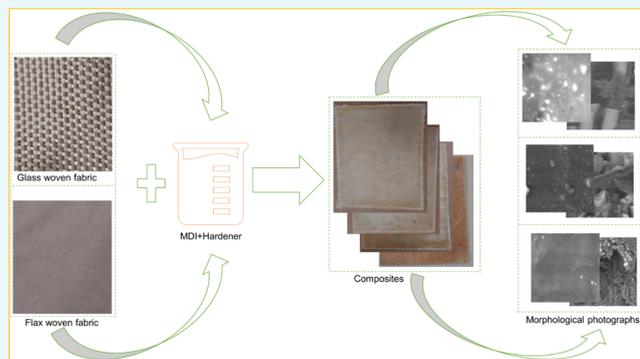
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ABSTRACT: The development of sustainable and innovative products through solving the constantly raising demands of end users is one of the significant parts of research and development. Herein, the development of a green composite is reported with the reinforcement of naturally originated flax and artificial glass woven fabrics through incorporating with the methylene diphenyl diisocyanate (MDI) resin. The glass fabrics were treated with silane and flax fabrics by using NaOH before the composite production to increase the affinity of fibers toward the resin. Composite panels were developed with four different ratios of glass and flax woven fabric reinforcement (100/0, 83.33/16.67, 50/50, and 0/100) to investigate their performance with the MDI resin. The composites were characterized by tensile and flexural analysis to investigate the mechanical performances. The thermogravimetric characteristics of the composites were examined for checking the thermal stability of the produced composites. The surface morphology was investigated for observing the surfaces of the composites before and after applying tensile loads. Scanning electron microscopy (SEM) deployed EDX linear scanning was used for ensuring about the signals of different chemical constituents into the matrix. Fourier transform infrared spectroscopy (FTIR) was conducted for finding out the fingerprint of the chemical elements of the produced composites. Besides, the water absorption and moisture content tests were also conducted to examine the moisture absorption by the pure glass, flax, and hybrid composites. Further, statistical analysis of variances was performed to test the significance of the differences in the mechanical properties of the individual types of the composites developed. For investigating the relationship between the proportion of woven glass fabric in the reinforcement and the mechanical properties, regression analysis was used. The ANOVA test was also examined for checking the significance of the mechanical properties of the composites.



INTRODUCTION

Naturally originated fiber reinforced composite materials have drawn significant attention since the last few decades for commercial applications in the field of automobiles, construction, biomedicine, packaging, and aerospace.¹ Natural fibers are showing greater potentiality to reduce the dependency on artificial fiber-based composites/products in terms of sustainable features. The composite materials developed from renewable and green natural fibers are termed as biocomposite, which is also getting more popularity among the researchers and industrialists nowadays. There are several plant-based fibers such as flax,² hemp,³ coir,⁴ ramie,⁵ jute,⁶ kenaf,⁷ and so forth drawing attention to be potential raw materials for biocomposite manufacturing. The outstanding features of natural fibers such as biodegradability, renewability, and recyclability have made them prominent raw materials in the emerging composite world.⁸

Polyurethane (PU)-based resins are derived from vegetable-/petroleum-based oils⁹ having widespread applications such as coating materials, adhesives, automotive parts, and

different infrastructures. However, they are more popular for producing durable, lightweight, and cost-effective composite products.¹⁰ PU is highly compatible with plant-based fibers as the isocyanate group present in PU interacts with the –OH (hydroxyl group) of natural fibers.^{9,11} Besides, PU also exhibits some outstanding properties such as zero volatile organic compound emission, comparatively cheaper price, feasible processing technology, higher reactivity, and so on.¹² In this regard, MDI is widely used for particle board productions by manufacturers. However, it is not yet getting attention for laminated multilayered hybrid composite productions. Therefore, research studies are needed to find out novel routes of

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Table 1. Stacking Sequence, Thickness, and Density of Developed Hybrid Composites (G1, GF2, GF3, and F4)^a

laminates	sequence of stacking	thickness (mm)	ultimate thickness (mm)	density (kg/m ³)
G1 (100% glass)	G,G,G,G,G,G	1.56 (0.012)	1.06 (0.004)	1727.62 (29.01)
GF2 (83.33% glass/16.67% flax)	G,G,F,G,G,G	1.9 (0.005)	1.52 (0.014)	1401.32 (60.28)
GF3 (50% glass/50% flax)	G,F,G,F,G,F	2.58 (0.007)	2.23 (0.007)	1091.53 (146.7)
F4 (100% flax)	F,F,F,F,F,F	3.6 (0.19)	2.51 (0.005)	1195.02 (32.10)

^aComposites were developed with different densities and thickness. (Mean values with standard deviations in parentheses.) G—glass, F—flax.

producing economical and feasible laminated composite panel manufacturing.

Although there are enormous advantages of using plant fibers, still there are some limitations for the inherent structural properties. The polymeric resins are hydrophobic and nonpolar while the natural fibers contain –OH groups, so they are hydrophilic and polar in nature.¹³ The compatibility problem hampers the bonding when these two different properties of materials are mixed together in the polymeric matrix. The mechanical properties of biocomposites are affected by this kind of incompatibility because the affinity toward the water is enhanced. The incorporation of synthetic fibers such as glass or carbon with the natural fibers (such as flax, hemp, kenaf, coir, jute, and so on) could overcome this challenge. Kumar et al.¹⁷ have reported that when the hybrid composites are developed through reinforcing glass with flax, the tensile strength was increased from 85.6 to 143.21 MPa. The same study¹⁷ also found a significant improvement in impact strength and flexural properties of the hybrid composites. Bajpai et al.¹⁴ described in another study that the reinforcement of three-layered jute and single-layer glass with epoxy resin provided the highest flexural strength (107.78 MPa) and impact strength (72.24 J/m), making it useful for safety helmets in industrial applications.

