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Semi-dry technology-mediated coir fiber and Scots pine particle-reinforced sustainable cementitious composite panels

K.M. Faridul Hasan [a,*], Péter György Horváth [a], Zsófia Kóczán [b], Miklós Bak [a], Tibor Alpár [a,*]

[a] Simonyi Károly Faculty of Engineering, University of Sopron, Sopron 9400, Hungary
[b] Paper Research Institute, Simonyi Károly Faculty of Engineering, University of Sopron, Sopron 9400, Hungary

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ABSTRACT

The demand for natural fiber-reinforced sustainable products is increasing continuously worldwide. The technology to produce lignocellulosic fiber-reinforced cementitious materials is currently limited; hence, new routes for composite development are being explored. The current study presents an innovative semi-dry technology implemented on lignocellulosic material (coir fiber and Scots pine particle) reinforced composite panels. Composite panels of 12 mm thickness and 1200 kg/m^3 nominal densities were prepared from lignocellulosic materials and OPC (Ordinary Portland cement). Measurements determined the dimensions of the lignocellulosic materials to be the following: coir fibers were within 0.1 to 1.25 mm in length while Scots pine particles ranged from 0.355 to 1.6 mm in length. The lignocellulosic materials were loaded in different proportions (100% Scots pine, 60% Scots pine/40% coir, 50% Scots pine/50% coir, 40% Scots pine/60% coir, and 100% coir) to produce five composite panels. The proportions of water glass additive and OPC remained constant. A cheaper and more convenient novel fabrication approach (semi-dry technology) was utilized to produce the composite panels via pressing implementation (3.2 to 7.1 MPa). Finally, the thermal, physical, morphological, and mechanical properties of all the composite panels were tested. The experimental results were significant and could facilitate composite manufacturers with potential and sustainable products possessing superior performance characteristics. The most promising finding of this research is the increased thermal and mechanical properties of the composites along with significant dimensional stability against moisture with the increase in loading of coir fibers in the matrix system. The SEM (scanning electron microscopy) and SEM-mediated EDX (energy-dispersive X-ray spectroscopy) also provided the morphological and elemental analysis of the fibers, cements, and associated composites. The FTIR (Fourier transform infrared spectroscopy) study also further confirmed the successful reinforcement of the lignocellulosic materials and OPC. Moreover, the perceived thermal conductivity of the composite materials also indicates promising routes toward the development of insulation materials using renewable lignocellulosic materials.

1. Introduction

Plant-based renewable materials exhibit notable potential to help shift the world from synthetic products to green and eco-friendly products. Cementitious materials are extensively used in building, construction, and structural applications; however, the increasing global demand for sustainable products has intensified the pressure on scientists and manufacturers to keep up with demand. In this regard, plant-based renewable lignocellulosic materials like coir, jute, sisal, abaca, hemp, flax, ramie, spruce, Scots pine, beech, and Turkey oak [1-4] could all be crucial choices. The chemical constituents of naturally derived plant materials like lignin, cellulose, hemicellulose, and other materials [5-8] are all potential candidates for cementitious reinforcements with reagents like OPC. Current efforts to develop OPC bonded lignocellulosic composite materials are tremendous. However, drawbacks continue to linger, particularly in selecting optimal reinforcement materials (lignocellulosic) to achieve satisfactory thermo-mechanical performances. Our previous study [9,10] attempted to utilize lignocellulosic saw dust (industrial byproducts) to develop cementitious materials, but our experiments could not attain internal bonding strengths higher than 0.5 MPa. Consequently, we began investigating lignocellulosic materials that are more convenient and discovered the following: both coir fiber and pine

* Corresponding authors.
E-mail addresses: faridulwtu@outlook.com (K.M.F. Hasan), alpar.tibor@uni-sopron.hu (T. Alpár).

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particles reinforced composites provide internal bonding strengths higher than 0.5 MPa even when mixed together. Moreover, cement incompatibility with lignocellulosic materials [11] is another significant drawback that needs to be overcome to attain satisfactory performances. Generally, cement is alkaline in nature and on its surface it possesses the hydroxyl (–OH) group, which intensifies in wet conditions that have higher pH values [9,12]. This phenomenon is responsible for the decay of hemicellulose from the polymeric structures of plant-based materials, resulting in significant strength loss.

Additionally, clinker materials like dicalcium silicate and tricalcium silicate help the cement set by creating calcium silicate hydrate through exothermic reactions in the presence of water. Water-soluble polymeric compounds like hemicellulose, sugar, and tannin also restrict the compatibility between the cements and plant-derived lignocellulosic materials. Hence, it is better to use additive materials like Na_2SiO_3 (water glass/ sodium silicate), $MgCl_2$ (magnesium chloride), $CaCl_2$ (calcium chloride), and $Al_2(SO)_4)_3$ (aluminum silicate) [11,13]. The pretreatment of the lignocellulosic materials could help facilitate improved compatibility between natural fiber/wood and cements. However, pretreatment is unnecessary if the reinforcement materials have no impurities or are below the acceptable limits of 0.4% for tannin content and 0.5% for sugar content [11]. These limits necessitate the examination of sugar and tannin contents of the materials prior to composite panel manufacturing. However, the minimized inhibition between lignocellulosic materials and cement facilitates higher strengths in the composite panels. Moreover, two ratios – the type of wood species/lignocellulosic materials and cement ratio, and the water to cement ratio – also play vital roles in cementitious composite panels [14].

