

Flame retardant hybrid composites 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 carbon fibers within mm 5 to 8 mm. Thickness of the boards were 10 mm and nominal density 730 kg/m³ whereas the dimensions were 400 mm × 400 mm. The developed panels were undergone for internal bonding strength and flexural properties to investigate the mechanical performances. Furthermore, SEM (Scanning electron microscopy) test was conducted to examine the morphologies of the products before and after fracture and found the significant presence 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, The FTIR (Fourier transform infrared spectroscopy) study shown a strong chemical interactions among the lignocellulosic and carbon fibers with the thermosetting epoxy polymers. The flame retardancy tests of the composite materials also shown significant thermal stability especially after loading carbon fibers in the composite system. Moreover, significant dimensional stability also noticed which was showing an improving trend with the increase of carbon fibers as the reinforcement. Overall, the composite materials developed from lignocellulosic and carbon fibers with epoxy resin is showing a novel route to the composite community with high mechanical performance and significant flame retardancy.

Introduction

In the field of composites, both synthetic and natural fibers have been extensively studied. However, there is also another brunch of sustainable cellulosic materials like fibers from woods/plants are not yet investigated like as different types of natural fibers namely flax, sisal, hemp, ramie, and so on (K. F. Hasan, Horváth, Miklos, & Alpár, 2021; K. F. Hasan, Horváth, & Alpár, 2021a; Kamau-Devers & Miller, 2020). 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. Moreover, wood-based cellulosic fibers provide lower density, lower cost, and eco-friendly features. Recently, much research effort has been focused to investigate the development of hybrid composites through reinforcing natural/synthetic fibers with various thermoplastic/ thermosetting polymers (Akpan, Wetzel, & Friedrich, 2019; K. M. F. Hasan, P. G. Horváth, & T. Alpár, 2021; Kibleur, Aelterman, Boone, Van den Bulcke, & Van Acker, 2022; Zhang et al., 2019). Previously, composite community was much dependent on synthetic fibers like carbon/glass for producing fiber reinforced plastic products. However, with the span of time, natural fibers also gaining significant attentions for producing composite panels due to their enhanced sustainability. In many cases, the hybridization of synthetic fibers with natural fiber could facilitate to increase the mechanical properties and thermal stability. Therefore the hybrid composites are showing novel potentiality in the field of aerospace, transportation, furniture, marine, military/defense, structural engineering, and so on (K. Hasan, P. G. Horváth, & T. Alpár, 2021; K. F. Hasan, Horváth, Zsolt, & Alpár, 2021; Mahmud et al., 2021; Tibor, Péter, & Hasan, 2021; Tripathi & Behera, 2021).

The lignocellulosic materials are cheaper compared to inorganic fibers especially for biodegradability, renewability, eco-friendliness, and abundantly availability in the nature. When the lignocellulosic fibers are reinforced with different thermosetting polymers, they provide strong interfacial adhesions between the fibers

and polymers, if the fibers are free from impurities presence in their polymeric structure. It is already solid proof that, by using plant fibers as the reinforcement material, an environment-friendly composite product could be manufactured. Nowadays, beside the usual natural fibers different plants are also used as the prominent cellulosic material for producing polymeric composites. Therefore, plant fibers derived from seven plants were selected to produce composite panels from mixed wood fibers. The mixtures of woods entailed *Pinus spp.*, *Fagus sylvatica*, *quercus cerris*, *Quercus robur*, *Carpinus betulus*, *Populus spp.*, and some other types of hardwoods available in the central European countries like Hungary. In our previous study, we explored the composites fabrication potentiality with OPC (ordinary Portland cement) and mixed lignocellulosic materials providing significant thermomechanical performances and dimensional stability (K. Hasan et al., 2020; K. F. Hasan, P. G. Horváth, & T. Alpár, 2021). However, the possibility to reinforce the mixed fiber with thermosetting polymers like epoxy resin is not yet investigated. Moreover, the loading of carbon fibers in the composite system with mixed cellulosic fibers is not also reported yet having significant mechanical and flame retardancy. Chemically, carbon fibers are comprised of carbon atoms which are bonded together through forming a long polymeric chain. Carbon fibers possess higher stiffness and tensile strength, highest resistance against chemicals, lighter in weight, tolerance to higher temperature, and lower expansion against thermal treatments (Bhatt & Goe, 2017). Previously, wood fibers were also practiced by researchers for thermoplastic polymers i.e. wood fiber/ HDPE (high density polyethylene) reinforced composites through using injection molding technique (Guo, Finkenstadt, & Nimmagadda, 2019). However, still now, according to our knowledge, no research conducted yet to for producing flame retardant composites from mixed wood fibers loaded with or without carbon fibers along with the epoxy resin.

There are different thermosetting and thermoplastic resins available in the market. However, thermosetting polymers are comparatively advantageous for panel/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 provide some magnificent advantages like filling the gaps and resisting water absorbency and chemicals, which are facilitating to generate higher durability's toward the products (K. F. Hasan, Horváth, Baş, & Alpár, 2021; K. F. Hasan, P. G. Horváth, K. Zsolt, Z. Kóczán, et al., 2021; K. M. F. Hasan, Péter György, & Tibor, 2020). Primarily, epoxy resins also provide higher mechanical properties, lower shrinkage during curing, and after all easy to process and better adhesion capabilities with the fibers. Therefore, epoxy resin was selected for this this current 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 materials production to the composites community for 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 (*Pinus spp.* (20%), *Fagus sylvatica* (30%), *quercus cerris* (2%), *Quercus robur* (5%), *Carpinus betulus* (15%), *Populus spp.* (20%), and other types of hardwoods in different proportions). It is found that the beech contains the highest proportions of fiber content, whilst the poplar and pines are accumulating the second highest fiber content. The lignocellulosic fibers were pretreated using NaOH (VWR international KFT., Debrecen, Hungary) before going to produce the

composite panels to remove the unexpected impurities as per Eq. 1. The carbon fibers were collected from Zoltek corporation, Nyergesújfalú, Hungary. The epoxy resin used to reinforce the lignocellulosic and carbon fibers were procured from Liebe Engel, China (colorless and transparent resin, viscosity 2500 to 3000 cps). The mixing ratio of epoxy and curing reagent was 3:1, whereas they were mixed uniformly before the fabrication into composites.