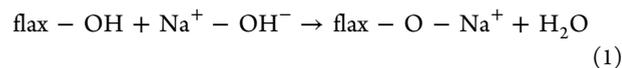
Besides, composites made from woven fabrics are also getting importance to the researchers and industrialists for better comfort, dimensional stability, strength, stiffness, lower fabrication cost, and so on.¹⁵ Glass and flax woven fabrics were selected for this research. Some studies also revealed that pretreatment of both the flax and glass fibers could improve the interfacial bonding in the polymeric composite;^{1a,16} so both the flax and glass woven fabrics were treated with NaOH and silane, respectively, before producing the hybrid composite. Different percentages of flax and glass woven fabrics were used as a reinforcement to fabricate the composites through reinforcing with the MDI matrix. According to our knowledge, no research studies have been conducted yet on pretreated flax and glass woven fabric-based laminated composites reinforced with the MDI polymeric resin. As MDI is used widely in industrial particle board manufacturing process, we hope our current research could facilitate the bulk productions of glass/flax reinforced MDI composites. The mechanical, physical, morphological, thermal, and statistical analyses have provided significant information on the produced hybrid composites.

MATERIALS AND METHODS

Materials. The flax woven fabrics (article number: LV06506, density: 230 g/m², composition: 100% flax, Twill structure) were purchased from Málitex (Pecs, Hungary). The glass woven fabric [with a measured density of 255 g/m², 100% glass, and plain weave structure (grid size 4.4 × 4.2 mm²)] was procured from Tolnatek located in Tolna, Hungary. The alkaline NaOH was bought from VWR International Kft. (Debrecen, Hungary) and vinyltrimethox-

ysilane, C₅H₁₂O₃Si (L#MKBZ5796V, 98%, molecular weight 148.23 g/mol), from Sigma-Aldrich Co. (St. Luis, MO, USA). The MDI (Ongronat XP-1161) was collected from Borsodchem Zrt. (Kazincbarcika, Hungary). The hardener (H-240) was collected from SC Kronospan Sebes SA, Hungary. The hardener (10%) and MDI (90%) were mixed together to make the adhesive paste for applying onto the stacked fabric layers into the composite panels. The Formula Five mold release wax was procured from Novia (Hungary) to use as a coating material between the composites and Teflon paper to avoid stickiness of resin with the Teflon.

Methods. Initially, the flax woven fabrics were pretreated with 0.5% NaOH solution (the material liquor ratio was 1/20) for 30 min at a temperature of 100 °C to enhance the interaction of fibers with polymeric resin. The reaction mechanism is shown in equation 1. The glass fabrics were treated by vinyltrimethoxysilane at room temperature for 30 min (the material liquor ratio was 1/10). After the pretreatment, the fabrics were rinsed and washed three times to remove the alkaline mucus, vinyltrimethoxysilane solutions, and other impurities from the surface. The fabric samples were then dried in an oven dryer at 60 °C for 6 min. After that, six layers (Table 1) of glass/flax woven fabrics (G1, GF2, GF3, and F4) coated with MDI resin were stacked up by hand-layup method. The ratio of MD resin and hardener was 10:1. The sequence of layers in the laminates were (G,G,G,G,G,G/G,G,F,G,G,G/G,F,G,F,G,F/F,F,F,F,F,F) with thicknesses of 1.56, 1.9, 2.58, and 3.6 mm for G1, GF2, GF3, and F4 composites, respectively. The produced laminates were pressed (3.5 MPa pressure) by a pressing machine for 15 min at room temperature. Later on, the composites were then cured for 24 h at ambient conditions.



Characterization of the Composites. The tensile and flexural properties of the produced composites were measured by using the universal testing equipment Instron 4208 (Instron corporation, USA). The tensile test was conducted as per the EN 310 procedures, whereas flexural properties were also adopted by the EN 310 standard. Six samples from each composite were selected for conducting the test. The FTIR characterization of the composites was performed using a FTIR-6300 (Jasco, Japan) spectrometer at 4000–500 cm⁻¹. The morphological investigation was performed by using an SEM equipment (SEM, S 3400N, Hitachi, Japan) at a 15.0 kV voltage within the magnifications of 2000 times and 1000 times. Thermogravimetric analysis (TGA) and derivative TG (DTG) analysis were conducted using a Themys thermal analyzer (Setaram Instrumentation, France) within 25 to 850 °C at a 10 °C/min temperature gradient under nitrogen (N₂) conditioning. The water absorbency was tested at 2, 24, and 240 h time intervals as per the MSZ 13336-4:13379 method, which is a Hungarian national standard. Samples of 50 mm by

50 mm dimensions were prepared to execute this test. The composite samples were emerged into 30 mm depth of water. The moisture content of the composite boards was investigated in line with the EN 322 methods. The dimensions of the samples were kept the same (50 mm by 50 mm).

RESULTS AND DISCUSSION

Mechanical Properties of Composites. The tensile properties of composites are highly influential to assess the strength of the produced materials. The tensile features of the manufactured composites (Figure 1) are given in Table 2. The

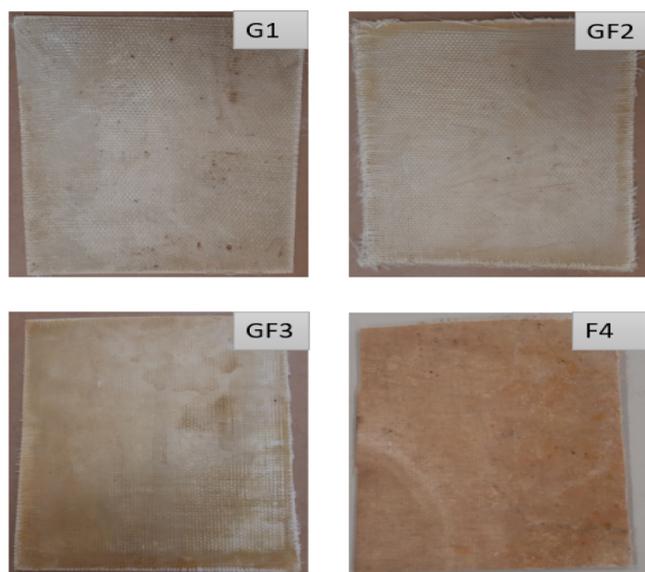


Figure 1. Representation of hybrid biocomposites: G1 (pure glass composite), GF2 (hybrid composite from glass/flax), GF3 hybrid composite from glass/flax, and F4 (pure flax composite).