The proper selection of lignocellulosic materials like plants is important as not all plant-derived materials are suitable for cementitious materials production due to the differing chemical structures of various plants [15,16]. Additionally, plant growth, plant maturity, and the harvesting period/season also determine the strengths of lignocellulosic materials and composited materials. Research studies exploring the reinforcement possibilities of Scots pine with OPC [13,17] have been conducted. However, according to our knowledge, no research studies concentrating on the hybridization of Scots pine with natural lignocellulosic fibers like coir have been completed yet. Scots pine is extensively available in central European countries like Hungary. Csoka et al. reported that Scots pine comprised around 9% of Hungary's forested area in 1993 [18]. On the other hand, coir fibers are also popular seed fibers that have been widely grown in some Asian countries for a long time [19]. Coconuts are primarily grown for their water and fruit, not their shells. Once the water and fruit have been extracted from the coconut, the husks, which do not biodegrade quickly or easily, are conventionally disposed of as waste. This practice creates numerous environmental burdens. However, coconut husks could be used as a potential raw material for biocomposite and cementitious materials as well. It has been reported that coconut husks contain around 30% fiber materials and 70% coir piths [20]. Coir has significant potentiality to improve the thermomechanical properties of hybrid composite panels when mixed with Scots pine and OPC, which is the focus of the current research study. Moreover, both coir and Scots pine contain significant amounts of cellulose, hemicellulose, lignin, and pectin in their polymeric structures (Table 1). As Table 1 shows, coir fiber contains more lignin (41 to 45%) than Scots pine (27 to 29%), which may facilitate coir with higher mechanical properties in the produced composite panels. Moreover, the higher lignin content in coir also makes it more durable, rot-resistant, and elastic [20].

Table 1
Chemical components present in coir fiber and Scots pine material.

Constituent polymers	Coir fiber (wt%) [26]	Scots pine (wt%) [27]
Cellulose	36–43	44–46
Hemicellulose	10–20	25–28
Lignin	41–45	27–29
Pectin	3–4	15
Moisture content	8	12.1
Others	–	3–4

Many different types of cements – such as Portland, aluminate, sulfate, phosphate, and sulfo-aluminate – are currently available [21]. Some derivatives of Portland, like OPC, Portland blast furnace, Portland pozzolana, fly ash, etc., are among the most widely and commonly used cement matrixes [21]. OPC is a widely used binding reagent due to its cost effectiveness and availability [22]. The current study used OPC type I cement to develop the composite materials. CaO and SiO_2 are the two main chemical constituents of cements and comprise 64.18% and 21.02%, respectively. Al_2O_3, Fe_2O_3, MgO, SO_3, Na_2O, K_2O, TiO_2, Cl, P_2O_5, and Cr_2O_3 are also present in varying proportions (Table 2).

Moreover, the utilization of renewable lignocellulosic materials for flooring, wall, and roofing application in the building and construction sector could expedite lower costs, lower energy consumption, lower environmental burdens, and thermal comfort. Consequently, cementitious materials developed from renewable plant-derived resources and OPC could encourage construction that is more environmentally sustainable. Though forested areas in the world continue to shrink, the combination of natural fibers with plants [23] could accelerate potential alternative lignocellulosic materials for OPC bonding via hybrid composite production. The current study implemented a semi-dry technology to produce composite panels from coir fibers and Scots pine particles and their associated composites. These are similar to the wood particle/cement composite panels used by the manufacturing industry. Comparatively, semi-dry technology is cheaper and more convenient as it is less capital intensive and less labor intensive [9,24,25]. However, research on semi-dry technology oriented natural fiber-reinforced cementitious materials remains limited. In this regard, the current research developed cementitious composite panels using semi-dry technology, which is a novel and innovative fabrication method for Scots pine/coir fiber and OPC that, to our knowledge, has not been reported yet. Moreover, this research will facilitate manufacturers of engineered construction materials by means of an economic, green, and feasible method of hybrid production as well as composite panel production.

2. Materials and methods

2.1. Materials

The Scots pine (*Pinus sylvestris*) particles were provided by FALCO Woodworking Co., in Szombathely, Hungary. The coir (*cocos nucifera*) fibrous chips were obtained from a local company named Pro Horto Ltd. located in Szentes, Hungary. Table 1 lists the chemical components of Scots pine and coir fibers. The Scots pine particles were used as received

Table 2
Chemical compositions of OPC materials.

Chemical component	OPC type I (wt%) by Ferreiro et al. [28]
CaO	64.49
SiO_2	19.01
Al_2O_3	5.51
Fe_2O_3	3.81
MgO	0.99
SO_3	3.69
K_2O	0.43
Na_2O	0.28
Loss in ignition	2.34
TiO_2	0.27
Cl	0.01
P_2O_5	0.35
Cr_2O_3	0.01

*OPC- Ordinary Portland cement.

from the company, while the coir fibrous chips were defibrated before they were fabricated into composite panels. Typical OPC CEM I 42.5 was used as the cement matrix for reinforcing Scots pine and coir fibers in order to manufacture the composite panels. The FALCO Woodworking Co. also provided the cement, which was Imanufactured by Duna-Dráva Cement Kft. in Vác, Hungary. Table 2 provides the chemical constituents of typical OPC type I cement. Fig. 1 presents the physical and morphological photographs of the Scots pine, coir fibers, and OPC. The additive reagent (water glass, Na_2SiO_3) was procured from Sigma Aldrich, Hungary.

