



Flame-retardant hybrid composite manufacturing through reinforcing lignocellulosic and carbon fibers reinforced with epoxy resin (F@LC)

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Abstract Novel flame retardant hybrid composites were developed from lignocellulosic and carbon fibers reinforced with epoxy resin using hot pressing technology. The size of the lignocellulosic fibers was within 0.045 to 0.8 mm, and the carbon fibers ranged from 5 to 8 mm. The nominal thickness of the composites was 10 mm, whereas the nominal density was 730 kg/m^3 . The composite dimensions were $400 \times 400 \text{ mm}^2$. The developed panels were tested for internal bonding strength and flexural properties to investigate their mechanical performance. Furthermore, SEM (Scanning electron microscopy) test was conducted to examine the morphologies of the products before and after fracture and found substantial quantities of both types of fiber in the composite system. The EDX (Energy disruptive X-ray) analysis

also displayed the chemical elements present in the developed products. Moreover, an FTIR (Fourier transform infrared spectroscopy) study showed strong chemical interactions among the lignocellulosic and carbon fibers with the thermosetting epoxy polymers. The flame retardancy tests of the composite materials also showed significant thermal stability, especially after loading carbon fibers in the composite system. Moreover, dimensional stability showed an improving trend with the increase of carbon fibers as the reinforcement. Overall, the composite materials developed using lignocellulosic and carbon fibers with epoxy resin are showing a novel route to develop composites with high mechanical performance and considerable flame retardancy.

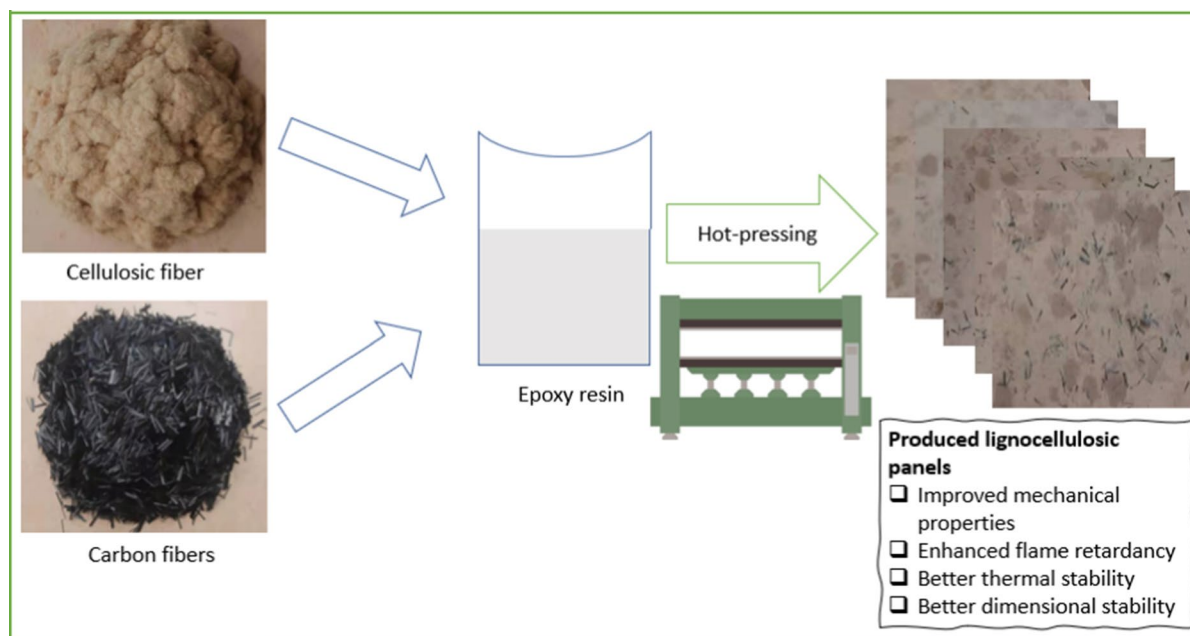
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Graphical abstract



Keyword Lignocellulosic fiber · Carbon filler · Epoxy resin · Hybrid composites · Flame retardancy · Mechanical properties

Introduction

In the field of composites, both synthetic and natural fibers have been extensively studied. However, there is also another type of sustainable cellulosic materials, like fibers from wood or plants, that have not yet been investigated much compared to other types of natural fibers, namely, flax, sisal, hemp, ramie, and so on (Aisyah et al. 2019; Kamau-Devers et al. 2020; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k; Hasan et al. 2022a, b). Additionally, the hybridization of wood-based fibers with synthetic materials like carbon or glass could help to enhance not only their mechanical properties but also their flame retardancy, which are inherent problems of typical wood-based composite products (Khalid et al. 2021). Moreover, wood-based cellulosic fibers provide lower density, lower cost, and eco-friendly features. Recently, much research effort has been focused on investigating the development of hybrid composites through reinforcing natural/synthetic fibers with various thermoplastic/thermosetting polymers (Akpan et al. 2019; Zhang

et al. 2019; Kibleur et al. 2022). Earlier, the composite community was much dependent on synthetic fibers like carbon/glass for producing fiber reinforced plastic products (Khalid et al. 2022). However, over time, natural fibers also gained important attention for producing composite panels due to their enhanced sustainability. In many cases, the hybridization of synthetic fibers with natural fiber could facilitate the increase in the mechanical properties and thermal stability. Therefore, hybrid composites are showing new potential in the fields of aerospace, transportation, furniture, marine, military/defense, structural engineering, and so on (Thyavihalli Girijappa et al. 2019; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k; Mahmud et al. 2021; Tibor et al. 2021; Tripathi et al. 2021).

Lignocellulosic materials are cheaper compared to inorganic fibers. Further advantages include biodegradability, renewability, eco-friendliness, and abundant availability in nature (Sanjay et al. 2019; Rangappa et al. 2022). When lignocellulosic fibers are reinforced with different thermosetting polymers, they