Table 2. Tensile and Flexural Properties of Produced Composites (G1, GF2, GF3, and F4)^a

laminated composites	tensile strength (MPa)	Youngs modulus, E (GPa)	flexural strength (MPa)	bending modulus, MOE (GPa)
G1 (100% glass)	78.61 (8.2)	6.82 (0.15)	211.9 (17.9)	54.4 (1.8)
GF2 (75% glass/25% flax)	69.63 (2.77)	7.59 (0.58)	147.7 (18.5)	40.4 (7.8)
GF3 (50% glass/50% flax)	49.44 (2.05)	6.73 (0.52)	58.9 (9.5)	39.9 (4)
F4 (100% flax)	21.19 (1.59)	2.54 (0.15)	43.9 (3.5)	3.9 (0.8)
coefficient of determinations (R^2)	0.66	0.57	0.41	0.89

^aPure glass and hybrid composites exhibited better mechanical performances in contrast to natural flax. (Means with standard deviations in parentheses.)

tensile strengths of pure glass (G1), glass/flax (GF2 and GF3), and pure flax (F4) reinforced MDI composites take the values of 78.61 (8.2), 69.63 (2.77), 49.44 (2.05), and 21.19 (1.59) MPa, respectively. While GF2 (hybrid composite) exhibited the highest tensile modulus [7.59 (0.58) GPa], pure flax reinforced composite exhibited the lowest modulus and tensile strength too. It is found from Table 2 that synthetic glass reinforced composites are stronger than naturally originated flax reinforced composites. However, the laminated composites

of glass/flax composites provided more strength than flax itself. Besides, it is also observed that the more loading of glass with flax enhances the composite strength. A similar phenomenon was also described by other researchers for different designs of fabric stackings (glass/flax)¹⁷ with vinyl ester used as the polymeric resin.

The flexural strengths followed the same trend as the tensile characteristics. The perceived flexural strengths were 211.9 (17.9), 147.7 (18.5), 58.9 (9.5), and 43.9 (3.5) MPa (Table 2), respectively, for G1, GF2, GF3, and F4 composites. Besides, Youngs modulus followed a similar pattern of flexural strengths (54.4 (1.8), 40.4 (7.8), 39.9 (4.4), and 3.9 (0.8) MPa). As expected, naturally originated flax reinforced composites provided the lowest strengths while those with pure glass reinforcement produced the highest values. However, the strength values started to increase with the incorporation of more glass fiber loading into the hybrid composites.¹⁷ Likewise, glass reinforced MDI composites provided higher bending modulus, with higher tendency to bend without breaking in contrast to flax reinforced composites.

The load versus displacement behavior of the test pieces is illustrated in Figure 2a,b both for tensile and flexural tests. In the case of tensile displacements, all the curves showed a linear region initially; a nonlinear region appeared whenever the cracking happened. Composite G1 attained a load of approximately 2000 N in the linear range in tension. After exhibiting a maximum load of around 2650 N at extended delamination, the load started to decline with the increase of displacement until failure. The decline of highest load depends on the onset of delamination and development of cracking in the laminates. In the case of composites GF2 and GF3, the highest observed loads in the linear range were 1500 and 750 N, respectively. Linearity for F4 ended at 50 N, although load continued to increase with the increased delamination up to 500 N, then started to drop. In the course of flexural tests, the highest load attained in the linear range by the composites G1, GF2, GF3, and F4 was 26, 21, 14, and 7 N, respectively. Similar trends for load and displacement patterns were also discussed in some other studies.

Statistical Analysis for Mechanical Performances.

Regression analyses of all the composites' mechanical performances were conducted in terms of glass fiber proportion in the composites. The R^2 values (Table 2) for all the composites are higher than 0.57, except for flexural strength with $R^2 = 0.41$. It seems that the presence of glass fiber results in higher mechanical performances of all the composites. The p values for tensile strength and tensile modulus (Tables 3–6) stand far less than 0.05 except for flexural strength and modulus, where the intercept parameter of the regression equation did not prove to be significant; see the corresponding p values shown in bold in Tables 5 and 6. These results support the existence of significant effects of glass fiber proportion on composite properties with slopes higher for strength than for modulus of elasticity.

The mechanical features of the produced composites were further analyzed conducting one-way ANOVA with the type of composites as a categorical factor. Overall F -tests of significance for all the four mechanical properties provided evidence of effect of all the composite types. For pairwise comparisons of the four types, Newman–Keuls tests (Tables 7–10) were used because the statistical assumptions of ANOVA were not always met. These tests showed that the

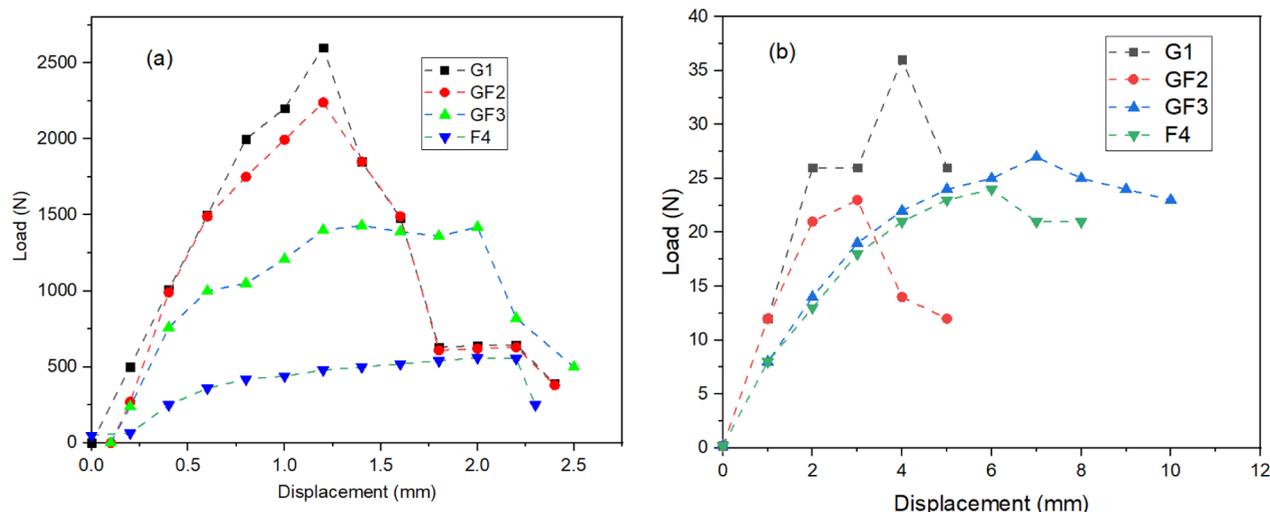


Figure 2. Load versus displacement graphs for G1, GF2, GF3, and F4 composites: (a) tensile test and (b) flexural test.