Fabrication of flame retardant hybrid composites

Initially, both types of fibers were sieved to understand their size distributions. The size distributions of lignocellulosic fibers are within 0.045 to 0.8 mm range. Whereas 8.6% fibers were belonged to 0.8 mm in length, 36% by 0.63 mm, 4.4% fibers 0.5 mm, 8.6% fibers 0.4 mm, 11.1% fibers 0.315 mm, 34.6% fibers belong to 0.1 mm, and 29% fibers 0.045 mm. Moreover, the 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 formulations. Moisture contents of carbon and lignocellulosic fibers were 0.9 and 7.3%, respectively. However, the nominal densities of the boards were 730 kg/ m³ and thickness 10 mm. However, their recipe as mentioned in Table 1 is the proportion of the fibers and resin materials used for producing the panels. However, the actual quantity of the materials was calculated according to the recipe, their properties, and designed dimensional characteristics. However, in 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). The panel 1 is termed as LE1, panel 2 as LE2, panel 3 as LE3, panel 4 as LE4, and panel 5 as LE5 for better expression of the products. Prior to produce the composites, both type of fibers and resins were mixed uniformly by a rotating drum (rotation speed 30 RPM). Firstly, the fibers were put in the drum and rotation was started. Secondly, epoxy resin was sprayed using a gun in the drum whilst it was rotating. 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. Produced mat was then prepressed by a wooden lid to ensure uniform shape of the materials before the final pressing. 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 second for each mm of thickness). Finally, the machine was cooled down and the pressed composites were removed from the machine. Similar protocol was followed for all the products.

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

Composite Materials	Lignocellulosic fiber	Carbon fibers	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			

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 EN322:1993 standard. Sugar and tannin contents of lignocellulosic materials were investigated to understand the presence of impurities in the raw materials using laboratory-based analytical methods. Flexural and internal bonding strengths were measured using Instron-4208 (United States) instrument. Standard adopted for both flexural and internal bonding strengths were EN 310 and EN 319, respectively whereas the crosshead movements were 5.0 and 0.8 mm/min, respectively. A circular saw cutting machine (DCS570N XJ, Pennsylvania, United States) was utilized to prepare the samples according to different standards. SEM micrographs were taken through an electron microscope (S 3400 N, High Technologies Co., Ltd., Hitachi, Japan) at 10.0 kV voltage. Moreover, chemical composition of both control and composited products were investigated applying SEM mediated EDX system. Furthermore, FTIR studies were carried out within 4000 to 400 cm^{-1} by FT/IR-6300, Jasco, Japan. Flame retardancy of the produced composites were tested by "The Single Source Test" as per EN ISO 11925-2:2011 standard using NETZSCH TAURUS Instruments GmbH (Weimar, Germany). In order to perform this test, the samples were prepared 250 mm by 90 mm to test ignitability. The samples were subjected directly to the impingement of a smaller flame. The samples were brought nearly 45° contact angle, whereas the flame height was 20 mm. The samples were exposed to the flame for 15 seconds, whereas the test was terminated right after 20 seconds of flame removal. The detailed testing protocols of different tests were provided in reference (K. Hasan, P. G. Horváth, Z. Kóczán, D. H. A. Le, et al., 2021; "Ignitability test EN ISO 11925-2").

Results And Discussions

Sugar and tannin contents create problems for better interactions between the lignocellulosic fibers and polymers, hence prior going to composites fabrication the sugar and tannin content test was carried out. Sugar content of the lignocellulosic fiber was 0.3% and tannin content 25%, which are within the range (0.4% tannin content and 0.5% for sugar content (K. Hasan et al., 2020; Tibor L. Alpar- Éva Selmeczi, 2012)). Density of the produced panels were found to have the values by 641.3 (42.59), 644.9 (20.1), 644.9 (20), 656.7 (20.9), and 739.8 (20.1) kg/m^3 , respectively for 100% lignocellulosic fiber, 5% carbon/ 95% lignocellulosic materials,

10% carbon/ 90% lignocellulosic materials, 15% carbon/ 85% lignocellulosic materials, 20% carbon/ 80% lignocellulosic materials whereas the nominal density was 730 kg/m³ (taken into considerations during recipe formulation). Thickness of the boards were also found for the panels by 9.8 (0.04), 0.1 (0.016), 0.11 (0.016), 0.135 (0.015), and 0.147 (0.005) mm, respectively for 100% lignocellulosic fiber, 5% carbon/ 95% lignocellulosic materials, 10% carbon/ 90% lignocellulosic materials, 15% carbon/ 85% lignocellulosic materials, 20% carbon/ 80% lignocellulosic materials reinforced epoxy composites, whereas the nominal thickness was 10 mm (Table 2). The variation in the thicknesses and densities happened maybe due to the manual operations starting from measurements of the materials as per recipe calculation to composites fabrications and samples preparation for testing of the products. However, the variation in density is found to have declining pattern with the increase in carbon fibers in the composite system. It maybe that lignocellulosic fibers are bulkier compared to carbon fibers; hence the process loss was higher.

Table 2
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					

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 of the internal bonding strength and flexural properties are displayed in **Fig. 2** (a and b). The curves shown a linear behavior at initial stage, however, became non-linear behavior after reached to the maximum load. However, the loading conditions causes a crack in the composite test specimens, which is continued with the extended level of displacements. As expected, 100% lignocellulosic fiber reinforced composites needed the lowest load to initiate the crack propagations, whereas the highest loads required by 20% carbon/ 80% lignocellulosic fiber reinforced epoxy resin. 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. However, the maximum load needed to break the 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. The similar consequences also noticed for flexural properties too. Highest loads need to bend the LE1 sample is 84.3 N, LE2 106.4 N for LE3 121.9 N, LE4 123.6 N, and LE5 is 131.6 N. The similar types of load/displacement results were also found for different other lignocellulosic panel products too (K. F. Hasan, Horváth, Kóczán, & Alpár, 2021; K. F. Hasan, P. G. Horváth, Z. Kóczán, M. Bak, & T. Alpár, 2021).

Like load-displacement curves, similar trends also seen for the mechanical properties of the products. 100% lignocellulosic fiber reinforced composites shown the lowest internal bonding strengths by 0.073 (0.005) MPa than that of 20% carbon fiber loaded composites with 80% lignocellulosic fiber by (0.147 (0.005)) MPa in the composite system. The other panels like LE2, LE3, and LE4 shown the intermediate strengths by 0.1 (0.016), 0.11 (0.016), and 0.135 (0.015) MPa, respectively. Additionally, the flexural strengths and modulus also followed the similar pattern of mechanical properties which is increased by the increase in carbon fibers in the composite system. MOR values of the composites were found to have the values by 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 case of flexural properties, strength and modulus values also increased with the increase in carbon fibers in the composite system whereas 100% lignocellulosic fiber reinforced composites shown the lowest values and 20% carbon fiber loaded composites with 80% lignocellulosic fiber shown the highest performances.