2.2. Methods

2.2.1. Preparation of coir fiber from chips and sieving of coir fiber and Scots pine

The coir fibrous chips were defibrated with a VZ 23412 model defibrating machine (Dinamo Budapest, Hungary). The defibration procedure was performed carefully by adjusting the grinders and grain distance to protect the extracted fibers from damage. Later, both the extracted fibers and Scots pines were sieved (sieve analyzer, ANALYSETTE 3Pro, Fritsch, Germany) to ensure uniform lengths for composite panel production. Our previous studies discuss the detailed sieving protocol for fiber/chips materials [9,29]. The length of the sieved coir fibers ranged within 0.1–1.25 mm, and for Scots pine this range was 0.355 to 1.6 mm. The sieve test revealed that around 42.3% of the coir fibers possessed a length of 1.25 mm (highest value), whereas the lowest proportion was 7.4%, corresponding to a 0.8 mm length (Fig. 2). However, two types of fiber lengths, 0.355 mm (26.6%) and 0.1 mm (22.5%), were also present. Conversely, the highest length obtained for Scots pine particles was 1.6 mm, which comprised 33.4% of the particles, whereas the lowest portions of Scots pine were 0.5 mm lengths, comprising 10.6%. Two other Scots pine lengths included 1.0 mm,

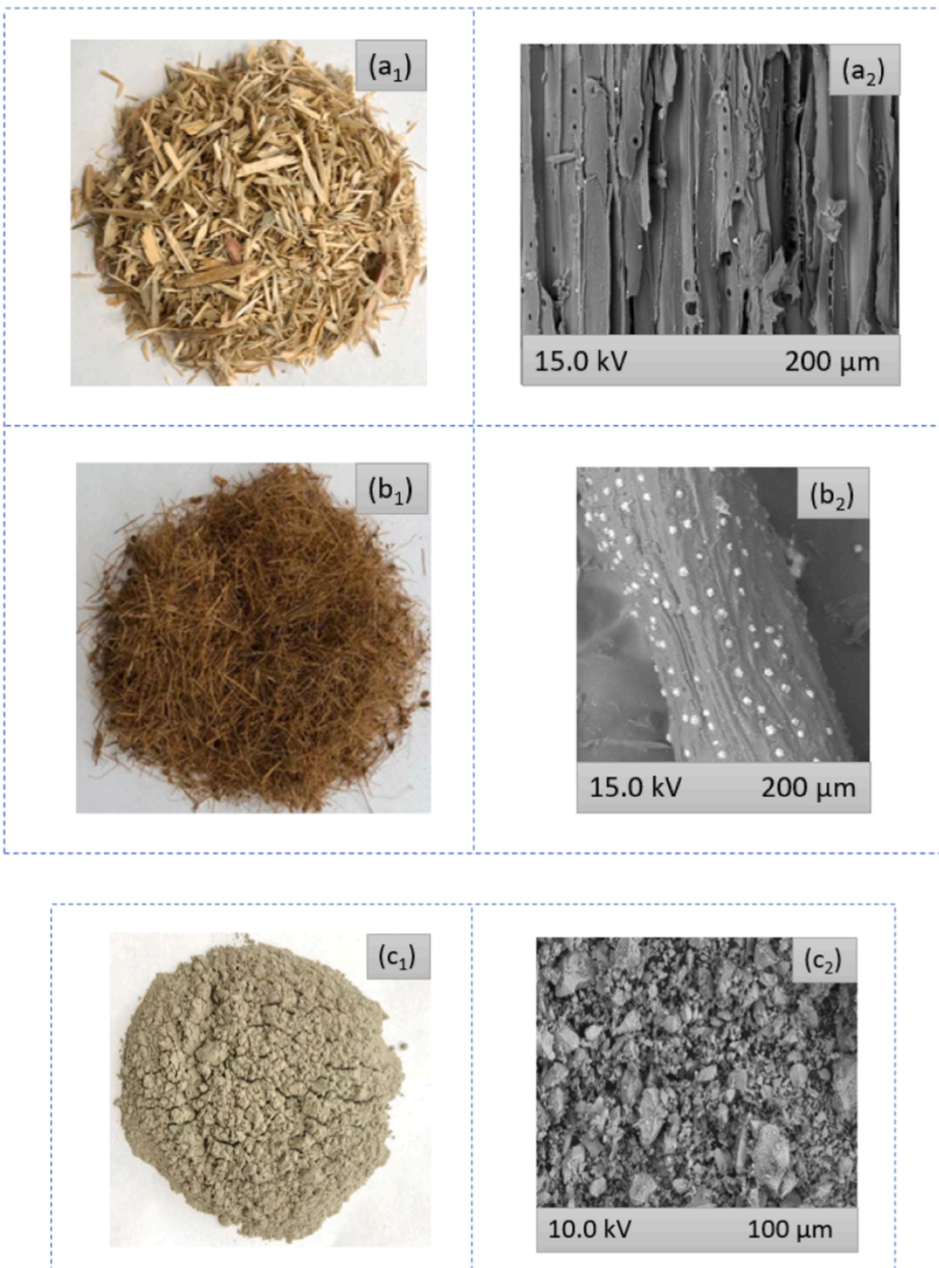

Fig. 1. Physical and morphological photographs of coir fiber, Scots pine material, and OPC material: (a_1) Physical photographs of Scots pine material; (a_2) SEM image of Scots pine material; (b_1) Physical photographs of coir fiber; (b_2) SEM images of coir fiber; (c_1) Physical photographs of OPC; (c_2) SEM image of OPC.