Table 3. Regression Analysis for Tensile Strength in Terms of Glass Fiber Compositions on Different Composites

effects	tensile strength parameter	tensile strength standard error	tensile strength t	tensile strength p
intercept	27.95864	6.970652	4.010908	0.002474
composition	0.43630	0.100053	4.360751	0.001419

Table 4. Regression Analysis for Tensile Modulus in Terms of Glass Fiber Compositions on Different Composites

effects	tensile modulus parameter	tensile modulus standard error	tensile modulus t	tensile modulus p
intercept	3.607239	0.771544	4.675349	0.000874
composition	0.040050	0.011074	3.616449	0.004717

Table 5. Regression Analysis for Flexural Strength in Terms of Glass Fiber Compositions on Different Composites

effects	flexural strength parameter	flexural strength standard error	flexural strength t	flexural strength p
intercept	44.25353	30.57797	1.447236	0.178442
composition	1.16265	0.43890	2.649016	0.024354

Table 6. Regression Analysis for Flexural Modulus in Terms of Glass Fiber Compositions on Different Composites

effects	flexural modulus parameter	flexural modulus standard error	flexural modulus t	flexural modulus p
intercept	7.653454	3.642373	2.101228	0.061949
composition	0.470820	0.052280	9.005672	0.000004

Table 7. Newman–Keuls Test Results for Tensile Strength in Terms of Different Composites (G1, GF2, GF3, and F4)

composites	{1}	{2}	{3}	{4}
G1		0.000201	0.003495	0.000231
GF2	0.000201		0.000223	0.000223
GF3	0.003495	0.000223		0.000201
F4	0.000231	0.000223	0.000201	

Table 8. Newman–Keuls Test Results for Tensile Modulus in Terms of Different Composites (G1, GF2, GF3, and F4)

composites	{1}	{2}	{3}	{4}
G1		0.500496	0.000352	0.000201
GF2	0.500496		0.000367	0.000223
GF3	0.000352	0.000367		0.000231
F4	0.000201	0.000223	0.000231	

Table 9. Newman–Keuls Test Results for Flexural Strength in Terms of Different Composites (G1, GF2, GF3, and F4)

composites	{1}	{2}	{3}	{4}
G1		0.000201	0.000283	0.000231
GF2	0.000201		0.001390	0.424337
GF3	0.000283	0.001390		0.001292
F4	0.000231	0.424337	0.001292	

Table 10. Newman–Keuls Test Results for Flexural Modulus in Terms of Different Composites (G1, GF2, GF3, and F4)

composites	{1}	{2}	{3}	{4}
G1		0.015094	0.011749	0.000233
GF2	0.015094		0.454250	0.000245
GF3	0.011749	0.454250		0.000250
F4	0.000233	0.000245	0.000250	

strength properties of the different composites are significantly different as the p values are less than the assumed level of significance of 0.05. However, the modulus of elasticity values in two cases shown in bold in the Tables 5 and 6 do not exhibit the significant difference; these are tensile modulus values for composites G1 and GF2 as well as flexural modulus for GF2 and GF3.

Morphological Studies of Composites. The SEM photographs clearly exhibit the uniform MDI polymer distributions on the respective glass and flax woven fabric reinforced composites. Although the stacked fibers cannot be observed in Figure 3(a₂,a₃,c₂,c₃,e₂,e₃,g₂,g₃) for strong polymeric overlapping/coating on the surface but could be clearly seen on the fractured surfaces of the composites, see Figure 4(b₂,b₃,d₂,d₃,f₂,f₃,h₂,h₃). The surfaces of the composites are flat,