provide strong interfacial adhesion between the fibers and polymers, if the fibers are free from impurities in their polymeric structure (Mahmud et al. 2021). It is already solid proof that, by using plant fibers as the reinforcement material (Ramesh et al. 2020; Vinod et al. 2020; Rangappa 2022), an environment-friendly composite product may be manufactured. However, natural fibers contain cellulosic chemical components like the hydroxyl group, which absorbs water from the atmosphere; hence, 100% biocomposites are not always considered as long-lasting materials like synthetic fiber-reinforced composites. To overcome such kind of drawbacks, wood fibers can be reinforced with carbon fibers (synthetic) and epoxy resin in smaller fractions. Nowadays, beside the usual natural fibers, different plants are also used as the prominent cellulosic materials for producing polymeric composites (Arif et al. 2022a, b). Therefore, plant fibers derived from seven plants were selected to produce composite panels from mixed wood fibers. The mixtures of woods included *Pinus spp.*, *Fagus sylvatica*, *Quercus cerris*, *Quercus robur*, *Carpinus betulus*, *Populus spp.*, and some other types of hardwoods available in the Central European countries such as Hungary. In our previous study, we explored the composites' fabrication potential with OPC (ordinary Portland cement) and mixed lignocellulosic materials providing good thermomechanical performance and dimensional stability (Hasan et al. 2020a, b; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k). However, the possibility of reinforcing the mixed fiber with thermosetting polymers like epoxy resin has not yet been investigated. Moreover, the loading of carbon fibers in the composite system mixed with cellulosic fibers has not been reported yet. This concept has considerable potential to improve mechanical properties and flame retardancy. Chemically, carbon fibers are comprised of carbon atoms that are bound together through the formation of a long polymeric chain. Carbon fibers possess higher stiffness and tensile strength, have higher resistance against chemicals, are lighter in weight, can tolerate higher temperatures, and have lower expansion against thermal treatments (Bhatt et al. 2017; Madhu et al. 2020; Puttegowda et al. 2020, 2021; TG et al. 2021). Previously, wood fibers were also used by researchers in conjunction with thermoplastic polymers, i.e., wood fiber/HDPE (high density polyethylene) reinforced composites, using injection molding (Guo et al. 2019). However,

according to our knowledge, no research has been conducted yet to produce flame retardant composites from mixed wood fibers with or without carbon fibers along with epoxy resin.

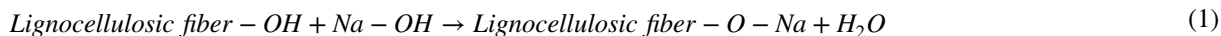
There are different thermosetting and thermoplastic resins available in the market. Thermosetting polymers are comparatively advantageous for panel and fiberboards production. The most common thermosetting polymers are epoxy resin, MUF (melamine urea formaldehyde), MDI (methylene diphenyl diisocyanate), and so on. Compared to other adhesives, epoxy resin provides some magnificent advantages like filling the gaps and resisting water absorbency and chemicals, which facilitates higher durability toward the composite products (Hasan et al. 2020a, b; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k). Epoxy resins also provide higher mechanical properties, lower shrinkage during curing, and are easy to process and have better adhesion capabilities with the fibers (Arif et al. 2022a, b). Therefore, epoxy resin was selected for this study to reinforce lignocellulosic and carbon fibers for producing sustainable composite products. This work will show a new milestone toward sustainable and flame-retardant hybrid structural material production for the composites community's advanced applications.

Materials and methods

The lignocellulosic materials were collected from Kronospan MOFA Hungary Ltd., Hungary. Lignocellulosic materials are comprised of seven wood plants found in Hungary, including *Pinus spp.* (20%), *Fagus sylvatica* (30%), *quercus cerris* (2%), *Quercus robur* (5%), *Carpinus betulus* (15%), *Populus spp.* (20%), and other types of hardwoods in various proportions. Beech was discovered to have the highest proportions of fiber, with poplar and pine coming in second place. The lignocellulosic fibers were pretreated using NaOH (VWR International KFT., Debrecen, Hungary) before producing the composite panels to remove unexpected impurities as per Eq. 1. The carbon fibers were collected from Zoltek Corporation, Nyergesújfalu, Hungary. The epoxy resin used to reinforce the lignocellulosic and carbon fibers were procured from Novia Kft., Hungary (MGS LR 235,

catalyst 35%, colored paste 3%, transparent resin, and viscosity 2500 to 3000 cps). The crosslinker (MGS LH 235) was also purchased from the same company. The mixing ratio of epoxy and curing reagent was 100:35, and they were mixed uniformly before the fabrication of composites.

0.045 mm, respectively. Carbon fibers were within 5 to 8 mm in length (measured randomly for 10 fibers). The moisture contents of both lignocellulosic and carbon fibers were checked before the recipe formulation. The moisture contents of carbon and lignocellulosic fibers were 0.9% and 7.3%, respectively. The



Fabrication of flame retardant hybrid composites

Initially, both types of fibers were sieved to establish their size distributions. The lignocellulosic fibers ranged in size from 0.045 to 0.8 mm. 8.6, 36, 4.4, 8.6, 11.1%, 34.6%, and 29% of the fibers belonged to size classes of 0.8, 0.63, 0.5, 0.4, 0.315, 0.1, and

nominal density of the boards was 730 kg/m³, with a thickness of 10 mm. The recipe used for producing the panels in terms of the proportion of the fibers and resin materials was according to Table 1. However, the actual quantity of the composite materials was calculated according to the recipe, their properties, and their designed dimensional characteristics. In the case of panel 1, 100% lignocellulosic fibers were used beyond the epoxy resin, 95% lignocellulosic fibers and 5% carbon fibers were used for panel 2, 90% lignocellulosic fibers and 10% carbon fibers were used for panel 3, 85% lignocellulosic fibers and 15% carbon fibers were used for panel 4, and 80% lignocellulosic fibers and 20% carbon fibers were used for panel 5 (Fig. 1). Panels 1 to 5 were denoted as LE1 to LE5 for better expression of the products. Prior to producing the composites, both types of fibers were mixed with epoxy resins uniformly by a rotating drum (rotation speed 30 RPM). Firstly, the fibers were put in

Table 1 Recipe for lignocellulosic fiber/carbon fiber reinforced epoxy composites

Composite Materials	Lignocellulosic fiber	Carbon fiber	Epoxy resin
LE1	100	0	7
LE2	95	5	7
LE3	90	10	7
LE4	85	15	7
LE5	80	20	7

*LE- Lignocellulosic fiber/epoxy

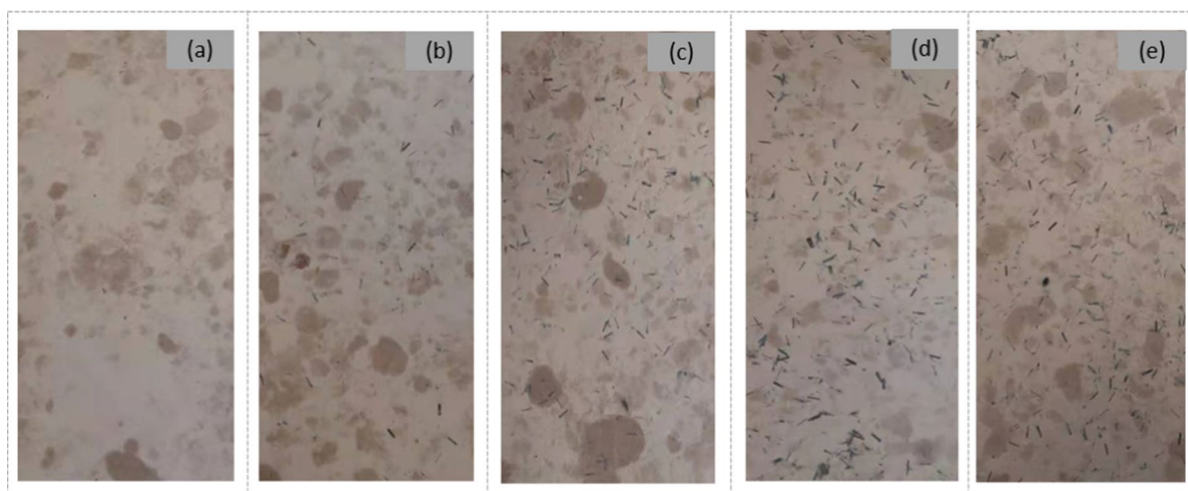


Fig. 1 Photographs of lignocellulosic fiber/carbon fiber reinforced epoxy composites: **a** LE1, **b** LE2, **c** LE3, **d** LE4, and **f** LE5. The visuals of the produced composites, where the domi-