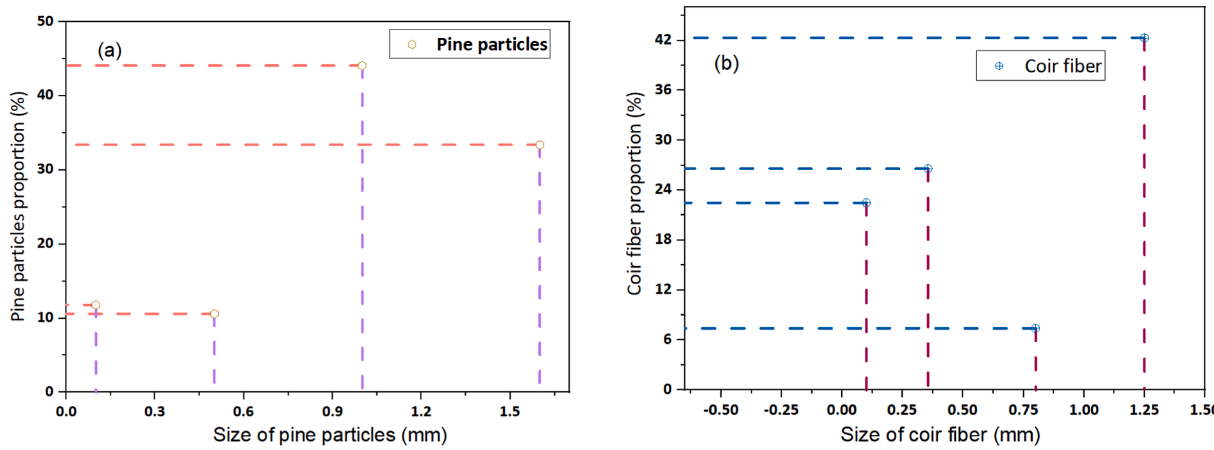

Fig. 2. Size distribution of Scots pine and coir fiber (a and b, respectively).

which made up 44.1%, and 0.1 mm, which encompassed 11.8%. Overall, the dominant lignocellulosic material size for coir fiber was 1.25 mm and 1.00 mm for Scots pine.

2.2.2. *Production of composites panels*

The moisture contents of both types of sieved materials were measured as per EN 322:1993 standards. The accuracy of the measurement was 0.001 g and dry temperature was 105 ± 0.3 °C. The investigated moisture content for coir was 11.8 (0.01%) and for Scots pine it was 35.8 (0.3%) for six consecutive tests. Later, all the lignocellulosic materials, OPC, and Na_2SiO_3 were measured proportionately, as noted in Table 3. Composite panel 1, denoted by P@C1, is made of 100% Scots pine, while composite panel 6, C@C1, is 100% coir fiber. The hybrid composites were denoted by PC@C2, PC@C3, and PC@C4, respectively and the proportions of Scots pine and coir fibers were 0.6/0.4, 0.5/0.5, 0.4/0.6, respectively. Since a main focus areas of this research was to investigate the effects of increased loading of coir fiber with Scots pine in the composites system, the proportions of OPC, Na_2siO_3, and cement stones were kept constant for all the recipes at 2.6, 0.052, and 0.52, respectively. Though the materials mentioned in Table 3 were weighed in g, they are reported here as proportions for all the composite panels individually. Another important material in this research is mixing water (H_2O), which was weighed by 50% of total dry matter content in every recipe. The water content was further measured in terms of composite panel density, dimensions, moisture content of Scots pine and coir fibers, and water glass. The nominal densities of the composite panels were set to 1200 kg/m^3 each and the dimensions were 400 × 400 × 12 mm^3. Later, the slurry was prepared by uniformly mixing lignocellulosic materials, OPC, water glass, and cement stone. To prepare the slurry, the lignocellulosic materials were placed into a steel drum into which the OPC was gradually poured while being constantly stirred with a magnetic stirrer. Once the OPC and lignocellulosic materials were optimally mixed, the water glass and water solution was poured in following the same protocol used for the cement additions. Water glass facilitates the perfect setting of OPC with lignocellulosic materials. Finally, the mixed ingredients in the drum were stirred for another three minutes to confirm the even mixing of all the materials to ensure a superior slurry. Later, a wooden frame of 400 × 400 mm^2 was placed on a table over a long polybag where the prepared slurry was uniformly spread. When the slurry spreading was complete, another wooden lid was used to manually press the materials in the mat to ensure they adhered without any deformations. After that, the material proceeded to mechanical pressing. Subsequently, the frame was withdrawn from the mat and covered by the same polybag. Two 12 mm steel rods were placed at the two sides of the mat and a steel plate was inserted over it. Finally, the mat containing the steel plate was placed to the platen of the pressing machine where 7.2 MPa pressure was applied on the mat. Pressure was applied constantly for 24 h durations at environmental temperature and humidity. Later, the pressure was gradually decreased and the panel was removed from the plate. A 28-day curing period followed. All the composite panels (Fig. 3) were manufactured following the same protocols.