Morphological images of the developed composites are shown Fig. 3. The presence of fibers is observed as uniformly distributed throughout the composite system. The traces of matrix are seen into and around the fibers. A strong bonding among the fibers and epoxy resin is noticed, whereas the 100% lignocellulosic fiber reinforced composites are seeming to have weaker adhesions. Therefore, the fractured samples are easy to separate the fibers from matrix system without any major cracking. However, it was difficult to separate the fibers from matrix as there is a hole created in the composites (5% carbon/ 95% lignocellulosic materials, 10% carbon/ 90% lignocellulosic materials, 15% carbon/ 85% lignocellulosic materials, and 20% carbon/ 80% lignocellulosic materials) when the samples were pulled out during internal bonding strength tests. Therefore, the composites reinforced with carbon fibers showing better internal bonding strength and flexural properties too. Overall, SEM micrographs provided a strong evidence regarding the fiber materials adhesion with epoxy matrix in the composite system. The similar morphological analysis were also reported for flax/ carbon fiber loaded epoxy hybrid composites (Bagheri, El Sawi, Schemitsch, Zdero, & Bougherara, 2013; Dhakal & Sain, 2020).

Additionally, to investigate the chemical elements present in the control materials (both carbon and lignocellulosic fibers) and their associated composites, EDX test was carried out further. As expected, EDX analysis of control lignocellulosic fibers displayed a dominant presence of carbon and oxygen by 49.6 and 40.4%, respectively whereas carbon fibers shown 88.7% carbon, 6.2% oxygen, and 4.9% nitrogen. As carbon is the main chemical compound for both types of control materials, hence their reinforced composites also displayed the dominance in 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 too. Therefore, the weight% of carbon for 100% lignocellulosic fiber/ epoxy composite was 53.1%, 95% lignocellulosic fiber + 5% carbon fiber/ epoxy composite was 66%, 90% lignocellulosic fiber + 10% carbon fiber/ epoxy composite was 67.4%, 85% lignocellulosic fiber + 15% carbon fiber/ epoxy composite was 68%, and 80% lignocellulosic fiber + 20% carbon fiber/ epoxy composite was 72.6% (Fig. 4).

The reinforcement effects of lignocellulosic and carbon fibers with epoxy resin are 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. 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 shown the peak at 3333 cm^{-1} , whereas composite materials displayed the peak at 3334, 3336, 3334, 3333, and 3333 cm^{-1} . Albeit the control carbon fiber did not show any peak like this at this wavelength like as the cellulosic materials. A shift/stable bands are observed maybe after reinforcement with epoxy resin and carbon fibers. However, these bands are associated with the absorption of water from the surrounding atmosphere, therefore maybe absent from carbon fibers (neat). The similar patterns of lignocellulosic material reinforced composites are also goes in line with previous researches (K. Hasan, P. G. Horváth, Z. Kóczán, M. Bak, & T. Alpár, 2021; K. F. Hasan, P. G. Horváth, Z. Kóczán, M. Bak, et al., 2021). Neat epoxy resin also gives peak at 3364 cm^{-1} (Cecen, Seki, Sarikanat, & Tavman, 2008), which can be shifted a bit after the reinforcement with fibers. There is an interaction happens between the -OH groups of the resin and lignocellulosic and the carbon fiber in the composite system. 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 (Barbosa et al., 2017; Cecen et al., 2008). However, variations are very little/not significant in the composite products. Interestingly, the formation of bonds between carbon fiber and epoxy resin could be detected by the bonds around 2361 to 2362 cm^{-1} wavenumbers (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, Takagi, Nakagaito, Li, & Waterhouse, 2016). The peaks 1028 to 1247 cm^{-1} are denoted for C = C functional group present both in the control cellulosic fiber and composited products (Rizal, Gopakumar, Thalib, Huzni, & Abdul Khalil, 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 the cases of control cellulosic fiber and composited products (Rizal et al., 2018). Overall, the peaks within 3200 to 3600 cm^{-1} and 2361 to 2362 cm^{-1} confirms about the successful reinforcements of lignocellulosic/ carbon fibers with epoxy resin.

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

Compared to all the composites, carbon fiber loaded products displayed the better flame retardancy, whereas 20% carbon fiber incorporated products shown 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 facilitate to gradually decline this burning tendency (Fig. 6). Generally, wood is prone to be burnt when exposed to flame but conversely carbon fibers exist superior resistance against the fire (Grange et al., 2019). Therefore, as indoor products like furniture items are at a big risk of fire-related burning, hence 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, Hadden, & Bisby, 2019). The overall trends of flame retardancy sequence of the composites is $LE5 > LE4 > LE3 > LE2 > LE1$.

The moisture susceptibility is considered as one of the critical challenges of natural fiber reinforced composites. The reason behind this is the presence of hydrogen bond in fiber cell walls molecules as the plant-based fibers are comprised of dominating cellulosic and hemicellulosic polymers. The cellulosic materials are also composed of -OH (hydroxyl) and C (carbon). However, although cellulose polymer is enriched with crystalline regions which could retard the moisture penetrations, albeit they also contain amorphous regions which facilitates the water molecules to be defused and to break the hydrogen bonds (Moudood, Rahman, Öchsner, Islam, & Francucci, 2019). Therefore, an intermolecular distance is created in the cellulose chains and consequently the fibers are swollen. On the other hand, carbon fiber does not absorb moisture from surrounding environment or absorb water when immersed under the water (Zhai, Feng, Liu, & Li, 2016). Therefore, when the carbon fiber is loaded in the composite system with lignocellulosic materials, the affinity to absorb moisture by the test specimens are decreased. It is seen that (Fig. 7) 100% lignocellulosic materials reinforced composites absorb highest moisture by 3.72 (0.12) % whereas the lowest moisture was absorbed by 20% carbon/ 80% lignocellulosic materials reinforced composites by 3.13 (0.11)% after 2 h of oven drying. The modest moisture was absorbed by 10% carbon/ 90% lignocellulosic materials by 3.38 (0.12)% after the same period of immersion. However, the similar trends of moisture content also noticed even after 24 h of immersion, albeit an increase is observed compared to 2 h for overall moisture absorptions. Interestingly, the similar phenomenon also followed by the composite samples for water absorbency and thickness swelling too. The highest water as absorbed by the 100% lignocellulosic materials reinforced composites too showing the value 14.89 (0.56)% and lowest by 20% carbon/ 80% lignocellulosic materials 12.22 (0.29)% after 2 h of immersion. Moreover, highest thickness swelling was found in case of 100% lignocellulosic materials reinforced composites providing the value 18.97 (0.55)% and lowest one is 14.99 (0.23%) by 20% carbon/ 80% lignocellulosic materials. This phenomenon also agrees with our previous study for glass/flax fiber reinforced with MDI composites (K. F. Hasan, Horváth, & Alpár, 2021b). It seems the physical properties are becoming more stable with the incorporations of carbon fiber in the composite system.