nance of carbon fiber is also noticed with the increase in carbon fiber loading in the composite systems as per the recipe

the drum and rotation was started. Second, epoxy resin was sprayed into the rotating drum with a gun. After complete mixing, all mixed materials were removed from the machine and a mat was produced in a wooden frame of 400 mm by 400 mm dimensions. Before the final pressing, the produced mat was pre-pressed by a wooden lid to ensure uniform shape of the materials. Finally, the steel plate containing the mat was placed in a hot pressing machine (G. Siempelkamp GmbH Co., Kg., Germany) and pressed at 135 °C through applying different pressures (7.1, 4.7, and 3.2 MPa). The total duration of pressing was 150 S (15 s for each mm of thickness). Finally, the machine was cooled down and the pressed composites were removed from it. A similar protocol was followed for all the products.

Characterization of composite products

Moisture contents of the control lignocellulosic and carbon fibers were measured using a moisture analyzer (Kern ULB 50-3N, KERN AND SOHN GmbH CO., Germany) through adopting of the standard EN322:1993. Sugar and tannin contents of lignocellulosic materials were investigated to understand the presence of impurities in the raw materials using laboratory-based analytical methods, which are discussed in detail in my PhD thesis and that of our colleagues. Flexural and internal bonding strengths were measured using an Instron-4208 (United States) universal testing machine. The standards used for flexural and internal bonding strength measurements were EN 310 and EN 319, respectively, and the crosshead movements were 5.0 and 0.8 mm/min, respectively. A circular saw (DCS570N XJ, Pennsylvania, United States) was utilized to prepare the samples according to standard specifications.

SEM micrographs were taken through an electron microscope (S 3400 N, High Technologies Co., Ltd., Hitachi, Japan) at 10.0 kV voltage. Moreover, the chemical composition of both control and composited products was investigated using an SEM-mediated EDX system. FTIR studies were carried out within the range of 4000 to 400 cm^{-1} by an FT/IR-6300 spectroscope from Jasco, Japan. Flame retardancy of the produced composites was tested by “The Single Source Test” as per the EN ISO 11925–2:2011 standard using NETZSCH TAURUS Instruments GmbH (Weimar, Germany). In order to perform this test, the samples were cut to the dimensions of 250 mm by 90 mm to test ignitability. The samples were subjected directly to the impingement of a smaller flame. The samples were placed at a nearly 45° contact angle, and the flame height was 20 mm. The samples were exposed to the flame for 15 s, and the test was terminated 20 s after flame removal. The detailed testing protocols of different tests were provided in reference (“Ignitability test EN ISO 11925–2”; Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k). Thermal stability of the control and composited samples was investigated using a Themys thermal analyzer (Setaram Instrumentation, France) at 10 °C/min, whereas the heating was continued within a range of 23 to 850 °C. The heating flow of nitrogen gas was 50 mL/min whereas the sample quantity was 16 to 24 mg.

Results and discussions

Sugar and tannin contents create problems for the interactions between the lignocellulosic fibers and polymers. Hence, prior to starting the composite fabrication process, the sugar and tannin content test

Table 2 Physical and mechanical properties of lignocellulosic fiber/carbon fiber-reinforced epoxy composites

Panels	D (kg/m^3)	T (mm)	IBS (MPa)	MOR (MPa)	MOE (GPa)
LE1	641.3 (42.59)	9.8 (0.04)	0.073 (0.005)	6.99 (1.2)	3.37 (0.52)
LE2	644.9 (20.1)	10.2 (0.15)	0.1 (0.016)	7.63 (0.43)	3.95 (0.22)
LE3	644.9 (20)	10.0 (0.08)	0.11 (0.016)	7.377 (0.74)	4.19 (0.24)
LE4	656.7 (20.9)	9.7 (0.11)	0.135 (0.015)	7.4 (0.98)	4.2 (0.34)
LE5	739.8 (20.1)	9.79 (0.01)	0.147 (0.005)	7.92 (0.16)	4.45 (0.26)

*D-Density, T- Thickness, IBS- Internal bonding strength, MOR- Flexural strength, MOE- Modulus of elasticity, EBS- Elongation at break (%), R^2 - Coefficient of variation

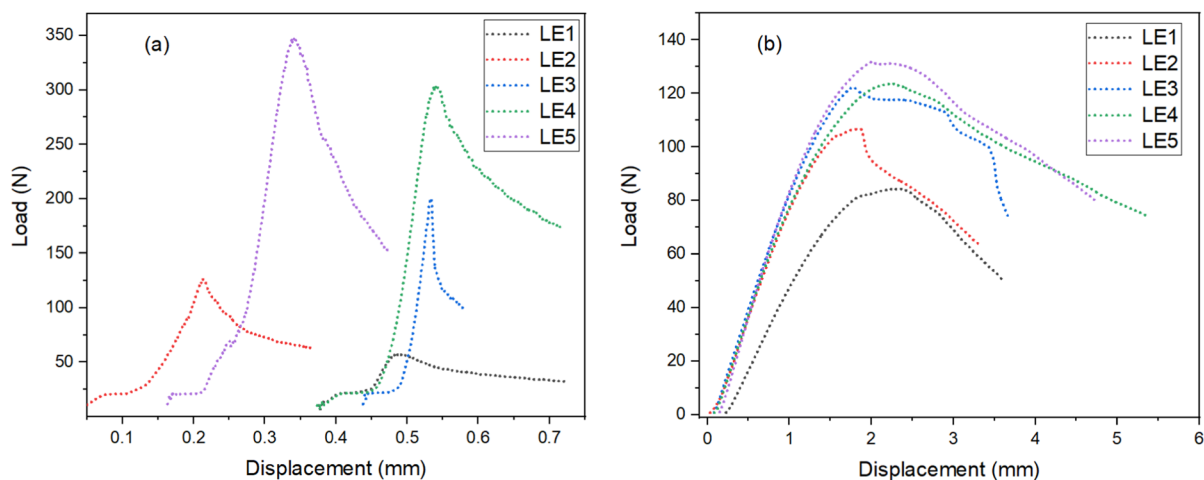


Fig. 2 Load–displacement curves for lignocellulosic/carbon fiber reinforced epoxy composites: **a** Internal bonding strength and **b** Flexural properties