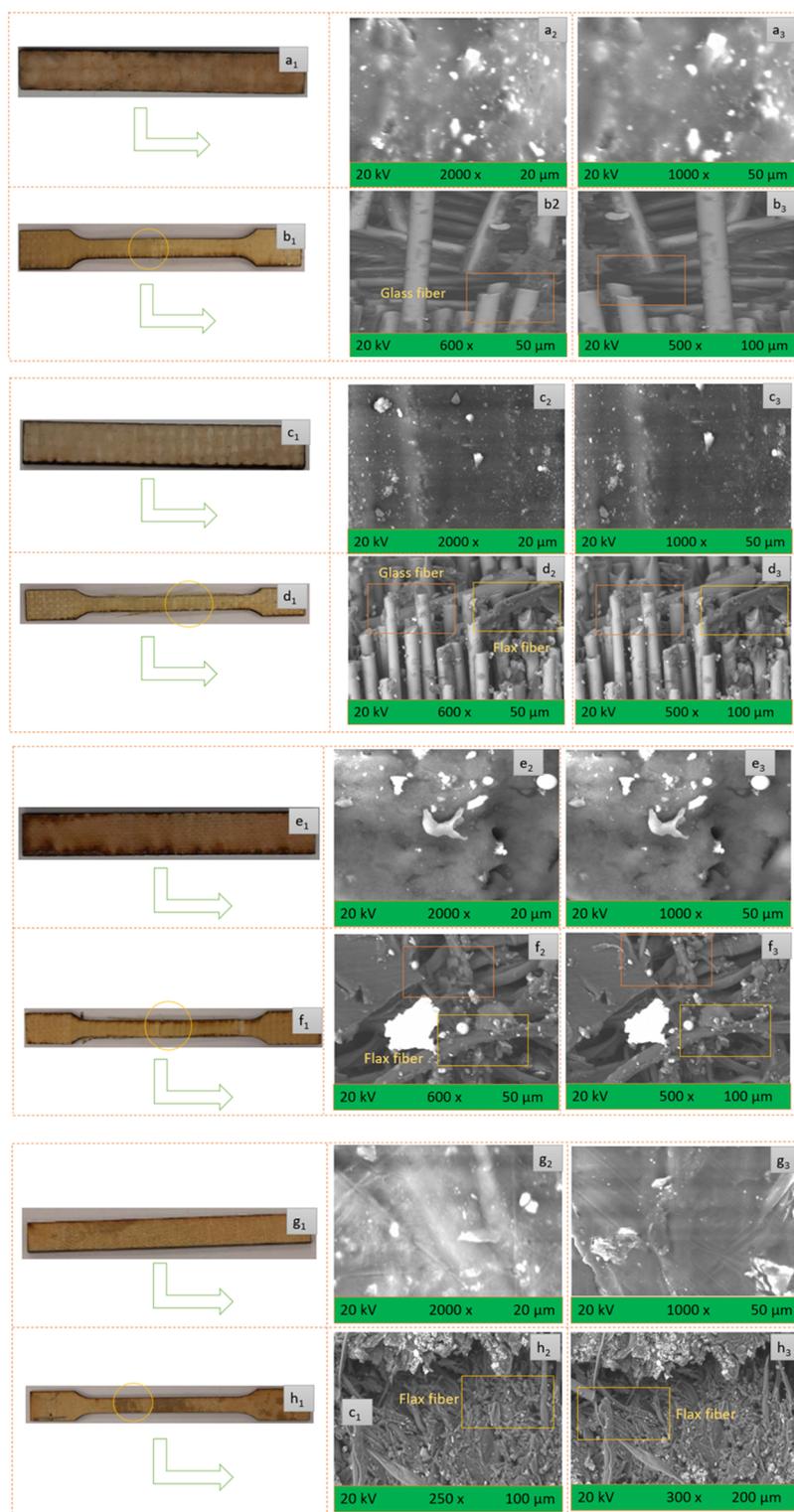


Figure 3. Morphological characterization of hybrid composites (a_1, c_1, e_1, g_1) for flexural test samples. Morphological characterization of fractured hybrid composites (b_1, d_1, f_1, h_1) for tensile test samples. Flat and uniform distribution of MDI resin on composites with reinforcement of pure glass (a_2, a_3), hybrid flax/glass (c_2, c_3, e_2, e_3), and pure flax (g_2, g_3) composites at different magnifications. Holes appeared for incompatibility between the MDI resin and woven fabrics (e_2, e_3). Fractured composites after applying tensile load on composites with reinforcement of pure glass (b_2, b_3), hybrid flax/glass (d_2, d_3, f_2, f_3), and pure flax (h_2, h_3) composites at different magnifications. Holes for incompatibility between the MDI resin and woven fabrics are presented through (e_2, e_3). SEM analysis of composites.

smooth, and uniform, which indicates the perfect bonding of MDI resin with the glass and flax woven fabrics. However, few holes could also be observed, which is indicating the weaker adhesion¹⁸ between the fabric and resin into the matrix system.

Such kind of holes were appeared only for Figure 3(e_2, e_3) showing test pieces of 50% flax and 50% glass with MDI. The surfaces of 100% glass, 100% flax, or 83.33% glass/16.87% flax reinforced composites did not display any weak adhesion/