Conclusions

Novel flame retardant composites were produced and reported in this current study produced from lignocellulosic materials and carbon fibers reinforced with epoxy resin. Pretreatment of the lignocellulosic fibers were performed before the fabrication methodology. The developed composites shown excellent flame

retardancy which increased with the increase in carbon fillers content in the composite system. The FTIR test results shown a successful bonding within the natural and synthetic fibers in the presence of epoxy resin. The morphological photographs further demonstrated a successful and efficient reinforcement effects in the composite system. However, the most prominent results were found in case of internal bonding strength, bending properties, which confirm a stronger material development. Moreover, fire retardant test also displayed confirmed excellent resistance capability against burning due to the incorporation of carbon fillers. SEM images of fractured surfaces further confirmed about the lower pulled out of carbon fiber reinforced composites from matrix system, therefore resulted for a better mechanical strengths compared to no carbon fiber loaded samples. Moreover, increased loading of carbon fibers also facilitate to absorb less moisture from surrounding environment by the composites. In summary, the developed products would facilitate the composite materials manufacturer and community with innovative approach of novel hybrid composites development.

Declarations

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Conflicts of Interest The authors declare that they have no conflicts of interest for the submitted work.

Compliance with ethical standards This research does not contain any investigation related to human participation or any animals conducted by the authors. In this regard, authors are claiming the compliance of this work with ethical standards.

References

1. Akpan EI, Wetzal B, Friedrich K (2019) Processing and properties of short wood fiber/acrylate resin composites. *Polym Compos* 40:91–98. doi:<https://doi.org/10.1002/pc.24604>
2. Bagheri ZS, Sawi E, Schemitsch I, Zdero EH, Bougherara H (2013) Biomechanical properties of an advanced new carbon/flax/epoxy composite material for bone plate applications. *J Mech Behav Biomed Mater* 20:398–406. doi:<https://doi.org/10.1016/j.jmbbm.2012.12.013>
3. Barbosa APC, Fulco APP, Guerra ES, Arakaki FK, Tosatto M, Costa MCB, Melo JDD (2017) Accelerated aging effects on carbon fiber/epoxy composites. *Compos Part B Eng* 110:298–306. doi:<https://doi.org/10.1016/j.compositesb.2016.11.004>
4. Bartlett AI, Hadden RM, Bisby LA (2019) A review of factors affecting the burning behaviour of wood for application to tall timber construction. *Fire Technol* 55:1–49. doi:<https://doi.org/10.1007/s10694-018-0787-y>

5. Bhatt P, Goe A (2017) Carbon fibres: production, properties and potential use. *Mater Sci Res India* 14:52–57. doi:<http://dx.doi.org/10.13005/msri/140109>
6. Cai M, Takagi H, Nakagaito AN, Li Y, Waterhouse GI (2016) Effect of alkali treatment on interfacial bonding in abaca fiber-reinforced composites. *Compos Part A Appl Sc Manuf* 90:589–597. doi:<https://doi.org/10.1016/j.compositesa.2016.08.025>
7. Cecen V, Seki Y, Sarikanat M, Tavman IH (2008) FTIR and SEM analysis of polyester-and epoxy-based composites manufactured by VARTM process. *J Appl Polym Sci* 108:2163–2170. doi:<https://doi.org/10.1002/app.27857>
8. Dhakal HN, Sain M (2020) Enhancement of mechanical properties of flax-epoxy composite with carbon fibre hybridisation for lightweight applications. *Materials* 13:109. doi:<https://doi.org/10.3390/ma13010109>
9. Grange N, Manescau B, Chetehouna K, Gascoin N, Lamoot L, Coppalle A, Reynaud I (2019) Fire resistance of carbon-based composite materials under both ideal and realistic normative configurations. *Appl Therm Eng* 159:113834. doi:<https://doi.org/10.1016/j.applthermaleng.2019.113834>
10. Guo G, Finkenstadt VL, Nimmagadda Y (2019) Mechanical properties and water absorption behavior of injection-molded wood fiber/carbon fiber high-density polyethylene hybrid composites. *Adv Compos Hybrid Mater* 2:690–700. doi:<https://doi.org/10.1007/s42114-019-00116-5>
11. Hasan K, Fatima Zohra B, Bak M, Horváth PG, Dénes L, Alpár TL (2020) *Effects of cement on lignocellulosic fibres*. Paper presented at the 9th Hardwood Proceedings, Sopron, Hungary
12. Hasan K, Horváth PG, Alpár T (2021) Potential fabric-reinforced composites: a comprehensive review. *J Mater Sci* 56: 14381–14415. doi:<https://link.springer.com/article/10.1007/s10853-021-06177-6>
13. Hasan K, Horváth PG, Kóczán Z, Bak M, Alpár T (2021) Colorful and facile in situ nanosilver coating on sisal/cotton interwoven fabrics mediated from European larch heartwood. *Sci Rep* 11:1–13. doi:<https://doi.org/10.1038/s41598-021-01914-y>
14. Hasan K, Horváth PG, Kóczán Z, Le DHA, Bak M, Bejó L, Alpár T (2021) Novel insulation panels development from multilayered coir short and long fiber reinforced phenol formaldehyde polymeric biocomposites. *J Polym Res* 28:1–16. doi:<https://doi.org/10.1007/s10965-021-02818-1>
15. Hasan KF, Horváth PG, Alpár T (2021) Development of lignocellulosic fiber reinforced cement composite panels using semi-dry technology. *Cellulose* 28:3631–3645. doi:<https://doi.org/10.1007/s10570-021-03755-4>
16. Hasan KF, Horváth PG, Baş S, Alpár T (2021) Industrial Flame Retardants for Polyurethanes. *Materials and Chemistry of Flame-Retardant Polyurethanes Volume 1: A Fundamental Approach*. ACS Publications, Washington, DC, USA, pp 239–264
17. Hasan KF, Horváth PG, Kóczán Z, Alpár T (2021) Thermo-mechanical properties of pretreated coir fiber and fibrous chips reinforced multilayered composites. *Sci Rep* 11:1–13. doi:<https://doi.org/10.1038/s41598-021-83140-0>
18. Hasan KF, Horváth PG, Kóczán Z, Bak M, Alpár T (2021) Semi-dry technology-mediated coir fiber and Scots pine particle-reinforced sustainable cementitious composite panels. *Constr Build Mater* 305:124816. doi:<http://dx.doi.org/10.1016/j.conbuildmat.2021.124816>