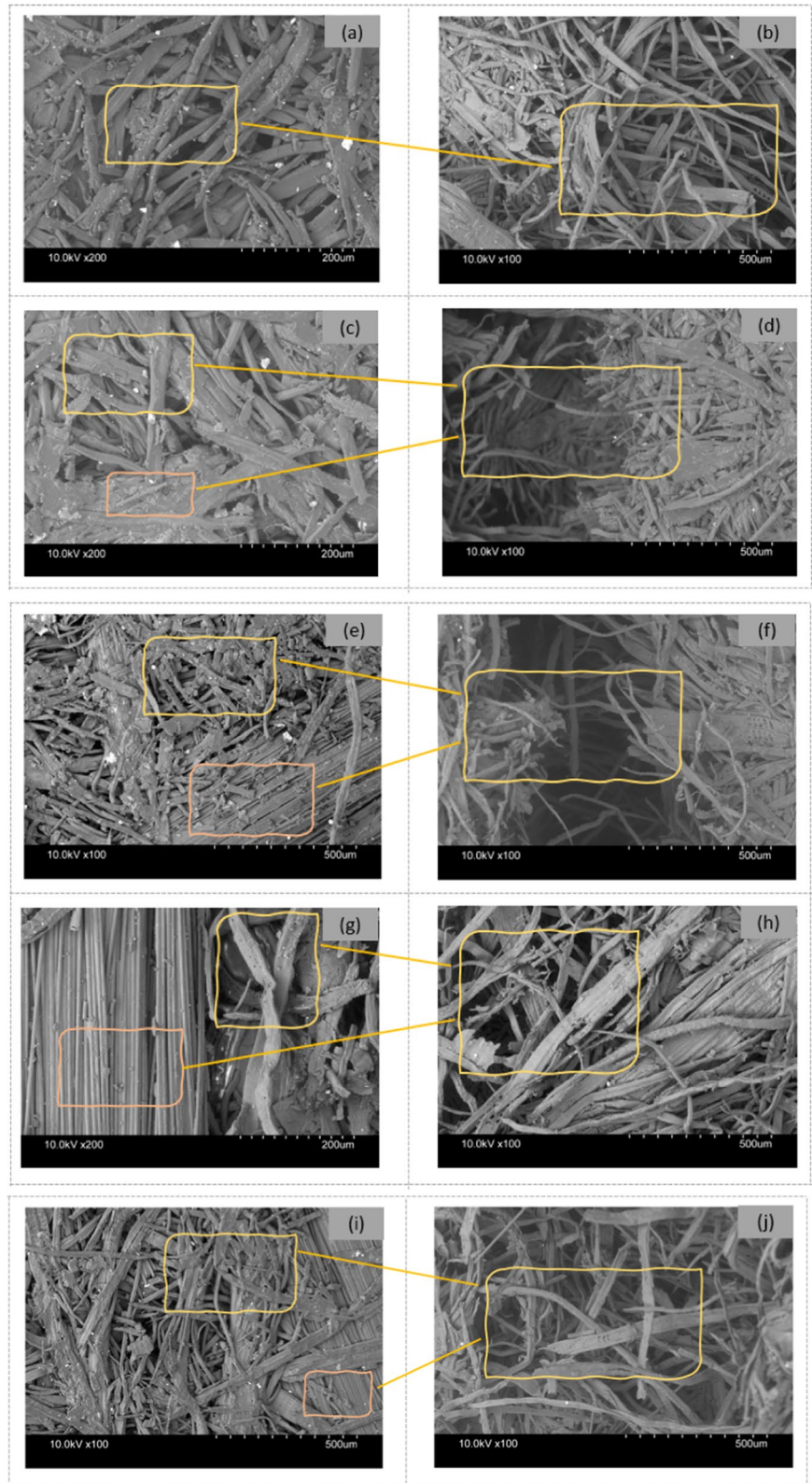
was carried out. The sugar content of the lignocellulosic fiber was 0.3% and the tannin content 0.25%, which are within the tolerable limits (0.4% tannin content and 0.5% sugar content (Tibor L. Alpar- Éva Selmeczi 2012; K. Hasan et al. 2020a, b)). The densities of the produced panels were 641.3 (42.59), 644.9 (20.1), 644.9 (20), 656.7 (20.9), and 739.8 (20.1) kg/m³ for panels LE1 through LE5, respectively, whereas the nominal density was 730 kg/m³ (taken into consideration during recipe formulation). The respective thicknesses of the boards were 9.8 (0.04), 10.2 (0.15), 10.0 (0.08), 9.7 (0.11), and 9.79 (0.01) mm, whereas the nominal thickness was 10 mm (Table 2). The variations in thicknesses and densities were possibly caused by manual operations ranging from material measurements according to recipe calculation to composite fabrication and sample preparation for testing the products. However, the variation in density has an increasing pattern with the increase in carbon fibers in the composite system. It may be that lignocellulosic fibers are bulkier compared to carbon fibers; hence, the process loss was higher.