2.3. *Characterizations*

The moisture contents of coir fiber and Scots pines were measured according to EN 322:1993 using a Kern ULB 50-3N model moisture analyzer by the KERN AND SOHN GmbH company, Germany. The sugar and tannin contents of the fibers were investigated by following analytical methods in the laboratory. A sand bath was used for boiling the water with coir fiber and Scots pine during the tannin and sugar contents testing protocol. The thermal conductivity of the developed cementitious composites was tested according to the MSZ EN ISO 10456 [30] (goes in line with Hungary, Europe, and ISO) standard following hot plate method. A custom made equipment by Technical University Budapest, Hungary was designed and assembled for measuring the thermal conductivity. The thermal conductivity was measured for homogeneous materials. Before the test, a standard calibration of the machine was performed. The composite of 300 mm × 300 mm was placed between the hot and cold plates. The temperature of the plates was set prior the test, hot surface, T_1 and cold surface T_2. These are heated by using different heating circuits. The steady state condition attainments were checked through installing various thermocouples on both hot and cold sides. The thermal conductivity was calculated by

Table 3
Experimental design of Scots pine and coir fiber-reinforced OPC composite panel manufacturing.

Composite materials	SP (proportion)	CF (proportion)	OPC (proportion)	Na_2SiO_3 (proportion)	CS (proportion)
P@C1	1.00	0	2.6	0.052	0.52
PC@C2	0.60	0.40	2.6	0.052	0.52
PC@C3	0.50	0.50	2.6	0.052	0.52
PC@C4	0.40	0.60	2.6	0.052	0.52
C@C5	0	1.00	2.6	0.052	0.52

*SP- Scots pine, CF- Coir fiber, and OPC- Ordinary Portland cement, Na_2SiO_3-Water glass, CS-Cement stone.

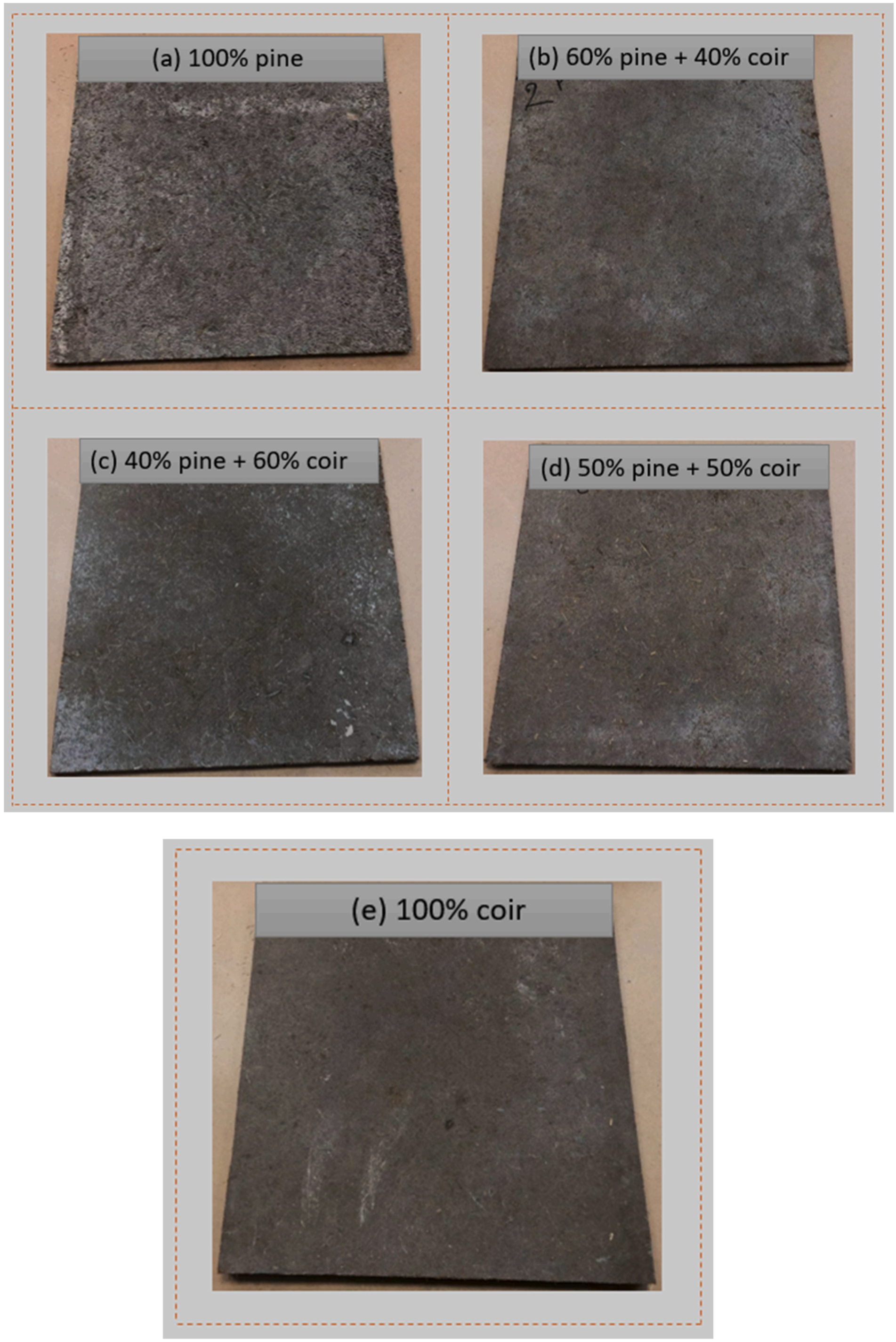

Fig. 3. Physical photographs of produced composite panels from coir fiber and Scots pines reinforced with OPC: (a) P@C1, (b) PC@2, (c) PC@3, (d) PC@4, and © C@5.