19. Hasan KF, Horváth PG, Miklos B, Alpár T (2021) A state-of-the-art review on coir fiber-reinforced biocomposites. RSC Adv 11:10548–10571. doi:<https://doi.org/10.1039/D1RA00231G>
20. Hasan KF, Horváth PG, Zsolt K, Alpár T (2021) Design and fabrication technology in biocomposites manufacturing. Value-added biocomposites: technology, innovation, and opportunity. CRC Press, Boca Raton, USA, pp 158–183
21. Hasan KF, Horváth PG, Zsolt K, Kóczán Z, Bak M, Horváth A, Alpár T (2021) Hemp/glass woven fabric reinforced laminated nanocomposites via in-situ synthesized silver nanoparticles from *Tilia cordata* leaf extract. Compos Interfaces 1–19. doi:<http://dx.doi.org/10.1080/09276440.2021.1979752>
22. Hasan KF, Horváth P (2021a) t. G. r., & Alpár, T. Nanotechnology for waste wood recycling. In Nanotechnology in Paper and Wood Engineering. Duxford, United Kingdom: Woodhead Publishing
23. Hasan KF, Horváth P, Alpár T (2021b) Thermomechanical Behavior of Methylene Diphenyl Diisocyanate-Bonded Flax/Glass Woven Fabric Reinforced Laminated Composites. ACS Omega 6: 6124–6133. doi:<https://doi.org/10.1680/jgrma.20.00052>
24. Hasan KMF, Horváth PG, Alpár T (2021) Lignocellulosic Fiber Cement Compatibility: A State of the Art Review. J Natur Fibers 1–26. doi:<https://doi.org/10.1080/15440478.2021.1875380>
25. Hasan KMF, György P, Tibor A (2020) Thermomechanical Behavior of Methylene Diphenyl Diisocyanate-Bonded Flax/Glass Woven Fabric Reinforced Laminated Composites. ACS Omega 6:6124–6133. doi:<https://dx.doi.org/10.1021/acsomega.0c04798>
26. Ignitability test EN ISO 11925-2. EUROCLASS, System Retrieved from <http://virtual.vtt.fi/virtual/innofirewood/stateoftheart/database/euroclass/euroclass.html#ignitabilitytest>
27. Kamau-Devers K, Miller SA (2020) The environmental attributes of wood fiber composites with bio-based or petroleum-based plastics. Int J Life Cycle Assess 25:1145–1159. doi:<https://doi.org/10.1007/s11367-020-01744-6>
28. Kibleur P, Aelterman J, Boone MN, Van den Bulcke J, Van Acker J (2022) Deep learning segmentation of wood fiber bundles in fiberboards. Compos Sci Technol 221:109287. doi:<https://doi.org/10.1016/j.compscitech.2022.109287>
29. Mahmud S, Hasan KF, Jahid MA, Mohiuddin K, Zhang R, Zhu J (2021) Comprehensive review on plant fiber-reinforced polymeric biocomposites. J Maters Sci 56:7231–7264. doi:<https://doi.org/10.1007/s10853-021-05774-9>
30. Moudood A, Rahman A, Öchsner A, Islam M, Francucci G (2019) Flax fiber and its composites: An overview of water and moisture absorption impact on their performance. J ReinfPlast Compos 38:323–339. <https://doi.org/10.1177%2F0731684418818893>
31. Rizal S, Gopakumar DA, Thalib S, Huzni S, Abdul Khalil H (2018) Interfacial compatibility evaluation on the fiber treatment in the Typha fiber reinforced epoxy composites and their effect on the chemical and mechanical properties. Polymers 10:1316. doi:<https://doi.org/10.3390/polym10121316>
32. Tibor L (2012) Alpar- Éva Selmeczi, L. C. March 26–27). *Advanced wood cement compatibility with nano mineral*. Paper presented at the International Scientific Conference on Sustainable Development & Ecological Footprint, Sopron, Hungary

33. Tibor LA, Péter GH, Hasan KMF (2021) Introduction to biomass and biocomposites. Toward the value-added biocomposites: technology, innovation and opportunity. CRC Press, Boca Raton, USA
34. Tripathi L, Behera B (2021) 3D woven honeycomb composites. J Mater Sci 56:15609–15652
35. Zhai Z, Feng L, Liu Z, Li G (2016) Water absorption test for carbon fiber epoxy resin composite based on electrical resistance. Polym Test 56:394–397. doi:<https://doi.org/10.1016/j.polymertesting.2016.10.020>
36. Zhang L, Chen S, Pan Y-T, Zhang S, Nie S, Wei P, Wang D-Y (2019) Nickel metal–organic framework derived hierarchically mesoporous nickel phosphate toward smoke suppression and mechanical enhancement of intumescent flame retardant wood fiber/poly (lactic acid) composites. ACS Sustain Chem Eng 7:9272–9280. doi:<https://doi.org/10.1021/acssuschemeng.9b00174>