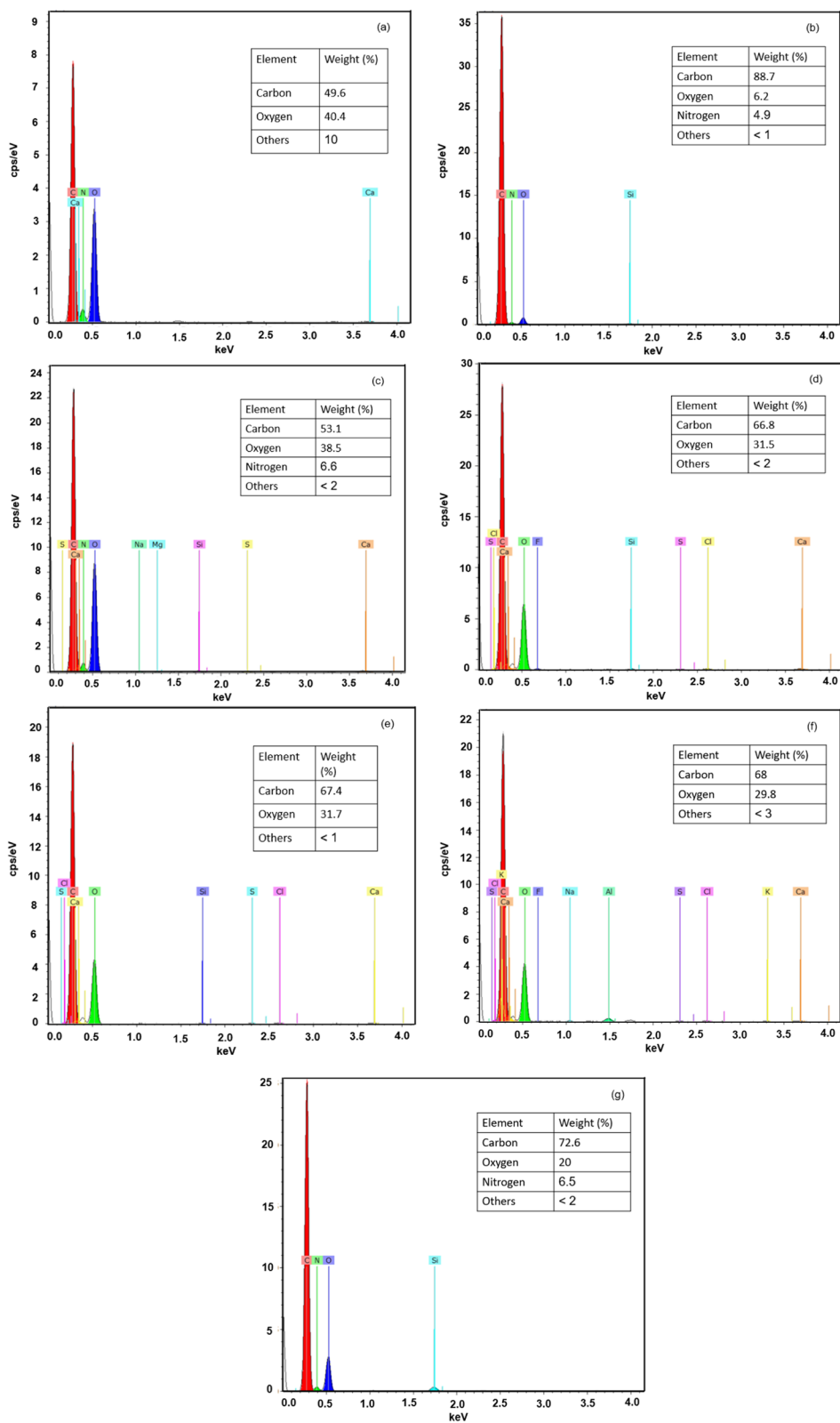
Mechanical properties of the composite products were investigated further in terms of flexural properties (MOR and MOE) and internal bonding strength. The load versus displacement curves for the internal bonding strength and flexural properties are displayed in Fig. 2 (a and b). The curves show linear behavior at the initial stage but become non-linear when approaching the maximum load. The loading

conditions, on the other hand, cause a crack in the composite test specimens, which continues with the extended level of displacements. As expected, 100% lignocellulosic fiber-reinforced composites needed the lowest loads to initiate crack propagation, whereas the 20% carbon/80% lignocellulosic fiber reinforced epoxy resin required the highest load. The sequence of load needed for crack propagation is LE1 < LE2 < LE3 < LE4 < LE5 for the composite samples, especially in the case of internal bonding strength. The maximum load needed to break sample 1 is 57.2 N for LE1, 126.3 N for LE2, 199.9 N for LE3, 304.7 N for LE4, and 348.1 N for LE5. Similar trends were also noticed for the flexural properties. The highest loads needed to bend the LE1 sample are 84.3 N, LE2 106.4 N, LE3 121.9 N, LE4 123.6 N, and LE5 is 131.6 N. Similar load/displacement results were discovered for other lignocellulosic panel products as well (Hasan et al. 2021a, 2021b).

Like load–displacement curves, similar trends are also seen for the mechanical properties of the products. 100% lignocellulosic fiber-reinforced composites showed the lowest internal bonding strengths of 0.073 (0.005) MPa as opposed to that of 20% carbon fiber-loaded composites with 80% lignocellulosic fiber in the composite system (0.147 (0.005) MPa). Other panels, including LE2, LE3, and LE4, showed intermediate strengths of 0.1 (0.016), 0.11 (0.016), and 0.135 (0.015) MPa, respectively. Additionally, the flexural strengths and moduli also followed a

Fig. 3 SEM analysis of lignocellulosic/carbon fiber reinforced epoxy composites: **a** LE1, **c** LE2, **e** LE3, **g** LE4, **i** LE5 before fracture, and **b** LE1, **d** LE2, **f** LE3, **h** LE4, **j** LE5 after fracture





◀**Fig. 4** EDX investigation of lignocellulosic fiber/carbon fiber reinforced epoxy composites: **a** fiber, **b** carbon, **c** LE1, **d** LE2, **e** LE3, **f** LE4, and **g** LE5

similarly increasing trend with the increase in carbon fiber content in the composite system. MOR values of the composites were found to be 6.99 (0.2), 7.63 (0.43), 7.377 (0.74), 7.4 (0.98), and 7.92 (0.16) MPa, whereas the MOE values were 3.37 (0.52), 3.95 (0.22), 4.19 (0.24), 4.2 (0.34), and 4.45 (0.26) MPa, respectively, for LE1, LE2, LE3, LE4, and LE5 panels. In the case of flexural properties, strength and modulus values also increased with the increase in carbon fibers in the composite system, where 100% lignocellulosic fiber reinforced composites showed the lowest values and 20% carbon fiber loaded composites with 80% lignocellulosic fiber showed the highest performances.

Figure 3. shows the morphological images of the developed composites. The presence of fibers is observed as being uniformly distributed throughout the composite system. Traces of the matrix are seen in and around the fibers. A strong bonding among the fibers and epoxy resin is noticed, whereas the 100% lignocellulosic fiber-reinforced composites seem to have weaker adhesion. Therefore, in the fractured samples, it is easy to separate the fibers from the matrix system without any major cracking. However, it was difficult to separate the fibers from the matrix as there was a hole created in the composites that contain carbon fibers when the samples were pulled out during internal bonding strength tests. Therefore, the composites reinforced with carbon fibers show better internal bonding strength and flexural properties too. Overall, SEM micrographs provided strong evidence regarding the fiber material's adhesion to the epoxy matrix in the composite system. Similar morphological analyses were also reported for flax/carbon fiber-loaded epoxy hybrid composites (Bagheri et al. 2013; Dhakal et al. 2020).

Additionally, to investigate the chemical elements present in the control materials (both carbon and lignocellulosic fibers) and their associated composites, an EDX test was carried out. As expected, EDX analysis of control lignocellulosic fibers displayed a dominant presence of carbon and oxygen of 49.6% and 40.4%, respectively, whereas carbon fibers showed 88.7% carbon, 6.2% oxygen, and 4.9% nitrogen. As carbon is the main chemical compound for both types

of control materials, their reinforced composites also displayed the dominance of carbon materials compared to others like oxygen and nitrogen. Moreover, the weight% of carbon also starts to increase with the increased loading of carbon fiber in the composite system. Therefore, the weight% of carbon in the LE1 composite was 53.1%, LE2 was 66%, LE3 was 67.4%, LE4 was 68%, and LE5 was 72.6% (Fig. 4).