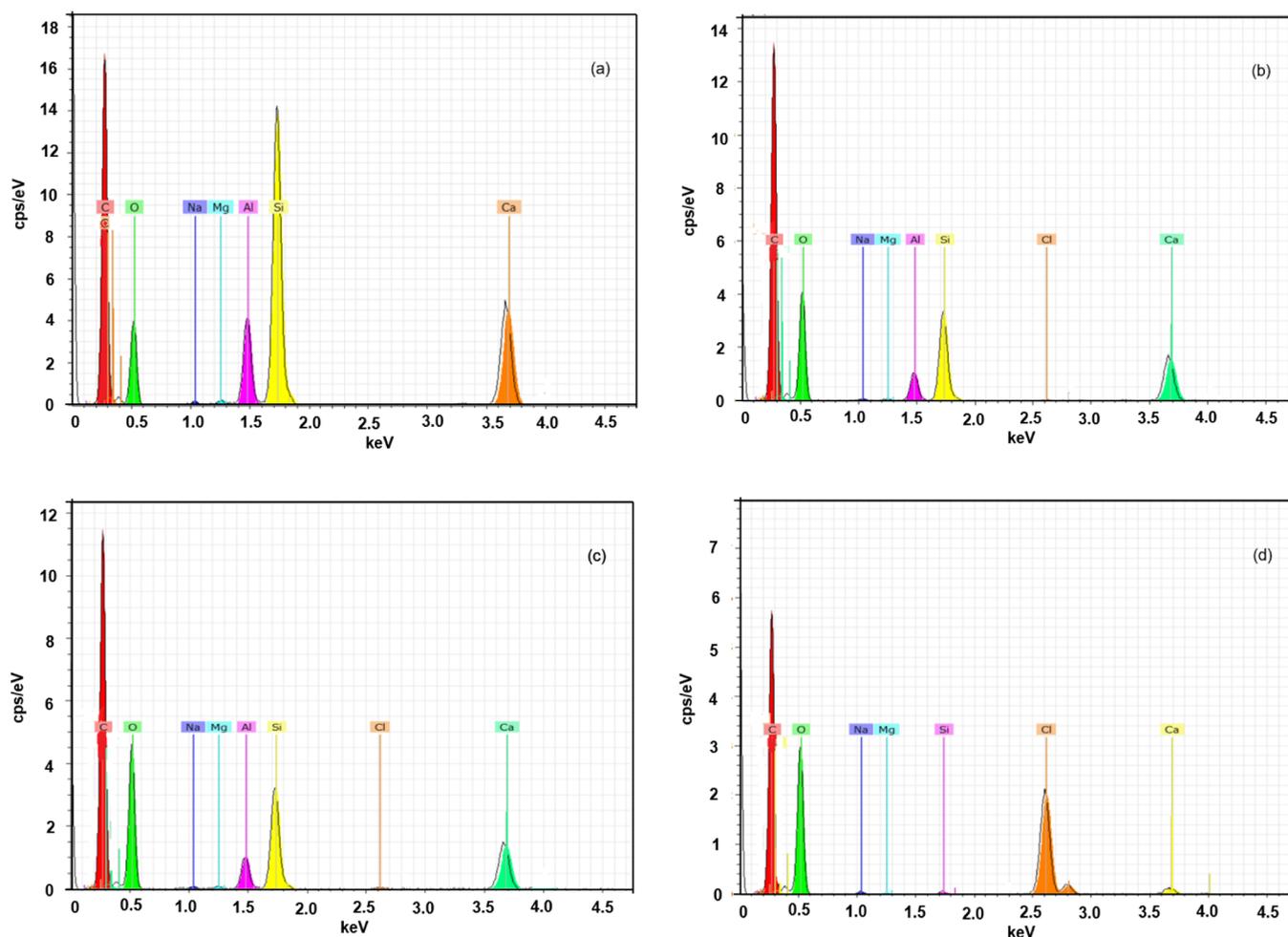


Figure 4. EDX spectrum of the composites (a) G1, (b) GF2, (c) GF3, and (d) F4. The presence of glass fiber is observed through the presence of Si (a–c), while the presence of flax is confirmed by the presence of C and O (b–d).

interactions. However in the case of Figure 4 ($b_2, b_3, d_2, d_3, f_2, f_3, h_2, h_3$), there are explicit breakage and presence of fibers (flax and glass marked through red and yellow color, respectively). Besides, as the glass fabrics were treated with silane, it helped to form an interpenetrated network between the silane-treated glass woven fabric and the MDI resin.¹⁹

EDX Analysis of Composites. The energy-dispersive X-ray (EDX) spectra provide the nature of glass, flax, and polymers embedded into the matrix. It is clearly observed from Figure 4 that silicon (Si) is one of the most significant chemical compound indicating the presence of glass into the composite (Figure 4a). Besides, the detection of calcium (Ca), magnesium (Mg), and aluminum (Al) also confirm the presence of different oxides into the glass woven fabrics of the composites (Figure 4a–c). At the same time, the broad peak of C and oxygen (O) denotes the good bonding of MDI. In the case of pure flax reinforced composite (Figure 4d), there is no peak observed for silicon (Si) but there are peaks for C and O (strong peaks for natural fibers) and chlorine (Cl). On the contrary, Cl did not show any peaks for G1 composites, while they are present in the spectra of GF2, GF3, and F4 composites. Presumably, this difference can be attributed to the presence of flax. The presence of C and O is detected for all the four composites, which may be an indication of the bond developed between the woven fabric and the MDI

polymeric resin. Thus, the EDX spectrum confirms the successful reinforcement of flax and glass with MDI.

FTIR Analysis of Composites. The chemical structures of MDI-based glass/flax reinforced composites were investigated by FTIR analysis. The broad absorption bands (Figure 5d) at 3331 cm^{-1} indicate the presence of cellulosic structure ($-\text{OH}$ unit) for flax fiber reinforced MDI composites. Besides, the peaks at 2851 and 2920 cm^{-1} are related to the existence of CH_2 groups in the fiber. The presence of cellulosic structure in the composite could be further confirmed by the peaks at 1654 and 1054 cm^{-1} . A similar phenomenon was described by another study.²⁰ The broad bands ranging from 1017 to 3340 cm^{-1} (Figure 5a) represent the glassy material-based composites.²¹ Specifically, the peak at 1017 cm^{-1} is responsible for the $\text{Si}-\text{O}-\text{Si}$ group and 1409 cm^{-1} for other types of oxides (boron oxide, aluminum oxide, calcium oxide, and so on), which are the specific chemical compositions of the glass fiber.²² There are also a few weaker bonds found at 1654 and 1409 cm^{-1} , which are related to water adsorptions occurring during the composite manufacturing process.²¹ In Figure 5 b,c, the peaks around 3340 cm^{-1} may be attributed to the bonding between the external hydrogen in glassy structures and cellulosic structures (flax) $-\text{OH}$ groups.²³ Besides, the peaks around 2850 cm^{-1} (Figure 5a–c) may be related to the treatment of glass surface with the silane. Further, the bonding of MDI with the cellulosic structure is also further