Figures

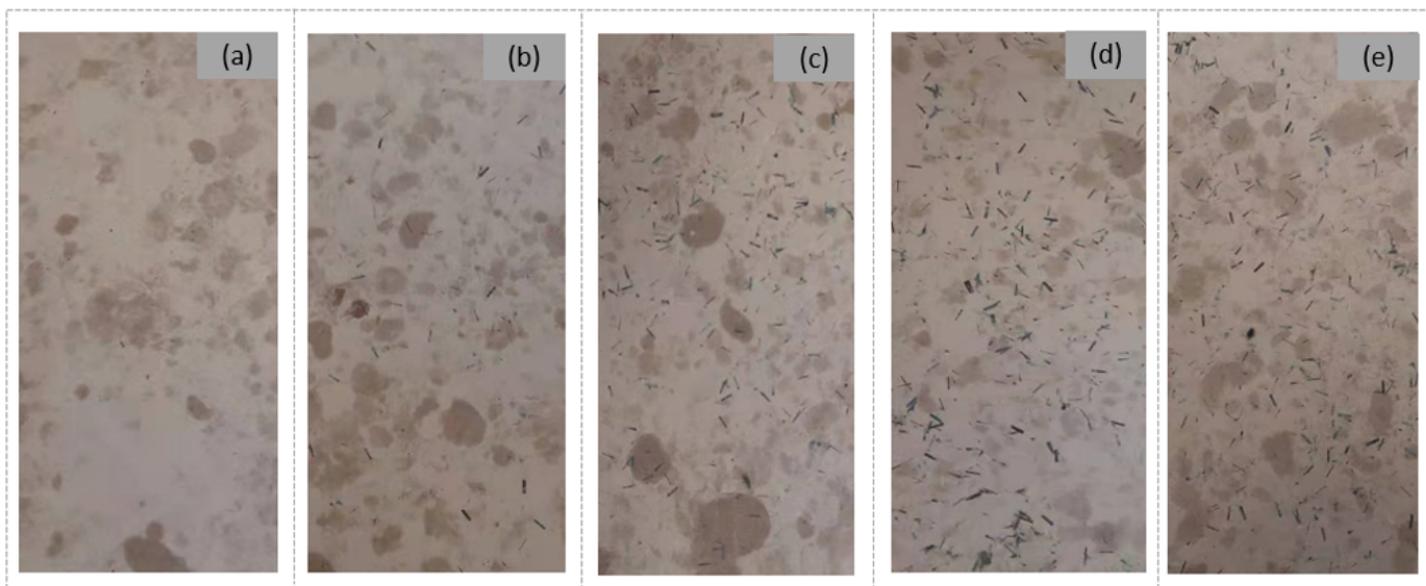


Figure 1

Photographs of lignocellulosic fiber/carbon fiber reinforced epoxy composites

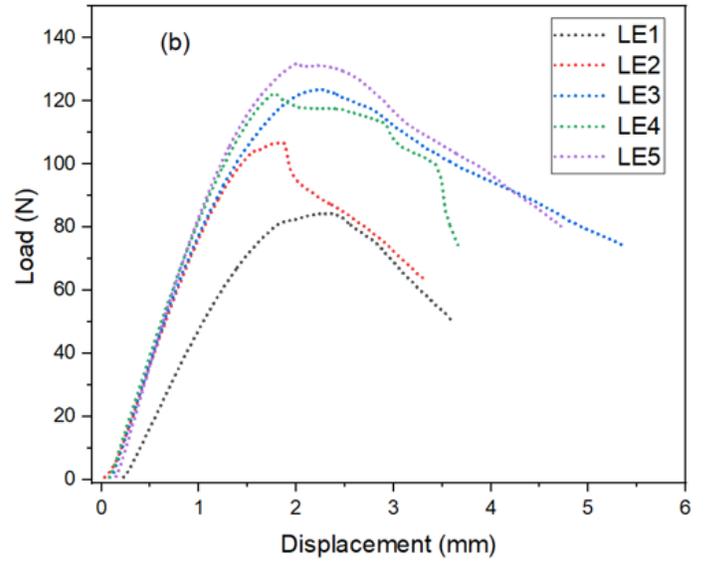
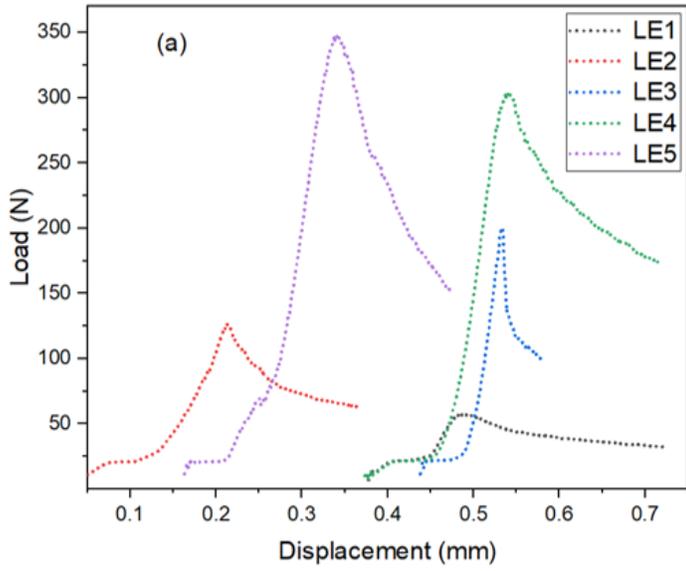


Figure 2

Load versus displacement curves lignocellulosic /carbon fiber reinforced epoxy composites: (a) Internal bonding strength and (b) Flexural properties