The reinforcement effects of lignocellulosic and carbon fibers with epoxy resin were investigated further using FTIR analysis for both control and composited products. The broad absorption band between 3600 and 3200 cm^{-1} is associated with the stretching vibrations of -OH groups of carbohydrates and lignin (Hasan et al. 2022a, b). However, there are no such peaks noticed for control carbon fibers within this range (Fig. 5). The peaks within 2880 to 2930 cm^{-1} are demonstrating the presence of C-H present in the cellulose and hemicellulose polymers. Control lignocellulosic fiber showed the peak at 3333 cm^{-1} , whereas composite materials LE1 through LE5 displayed the peak at 3334, 3336, 3334, 3333, and 3333 cm^{-1} , respectively. In contrast to lignocellulosic materials, the control carbon fiber did not show any peaks like this at this wavelength. After reinforcement with epoxy resin and carbon fibers, shift/stable bands may be observed. However, these bands are associated with the absorption of water from the surrounding atmosphere, therefore they may be absent from carbon fibers (neat). The patterns of lignocellulosic material reinforced composites are also in line with previous research results (Hasan et al. 2021a, 2021b). Neat epoxy resin also gives a peak at 3364 cm^{-1} (Cecen et al. 2008), which can be shifted somewhat after the reinforcement with fibers. In the composite system, an interaction happens between the -OH groups of the resin and lignocellulosic and the carbon fiber. The peaks of the control and composited products are summarized in Table 3. Additionally, -CH stretching vibrations (aliphatic) are noticed within 2800 to 3000 cm^{-1} bands (Cecen et al. 2008; Barbosa et al. 2017). However, there is very little/insignificant variation in the composite products. Interestingly, the formation of bonds between carbon fiber and epoxy resin could be detected by the peaks around wavenumbers 2361 to 2362 cm^{-1} (Cecen et al. 2008). Emerging peaks at 895 to 896 cm^{-1} are responsible for the C-OH groups due to the β -glycosidic linkages present in the monosaccharides (Cai et al. 2016).

Fig. 5 FTIR analysis of lignocellulosic/carbon fiber-reinforced epoxy composites

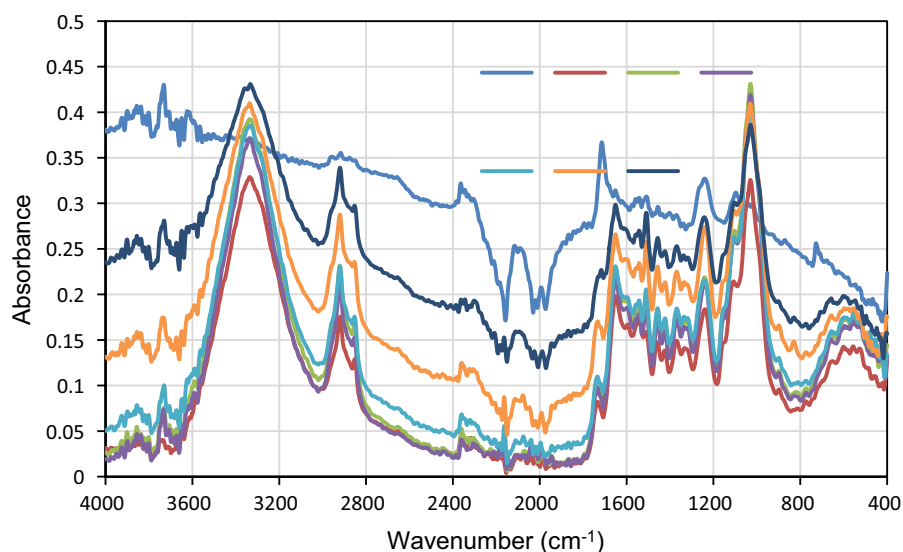


Table 3 Summarized peak information on FTIR analysis of lignocellulosic/carbon fiber reinforced epoxy composites

	Carbon fiber	Lignocellulosic fiber	LE1	LE2	LE3	LE4	LE5
-OH group		3333	3334	3336	3334	3333	3334
-CH stretching vibrations	2912	2919	2921 and 2851	2918 and 2851	2920 and 2851	2919 and 2851	2918 and 2851
Carbon/epoxy bonding (with/without)	2362		2359	2360	2361	2360	2361
C–O–C stretching		1028 and 1241	1028 and 1237	1029 and 1237	1028 and 1241	1031 and 1238	1029 and 1242
C–OH		896	895	896	895	895	895
C=C	1733	1734	1733	1733	1733	1733	1734

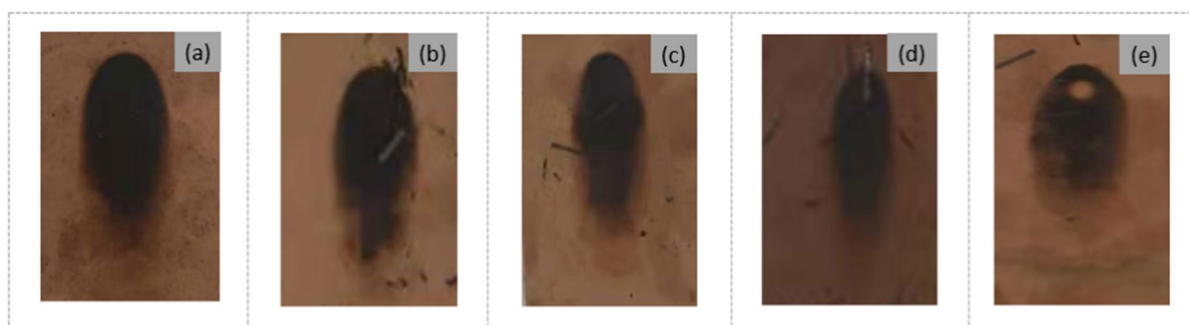


Fig. 6 Flame retardancy test of lignocellulosic fiber/carbon fiber reinforced epoxy composites: **a** LE1, **b** LE2, **c** LE3, **d** LE4, **e** LE5. The fire resistance of the composites increased with the increase in carbon fiber loading in the composite sys-

tem. As a result, composite panel 1 (LE) is found to be more susceptible to burning than LE5 (the sample with the highest carbon-fiber content)

The peaks in the range of 1028 to 1247 cm^{-1} are denoted for the C=C functional group present both in the control cellulosic fiber and composited products (Rizal et al. 2018). The stretching vibrations related to carboxylic acid and vinyl esters could be demonstrated by the bands at 1733 to 1734 cm^{-1} both for control cellulosic fiber and composite products (Rizal et al. 2018). Overall, the peaks within the 3200 to 3600 cm^{-1} and 2361 to 2362 cm^{-1} ranges confirm the successful reinforcement of lignocellulosic and carbon fibers with epoxy resin.