using **Equation (1)**.

$$\lambda = \frac{\phi.d}{(T_1 - T_2)A} \quad (1)$$

Where, ϕ indicates the heat flow which is the average electric power under steady state conditions, d is the composites thickness, and A is denoted for metering area. Thermal conductivity was measured through leaving the same temperature gradient, T_2-T_1 = 10 °C, where the tested conditions were T_2-T_1 = 10–20 °C. The density information of the panels are provided in and front view at Fig. 3. The protocols discussed above also goes in agreement with ISO 8302 standard [31].

The thermal conductivity was measured across the thickness of the composite panels with measuring the heat flow using a guarded hotplate method, where the difference between hot and cold side was 10 °C. A parallel heat flow was ensured to conduct this test by framing the composite samples with EPS insulation material of 15 cm width. The detailed testing protocol was discussed in some of the previous studies [29,32-34]. In brief, the composite panel trims were evenly cut. The panel surfaces needed to be flat and uniform to conduct this test. Once this was complete, the composites were placed in a standard atmospheric condition (65 ± 5% relative humidity and 20 ± 5 °C temperature) for 20 days to ensure that all the panels reached an equilibrium moisture level. It is necessary to secure uniform heat flow transfer; hence, the composites were surrounded by insulation boards (15 cm). The measurement readings were taken at steady rate and at least 100 data points were found to have less than 0.002 W/(m.K). The measurements were taken every minute and ultimate thermal conductivity was considered from last 100 measurement readings. The internal bonding strengths and flexural performances of the developed cementitious composites were characterized by Instron testing equipment (4208 model, United States) as per EN 319 and EN 310, respectively. The test specimens were prepared by cutting the samples with a circular saw (saw cut instruments, DCS570N XJ model, Pennsylvania, USA). The speed of crosshead movement for internal bonding strength was 0.8 mm/min and flexural properties were 5.0 mm/min. Furthermore, the SEM images were taken by SEM instruments (S 3400 N model manufactured by High Technologies Co., Ltd., Hitachi, Tokyo, Japan) at 15.0 kV under different magnifications. The EDX analysis was also performed by the same SEM machine. The FTIR analysis was conducted using FT/IR-6300 model instruments made by Jasco, Tokyo, Japan within a 4000 to 600 cm^{-1} wavenumber.

3. Results and discussion

3.1. Sugar and tannin contents

Sugar and tannin contents of both Scots pine and coir fibers were investigated before composite panel fabrications began in order to keep the presence of inhibiting materials in the lignocellulosic materials within an acceptable range. The perceived sugar content of Scots pine was less than 0.4% and tannin content was less than 0.5%. Conversely, the assessed values of coir fibers were found to be nearly 0.25% for tannin content and the sugar content was also found to be lower than 0.5%. However, the standard range of sugar content should be less than 0.5%, whereas the standards for tannin content should be less than 0.4% [11]. The current research skipped the pretreatment step to save on energy, associated chemicals, and costs as the sugar and tannin contents of both the materials are within the standard range.

3.2. Mechanical properties

Figure 4 shows the load-delamination curves in terms of internal bonding strength and flexural properties. The pulling out strengths for 100% coir/OPC composite was the highest compared to all other types, whereas 100% pine/OPC composites had the lowest value in this current study. However, the strengths started to increase with the increased coir fiber loading in the composite systems. It seems that coir fiber possesses higher bonding with the OPC matrix, which requires the highest loads for displacement compared to other composite panels. Furthermore, the increased values of load also demonstrate an effective and uniform transfer of stress throughout the composite materials. Moreover, a creation of hydrogen bonding between the fibers and OPC also facilitates the creation of a stronger interface in cementitious matrix systems. It is also worth mentioning that coir fibers developed stronger bonding with the OPC than the Scots pine materials did. This was also discussed by the scientists for different fiber-reinforced cementitious composites [35]. Accordingly, the highest value for loading, 1973.6 N, was observed in C@C5 composite panels and lowest, 1468.3 N, in P@C1 composites. The highest loads exposed by different composite panels are as follows: CP@C2, 1507.3; CP@C3, 1575.3; and CP@C4, 1676.54 N for the pulling out of the fiber materials from panels. Similar trends are also observed for flexural load–displacement curves. The highest load required to bend the specimen was in C@C5 samples (217.2 N) and the lowest load in P@C1 composites (81.2 N). Conversely, other composite panels also showed increasing trends from P@C1, which increases with the increase of coir fiber loading in the cementitious systems. Additionally, the stresses continued with the extended delaminations until total failure occurred.