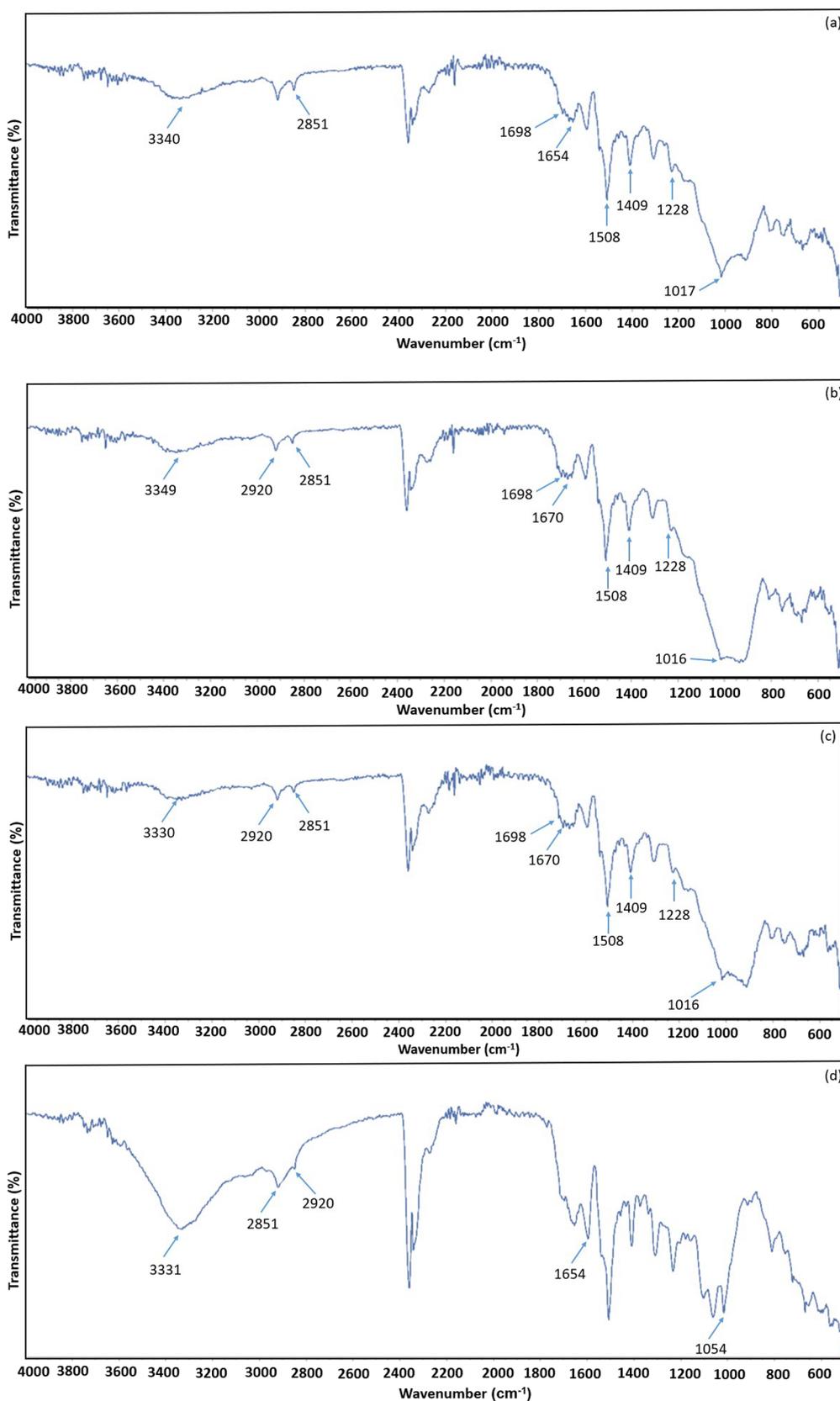


Figure 5. FTIR analysis of composites (G1, GF2, GF3, and F4). (a) Pure glass composite, (b,c) hybrid composites, and (d) pure flax.

confirmed by the peaks at 1508 cm^{-1} (CN–H, urethane holding secondary amide), 1698 cm^{-1} (carbonyl urethane,

–C=O), and 1228 cm^{-1} (–C–O–C , ether urethane) as can be seen in Figure 5b–d.²⁴

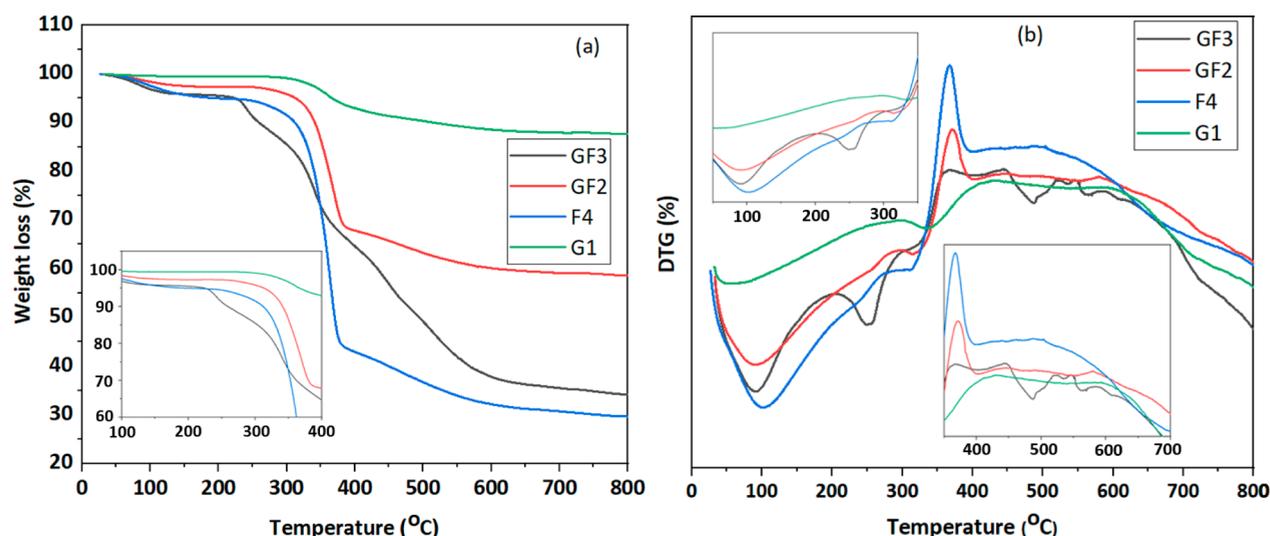


Figure 6. Thermal behavior of composites (G1, GF2, GF3, and F4): (a) TGA and (b) DTG. Pure glass and hybrid glass composites are more stable than natural flax reinforced composite (b–d).

Thermogravimetric Analysis. The thermal properties of glass/flax reinforced MDI composites are shown in Figure 6. Initially, all the composites except G1 displayed significant weight loss due to moisture evaporation probably because of the presence of MDI polymer or flax and glass fibrous material, as shown in Figure 6a. Weight loss is gradually increasing with the increase of flax fiber content in the composites. Temperatures belonging to 5% weight loss of the composites are provided in Table 11. As illustrated in Figure 6, the

Table 11. Onset and Maximum Temperature of Hybrid Composites (G1, GF2, GF3, and F4)^a

laminated hybrid composites	T_{onset} (°C)	T_{max} (°C)	residues at 650 °C (weight %)
G1	359	650	87.99
GF2	310	650	36.01
GF3	197	650	58.78
F4	162	650	30.82

^aGlass reinforced composites exhibit more residues compared to flax and hybrid composites.

maximum weight losses (10–60%) occurred at temperatures ranging from 315 to 450 °C. Besides, the residues of the composites G1, GF2, GF3, and F4 were amounted to 87.99, 36.01, 58.78, and 30.82%, respectively. In summary, it could be stated that the combustion in the case of glass reinforcement is of lower level than in the case of reinforcements containing flax. Also, more residues are found for glass as compared to flax. A similar phenomenon was also described by other studies on glass with natural fiber-based laminated hybrid composites.²⁵

DTG curves in Figure 6b, showing some small peaks ranging from 250 to 340 °C, which are related to the decomposition of organic components of hybrid composites. The second stage degradation (367–372 °C) is associated with the hemicellulose decompositions from flax constituents. Cellulose degradation is indicated by the peaks within the range of 490–582 °C. The variation in temperatures is caused by the different proportions of glass fiber present in the composite stackings (GF2 and GF3). A similar study was conducted by Atiqah et al.^{25c} for thermoplastic polymer-based sugar palm/

glass composites. The DTG curves have clearly displayed the decomposition pattern of glass/flax reinforced MDI polymeric composites.