Among all the composites, carbon fiber-loaded products displayed better flame retardancy. 20% carbon fiber-incorporated products showed the best stability and 100% lignocellulosic materials displayed the lowest one. The control samples started to burn immediately when exposed to flame. However, the incorporation of carbon fiber in the composite system

facilitates the gradual decline in this burning tendency (Fig. 6). When exposed to flame, wood is prone to burning, but carbon fibers exhibit superior fire resistance (Grange et al. 2019). Therefore, as indoor products like furniture items are at a big risk of fire-related burning, the use of carbon fiber along with lignocellulosic materials would not only enhance the increased sustainability but also improve the mechanical properties and fire resistance as well (Bartlett et al. 2019). The overall trends of flame retardancy sequence in the composites are LE5 > LE4 > LE3 > LE2 > LE1.

Moisture susceptibility is one of the critical challenges of natural fiber reinforced composites. The reason behind this is the presence of hydrogen bonds in fiber cell wall molecules, as the plant-based fibers consist of dominant cellulosic and hemicellulosic polymers. The cellulosic materials are also composed of -OH (hydroxyl) and C (carbon). However, although

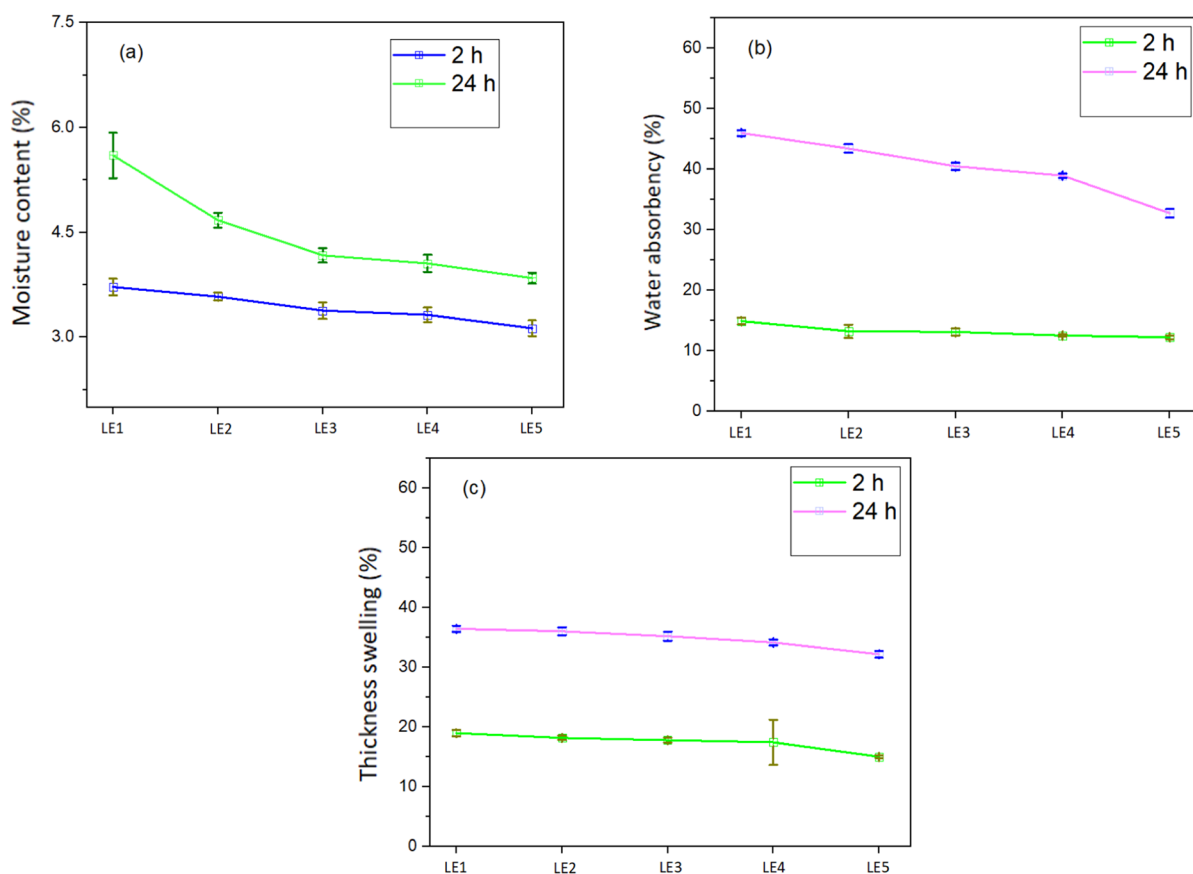


Fig. 7 Physical properties of lignocellulosic fiber/carbon fiber reinforced epoxy composites: **a** Moisture content, **b** Water absorption, and **c** Thickness swelling

the cellulose polymer is enriched with crystalline regions, which could retard the moisture penetration, they also contain amorphous regions, which facilitate the water molecules to be defused and break the hydrogen bonds (Moudood et al. 2019). Therefore, an intermolecular distance is created in the cellulose chains, and consequently, the fibers are swollen. On the other hand, carbon fibers do not absorb moisture from the surrounding environment, or absorb water when immersed (Zhai et al. 2016). As a result, when carbon fiber is mixed with lignocellulosic materials in a composite system, the test specimens' affinity to absorb moisture decreases. Figure 7 shows that 100% lignocellulosic materials reinforced composites absorb the highest amount of moisture 3.72 (0.12)%, whereas the 20% carbon/80% lignocellulosic material reinforced composites absorb the least, 3.13 (0.11)%, after 2 h of oven drying. A modest amount

of moisture was absorbed by the 10% carbon/90% lignocellulosic composite at 3.38 (0.12)% after the same period of immersion. Similar trends in moisture absorption were also noticed even after 24 h of immersion, where an increase was observed compared to 2 h for overall moisture absorption. Interestingly, the composite samples also followed the same pattern for water absorbency and thickness swelling. After 2 h of immersion, the 100% lignocellulosic materials reinforced composites absorbed the most water, with a value of 14.89 (0.56)%, and the 20% carbon/80% lignocellulosic material absorbed the least, with a value of 12.22 (0.29)%. Moreover, the highest thickness swelling was found in the case of 100% lignocellulosic material reinforced composites, providing the value of 18.97 (0.55)% and the lowest one is 14.99 (0.23)% for the 20% carbon/80% lignocellulosic composite. This phenomenon also agrees