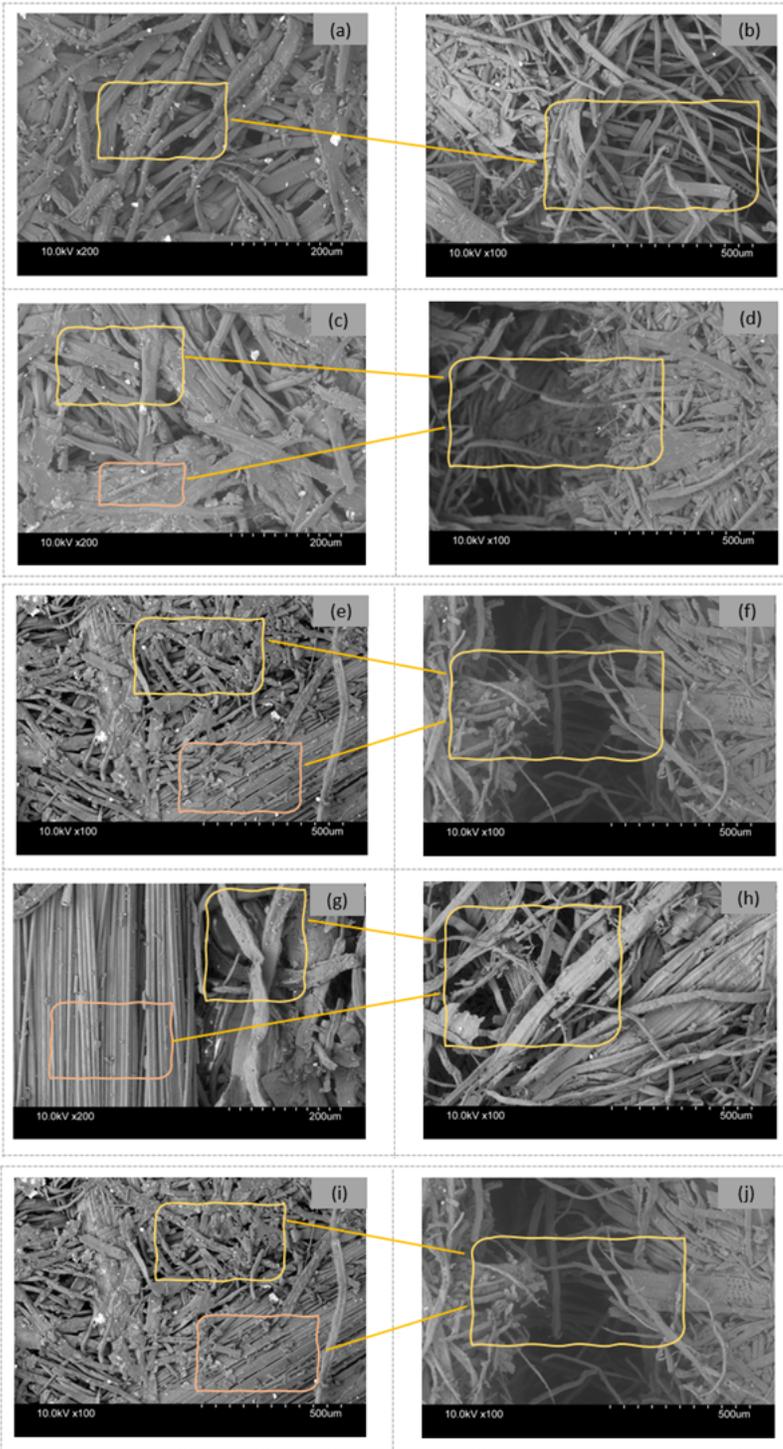


Figure 3

SEM analysis of lignocellulosic/ carbon fiber reinforced epoxy composites

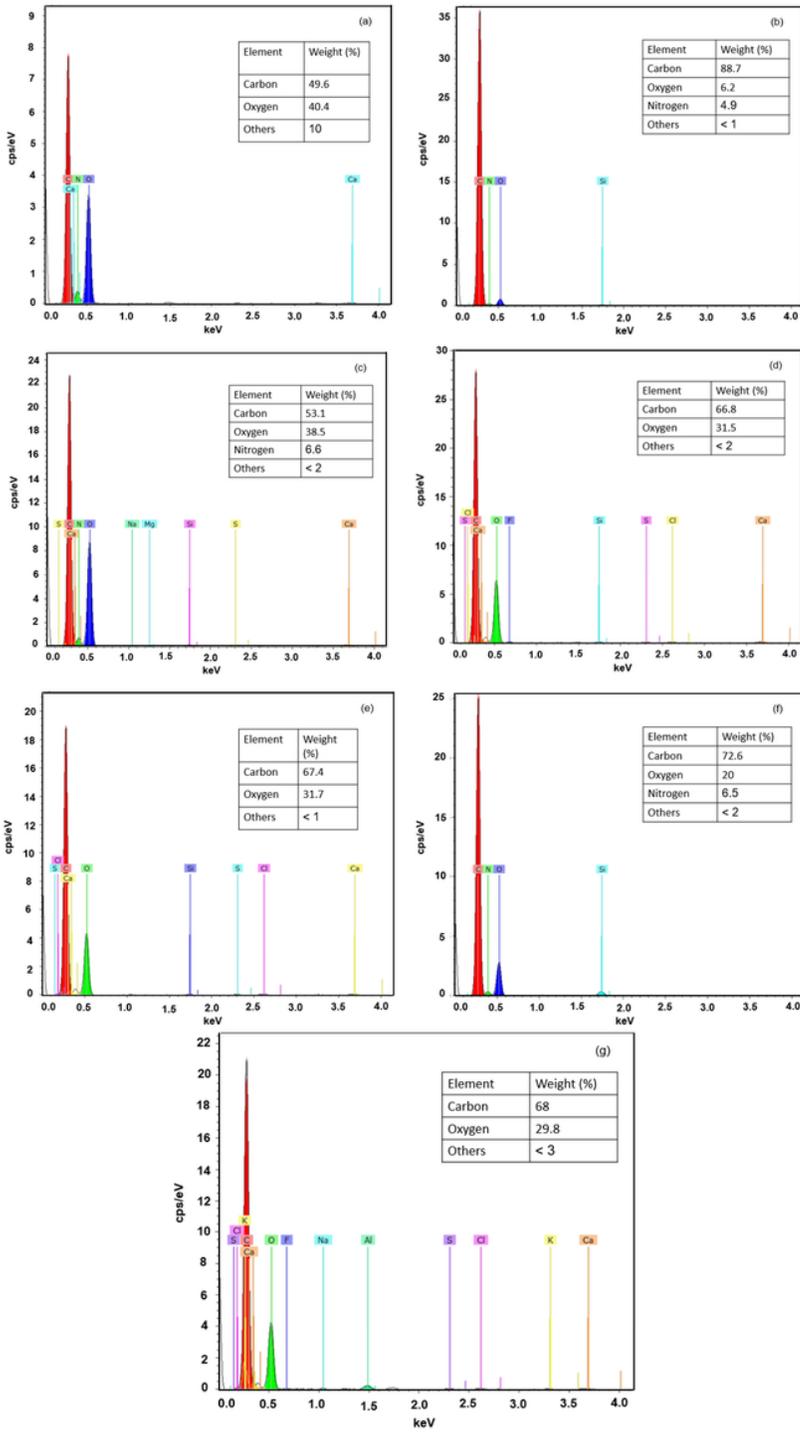


Figure 4

EDX investigation of lignocellulosic fiber/carbon fiber reinforced epoxy composites