The nominal densities of the composite panels were considered 1200 kg/m^3; however, actual panel densities were 1176.1 (35.76), 1021.03 (38.95), 1337.47 (65.64), 1118.4 (6.94), and 1150.17 (18.81) kg/m^3, respectively for P@C1, CP@C2, CP@C3, CP@C4, and C@C5 composites. The same operation protocol was maintained for all the composite panels, but the discrepancies may have been due to the manual mixing of materials, in-homogeneous mixing of materials to form the mat, the compression of mats, and the manual cutting of test specimens. On the other hand, the mechanical properties of the developed composites also displayed the same pattern with the load-delaminations curve. The flexural properties obtained for five composite panels were 6.22 (0.78), 6.77 (0.12), 6.78 (0.73), 7.97 (0.8), and 8.02 (0.87) MPa after 28 days of air curing in the laboratory. The flexural strengths of 100% Scots pine reinforced panel was the lowest, whereas the patterns increase with the increase of coir fiber contents in the OPC matrix system. Finally, 100% coir fiber-reinforced OPC composites exposed the highest values. A 29% increase was found in C@C5 panel compared to P@C1s. A similar trend was also observed for MOE results, with the exception of the CP@C2 composites. The reason behind the enhanced flexural properties could be the strong chemical and physical bonding [36] between the lignocellulosic materials and cement reagents (OPC). The incorporations of wood-based materials could play a significant role for the developed flexural properties in wood materials/cement composites [37].

The composite panels were also tested for internal bonding strengths (Table 4). P@C1 displayed 0.63 (0.08) MPa, whereas CP@C2 was 0.64

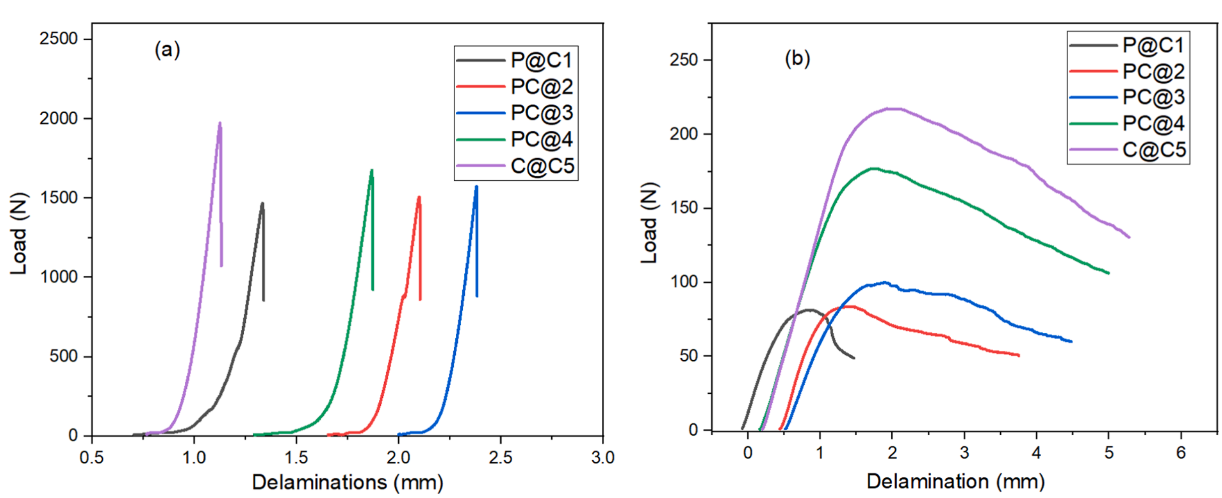

Fig. 4. Load versus displacement curves of composite panels produced from coir fiber and Scots pines reinforced with OPC: (a) IBS and (b) flexural properties.

Table 4
Mechanical characteristics of produced composite panels from coir fiber and Scots pines reinforced with OPC: (a) P@C1, (b) PC@2, (c) PC@3, (d) PC@4, and (e) C@5.

BCs	D (kg/m^3)	MOR (MPa)	MOE (GPa)	IBS (MPa)
P@C1	1176.1 (35.76)	6.22 (0.78)	6.78 (0.72)	0.63 (0.08)
CP@C2	1021.03 (38.95)	6.77 (0.12)	5.54 (0.74)	0.64 (0.03)
CP@C3	1337.47 (65.64)	6.78 (0.73)	7.62 (0.73)	0.66 (0.02)
CP@C4	1118.4 (6.94)	7.97 (0.8)	7.81 (0.42)	0.68 (0.03)
C@C5	1150.17 (18.81)	8.02 (0.87)	7.52 (0.87)	0.72 (0.06)

*BC-Biocomposites; D-Density; MOR-Modulus of rupture; MOE-Modulus of elasticity; IBS-Internal bonding strength.

(0.03), CP@C3 was 0.66 (0.02), CP@C4 was 0.68 (0.03) MPa, and C@C5 was 0.72 (0.06) MPa. Similarly, the properties show improved performances when coir fiber was induced in the composite systems. The most interesting finding is that all the panels show values higher than 0.5 MPa, which is the standard bottom line for internal bonding strengths [13]. Nevertheless, Ghofrani et al. notes that the standard of internal bonding strengths as per ISO (the International Organization for Standardizations) system is 0.45 MPa [38]. In this regard, the produced biocomposite panels could be implemented as prominent materials in the construction and building sector. Moreover, the 100% coir fiber-reinforced composite provided values that were roughly 14% higher than 100% Scots pine reinforced cementitious materials. Although density is a significant parameter for determining the mechanical performances of composite panels [39], the incorporation of different lignocellulosic materials played a key role in determining the ultimate performances of the composite materials in this current study.