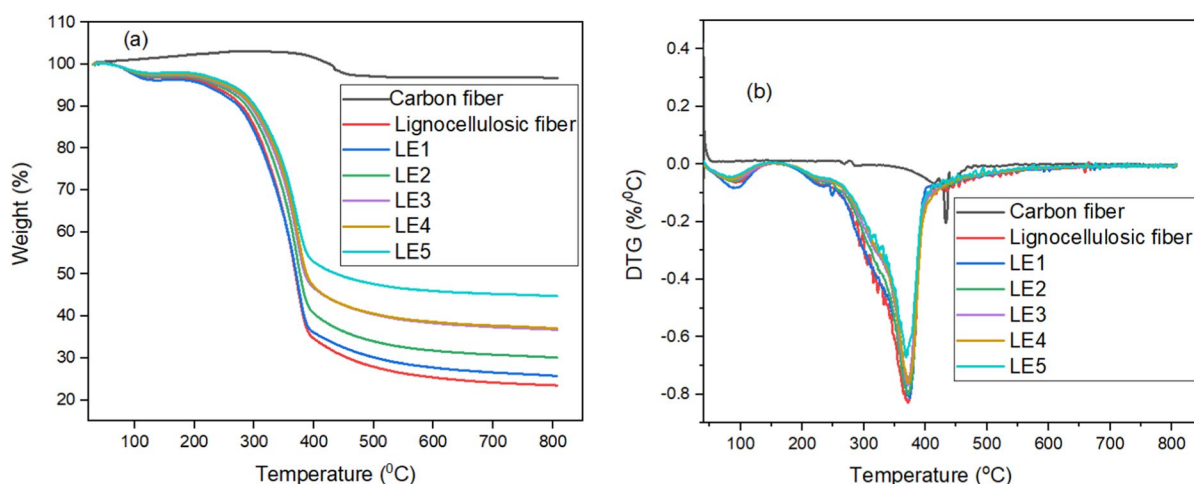


Fig. 8 Thermal properties analysis of lignocellulosic/carbon fiber reinforced epoxy composites: **a** TGA and **b** DTG

Table 4 Temperature characteristics of at elevated weight loss of the materials

Samples	T_{on} (°C)	T_{Max} (°C)	Weight loss (%)	Residues at 650 °C (Weight%)
Control carbon fiber	428	583	3.2	96.8
Control lignocellulosic fiber	190	408	72.4	24.51
LE1	191	409	64.71	29.96
LE2	192	409	61.22	30.96
LE3	193	410	57.3	37.77
LE4	195	415	54.55	37.93
LE5	198	426	49.39	45.59

with our previous study for glass/flax fiber-reinforced MDI composites (Hasan et al. 2021a, b, c, d, e, f, g, h, i, j, k). It seems that the physical properties become more stable with the incorporation of carbon fiber in the composite system.

The thermal properties of the control carbon and lignocellulosic fibers, along with the composited products, also investigated further in terms of TGA and DTG analysis (Fig. 8). The characteristics (onset temperature— T_{on} , temperature at decomposition— T_{max} , weight loss (%), and nonvolatile fragment of materials at 650 °C) of the materials are also tabulated in Table 4. The initial weight loss is observed around 100 °C in the case of control lignocellulosic fiber and composited products, which is due to the moisture evaporating from the samples. However, carbon fibers did not display any initial weight loss at this stage, maybe due to the absence of any water molecules in their polymeric chains. The onset degradation for carbon fiber is so high (428 °C), whereas the 100% lignocellulosic fiber is displaying the onset degradation at 190 °C. However, after increasing the loading of carbon fibers in the system, the temperature started to show an increasing trend to degrade the materials (Table 4). 5% carbon fiber-loaded sample shows the onset of degradation at 190 °C, whereas 20% carbon fiber loaded composites T_{on} at 198 °C. A similar pattern can be seen in the case of T_{Max} . Interestingly, control carbon fiber showed very little weight loss at the maximum level of degradation (3.2%), whereas control lignocellulosic fiber degradation was 72.4%. The composite loaded with 20% carbon fiber shown the maximum stability against the temperature (only 49.39% weight loss). The composite loaded with 20% carbon fiber demonstrated the greatest temperature stability (only 49.39% weight loss). However, all the samples loaded with carbon fiber showed better stability than the control lignocellulosic fiber-loaded composites. Moreover, the residues after 650 °C also show that carbon fiber is still showing 96.8% char yield, whereas 100% lignocellulosic fibers show 24.51%, which starts to escalate with the higher loading of carbon fibers in the composite system. The results also goes in line with different carbon- or vegetable-based natural fiber-reinforced polymeric composites (Aisyah et al. 2019; Dinesh et al. 2020).

Additionally, the degradations within 250 to 450 °C demonstrate the decomposition of cellulose,

hemicellulose, and lignin present in the lignocellulosic fibers (Kumar et al. 2014). The higher char yield is also responsible for the amount of lignin present in the materials (Kumar et al. 2014). Moreover, the degradations within 200 to 300 °C also correspond to the epoxy polymers present in the composite materials (Kumar et al. 2014).

Conclusions

Novel flame-retardant composites produced from lignocellulosic materials and carbon fibers reinforced with epoxy resin were reported in this study. Pretreatment of the lignocellulosic fibers was performed before the fabrication. The developed composites showed excellent flame retardancy, which improved with the increase in carbon fiber content in the composite system. The FTIR test results showed successful bonding among the natural and synthetic fibers in the presence of epoxy resin. The morphological photographs further demonstrated successful and efficient reinforcement effects in the composite system. However, the most prominent results were found in the case of internal bonding strength and bending properties, which confirms a stronger material development. Moreover, fire retardant tests also confirmed excellent resistance to burning due to the incorporation of carbon fibers. SEM images of fractured surfaces further confirmed the lower degree of separation of carbon fiber-reinforced composites from the matrix system, which resulted in better mechanical performance compared to non-carbon-fiber-loaded samples. Moreover, increased loading of carbon fibers also retards moisture absorption from the surrounding environment by the composites. Thermal properties investigation has also shown that the incorporation of carbon fiber in the composite system also improves the thermal stability of the composites. In summary, the developed products would facilitate the composite material manufacturers and community with an innovative approach to the development of novel hybrid composites.

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Author contributions KMFH generated the idea, conducted the experiment, data curation, and written the initial draft. ZK and MB cooperated for testing. PGH, MB, and TA helped for project administration, data validation, and final draft review.

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Data Availability of data and materials The data is available on request from corresponding authors.

Declarations

Conflict of interest The authors declare that they have no competing interest for the submitted work. This research does not contain any investigation related to human participation or any animals conducted by the authors. In this regard, authors have no statement about ethical approval.

Informed consent This research work did not involve any human participation therefore authors have no statement about informed consent.

Consent for publication The authors have given their consent to publish this research work.

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