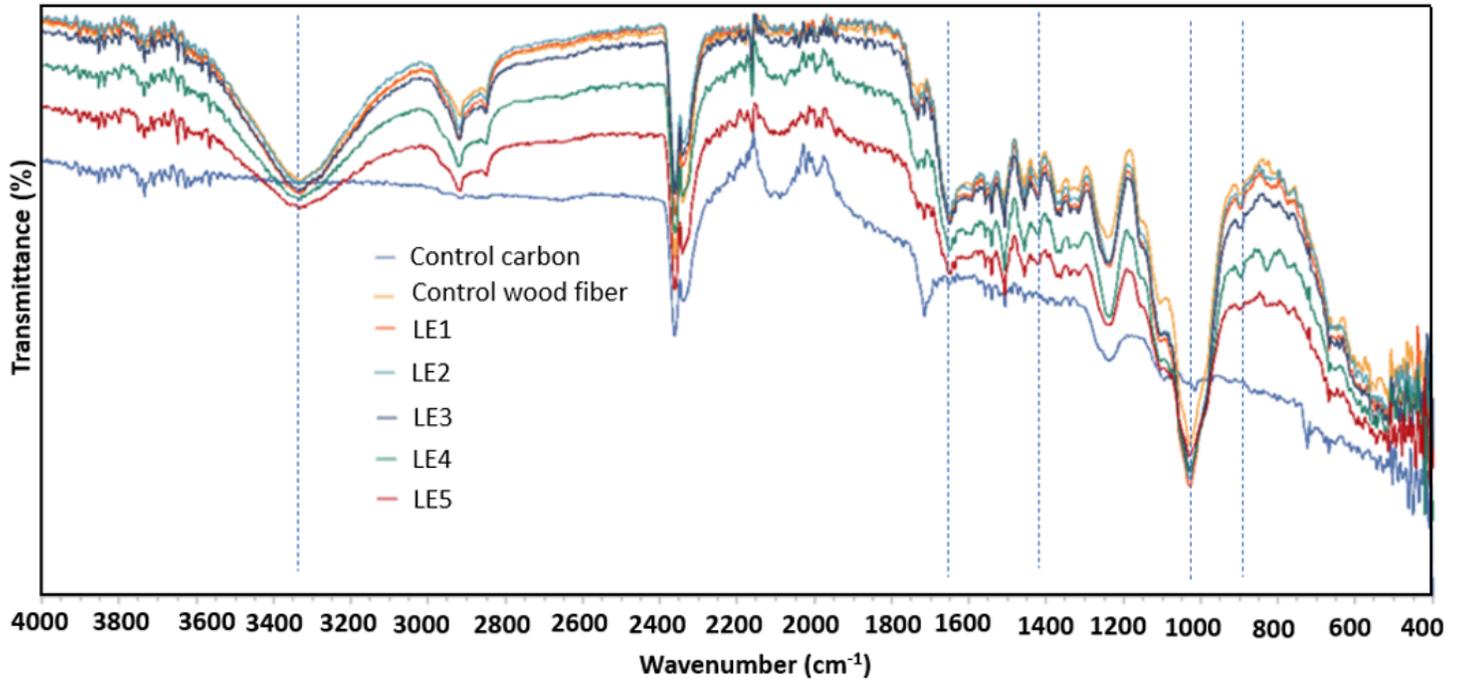


Figure 5

FTIR analysis of lignocellulosic / carbon fiber reinforced epoxy composites

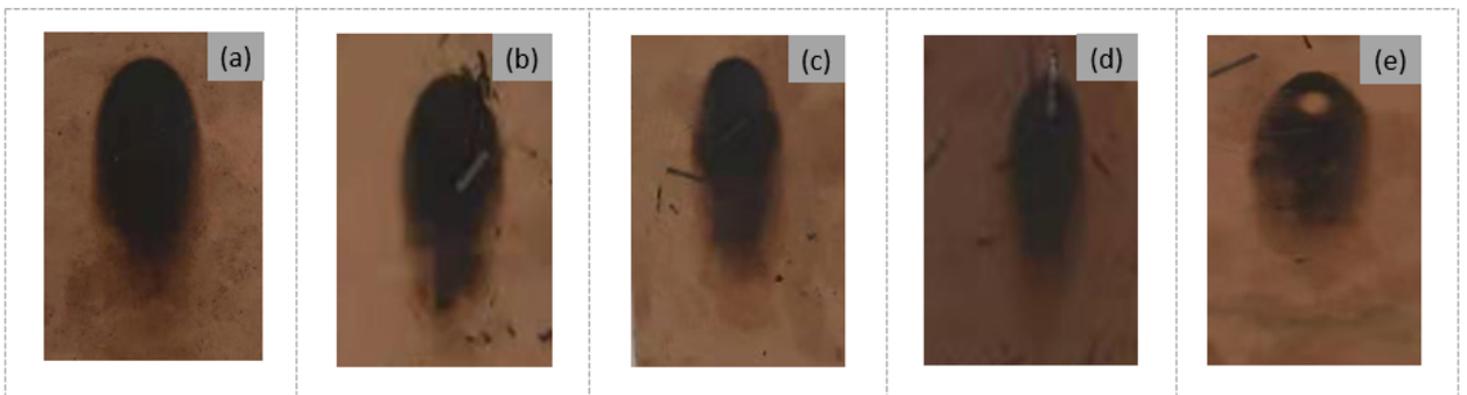


Figure 6

Flame retardancy test of lignocellulosic fiber/carbon fiber reinforced epoxy composites

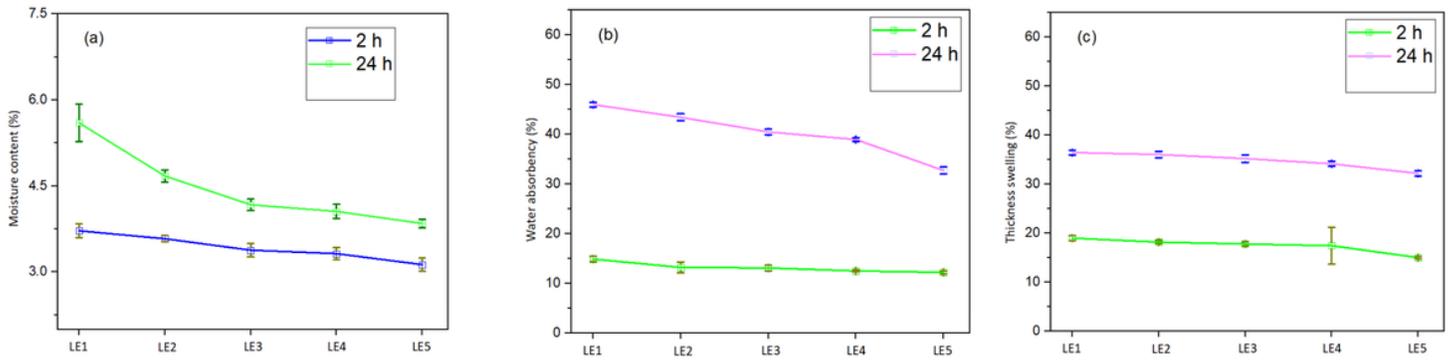


Figure 7

Physical properties of lignocellulosic fiber/carbon fiber reinforced epoxy composites

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