Physical Properties of Composites. The water absorption by composition of pure flax, glass, and hybrid reinforcement is demonstrated in Figure 7a. As expected, the naturally originated flax reinforced composites exhibited higher water absorption than reinforced with pure glass. This is because the natural fibers contain hydrophilic groups (–OH, –COOH, –CO, and –NH₂),^{25a} whereas the synthetic glass fibers do not. As a natural fiber, flax contains enormous amounts of –OH groups (also found by FTIR analysis). Therefore, the saturation point is higher for flax-based composites than glass. The saturation point of flax decreases with the loading of more glass woven fabrics into the hybrid composites. The sequence is G1 < GF2 < GF3 < F4. The water absorption was observed from 2, 24, and 240 h as illustrated in Figure 7a.

The moisture content of pure glass, flax, and hybrid composites also exhibited the same trend. The G1 (pure glass) sample absorbed the lowest moisture, whereas F4 (pure flax) attained the highest moisture content. The hybrid composites (GF2) attained a moisture content of 1.34 (0.32), 1.88 (0.29), and 2.15 (0.09)% after 2, 24, and 240 h, whereas GF3 showed 3.76 (0.08), 3.84 (0.33), and 3.97 (0.04)% moisture content within the same time period. Again, standard deviations are shown in parentheses. It is noticed that the moisture content of flax reinforced composites starts to decline with the increased loading of glass woven fabrics.

CONCLUSIONS

The fabrication of composites reinforced by laminated flax and glass woven fabric with the use of MDI polymeric resin was performed successfully. The glass woven fabric reinforcement with MDI resin provided highest flexural and tensile strengths, whereas the flax reinforced composites showed the lowest performance. However, when the loading of glass was increased into flax/glass reinforced hybrid composites, the strengths started to increase. The GF2 sample as a hybrid composites developed through reinforcing by natural and synthetic fibers together (83.33% glass and 16.87% flax) provided satisfactory mechanical performance [tensile strength

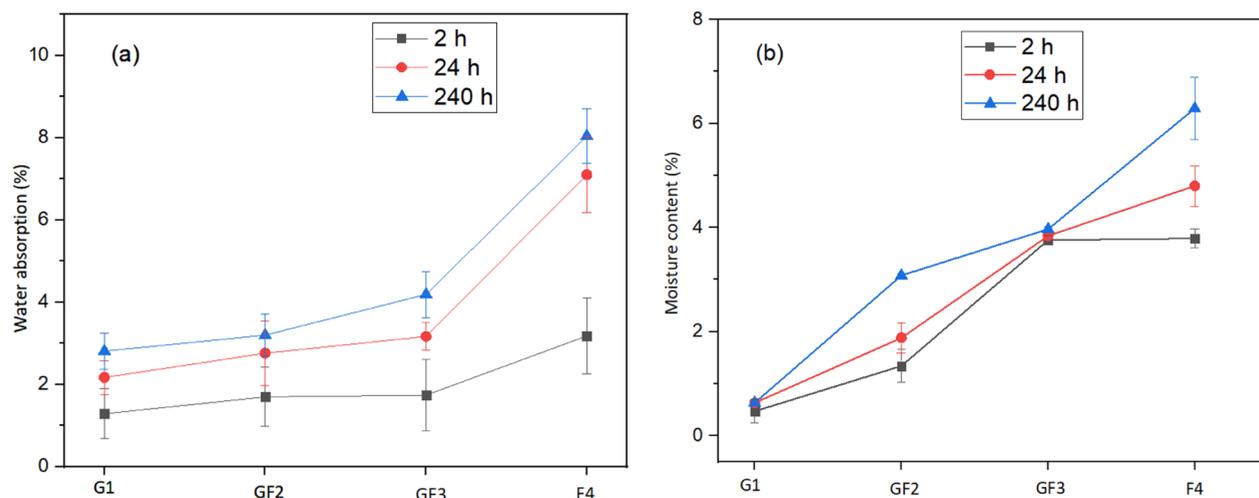


Figure 7. Water absorption (a) and moisture content (b) of composites (G1, GF2, GF3, and F4) within 2, 24, and 240 h time intervals. Natural flax reinforced composites absorb more water and exhibit higher moisture uptake compared to glass reinforced composites.

69.63 (2.77) MPa and flexural strength 147.7 (18.5) MPa]. The SEM micrographs also showed flat and uniform surfaces of the produced composites with homogeneous distribution of MDI resin into the woven fabric reinforced matrix. The EDX characterization of the composites confirmed the successful reinforcement of glass and flax woven fabrics with MDI resin through testifying their footprint of elemental compositions. The thermogram studies of the produced composites have proved the satisfactory thermal stability. The addition of glass on flax/glass hybrid composites also enhanced the thermal stability. The FTIR analysis provides the fingerprint of glass and flax fiber presence on the hybrid composites. The water absorption and moisture content investigation has shown that the natural flax reinforced composite contains higher moisture than that of pure glass. However, with the increased incorporation of synthetic glass, both the water absorption and moisture content started to decline. However, the incorporation of glass into the composites has significant influence on tensile strength, tensile modulus, and flexural modulus (regression analysis). The ANOVA test has further confirmed about the significance of mechanical properties with the produced composites. The incorporation of glass into the composites has significant influence on tensile strength, tensile modulus, flexural strength, and flexural modulus quantified by regression analysis. The ANOVA test has further confirmed about the significance of the improvement of mechanical properties with the produced composites. As MDI is popularly used by particle board manufacturing companies, this report could be a benchmark for hybrid composite manufacturing to the industries.

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Author Contributions

K.M.F.H. conceptualized and investigated the study; T.A. and P.G.H. validated the study; T.A. and P.G.H. involved in funding acquisition; T.A. and P.G.H. involved in project administration; T.A. and P.G.H. dealt with the resources; T.A. and P.G.H. supervised the study; K.M.F.H. dealt with writing and original draft; and P.G.H. and T.A. dealt with writing, reviewing, and editing of the manuscript.

Notes

The authors declare no competing financial interest.

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