3.3. Morphological observation

In addition to the thermomechanical and physical properties of cementitious materials, morphological photographs were also investigated further. Fig. 5 contains the SEM pictures of the composite panels where the interface zones of coir fiber and Scots pines bonded OPCs are explicitly noticeable. Two phases could be observed in the composite systems: (a) a continuous fiber like layer (attributed to lignocellulosic materials) and (b) aggregated particle-like appearances (corresponding to OPC) [40]. Similar fiber-like appearances also appeared in the developed composites; see Fig. 5(a_1 and a_2). Furthermore, other images also displayed both the fiber and particle like aggregates in the composite systems. The appearances of lignocellulosic materials appeared more in P@C1 and PC@C2 than in other composites. This could be because the bigger Scots pine particle contents were higher (100 and 60%) in these panels than they were in the other three types of panels. Conversely, the coir fibers were used here in fiber forms, which is why they were in closer contact with the OPC (due to smaller fiber-like materials); hence, they nearly disappeared in the strongest bonding in the composite systems. This phenomenon also illustrates that lignocellulosic materials are compatible with OPCs. The strongest bonding could also be proved by the increased mechanical properties of the composite panels with higher loadings of coir fibers. However, the presence of lignocellulosic materials could be clearly seen in the fractured surfaces of composites, although OPC was strongly adhered in the surfaces of lignocellulosic materials (Fig. 6). That is why more load is required to break the composites. This is also reflected in load versus delamination curves (Fig. 4) and internal bonding strengths (Table 4). In addition, the interaction between the lignocellulosic materials and the OPC matrix plays a significant role for the initiation and determinations of crack propagations [41]. A similar phenomenon was also observed in our previous study in which the lignocellulosic materials derived from seven Hungarian plants were bonded with OPC [9,42].

3.4. EDX analysis

The detected chemical elements in the cementitious composites could be easily discerned by differently colored scaling of EDX spectra. The EDX analysis of the cementitious composite exhibits the distribution of Ca and Si in different fragments that are the principal chemical constituents of OPC. The presence of Si and Ca signals were also detected by Wei et al. [43] for wood/cement composite panels at their interface. Furthermore, the significant presence of some other chemical elements like as Na, Mg, Al, Ti, K, and Fe, also reflects a successful bonding of OPC with the lignocellulosic Scots pine and coir fiber materials. The presence of these materials also corresponds with the constituent materials of OPC as shown previously in Table 2. The presence of C and O at different fragments in Scots pine and coir fiber control (Figure 7) was also notable, even after the bonding with OPC. Furthermore, the bonding of lignocellulosic materials with OPC could also be further confirmed by the FTIR analysis. Interestingly, the presence of C is higher in Scots pine (55.25%) than in coir fiber (52.41), which also applies to the associated composite panels at 100% Scots pine-reinforced OPC composite. In addition, this shows that the presence of C that is higher than it was in the 100% coir fiber-reinforced cementitious materials. Alternatively, a similar effect was noticeable in O, which was higher in coir fibers and associated composites. Moreover, the coir fiber and Scots pine controls displayed the dominance of C and O in their polymeric structure, but the phenomenon changed after the bonding with OPC for all the five composite panels, where the dominance of Si and Ca is also apparent due to the strong influence of OPC in the composite systems. The presence of cementitious chemical agents in the composite systems also accords with the reports of other scientists [40,44,45].

3.5. Thermal conductivity investigation

Thermal conductivity values of different cementitious composite panels (Fig. 8) were also investigated to assess their insulation performances. The thermal conductivity of air is 0.026 W/(m.K), water is 0.6 W/(m.K), and the cement stones is 1 to 3 W/(m.K) [51]. Natural plant fibers contain a thermal conductivity of around 0.2 W/(m.K) [36], whereas coir fiber possesses 0.58 W/(m.K) in case of 85 kg/m^3 fiber density at 39 °C [52], and Scots pine 0.132 (0.008) W/(m.K) in case of 550 kg/m^3 material density [53]. A study was also conducted in our University by another research group to investigate the thermal conductivity of coir fiber materials through following the same machine and protocol and found the results 0.057 W/(m.K) [54] which also goes in agreement with the other typical thermal conductivity values reported by different research team 0.58 W/(m.K) [52], which is also validating our current testing setup and operational protocols too. Similarly, by using the same testing equipment for some other wood/fiber based cellulosic material derived materials/products, the results also shown to meet the accuracy of test. Like as, thermally treated kiri wood was providing thermal conductivity values by 0.064 W/(m.K) at 220 °C [34], bark-based insulation panels providing 0.067 to 0.070 W/(m.K) [55], glass fiber reinforced bark particleboards 0.059 (±0.002) to 0.079 (±0.003) W/(m.K) [56], wood shaving made insulation panel 0.05 to 0.06 W/(m.K) [57], coir material reinforced MUF composite 0.9302 ± 0.00999 to 0.1078 ± 0.0072 W/(m.K) [29], and so on. The overall reports also clarifying the accuracy of our testing setup for thermal conductivity. The distribution and volumes of pores in the cementitious composite also affect thermal conductivity. As the thermal conductivity of water and cement is also excessively higher than air, they also significantly influence the thermal conductivity of lignocellulosic materials bonded with OPC composite panels. However, thermal conductivity values obtained in our current study (average of 100 readings) exhibited values of 0.17045 (0.00352) W/(m.K) for P@C1, whereas CP@C2 was 0.16651 (0.00265), CP@C3 was 0.11646 (0.00263), CP@C4 was 0.11265 (0.00409), and C@C5 was 0.10547 (0.00657) W/(m.K). Notably, when the coir fiber was induced in composite systems,

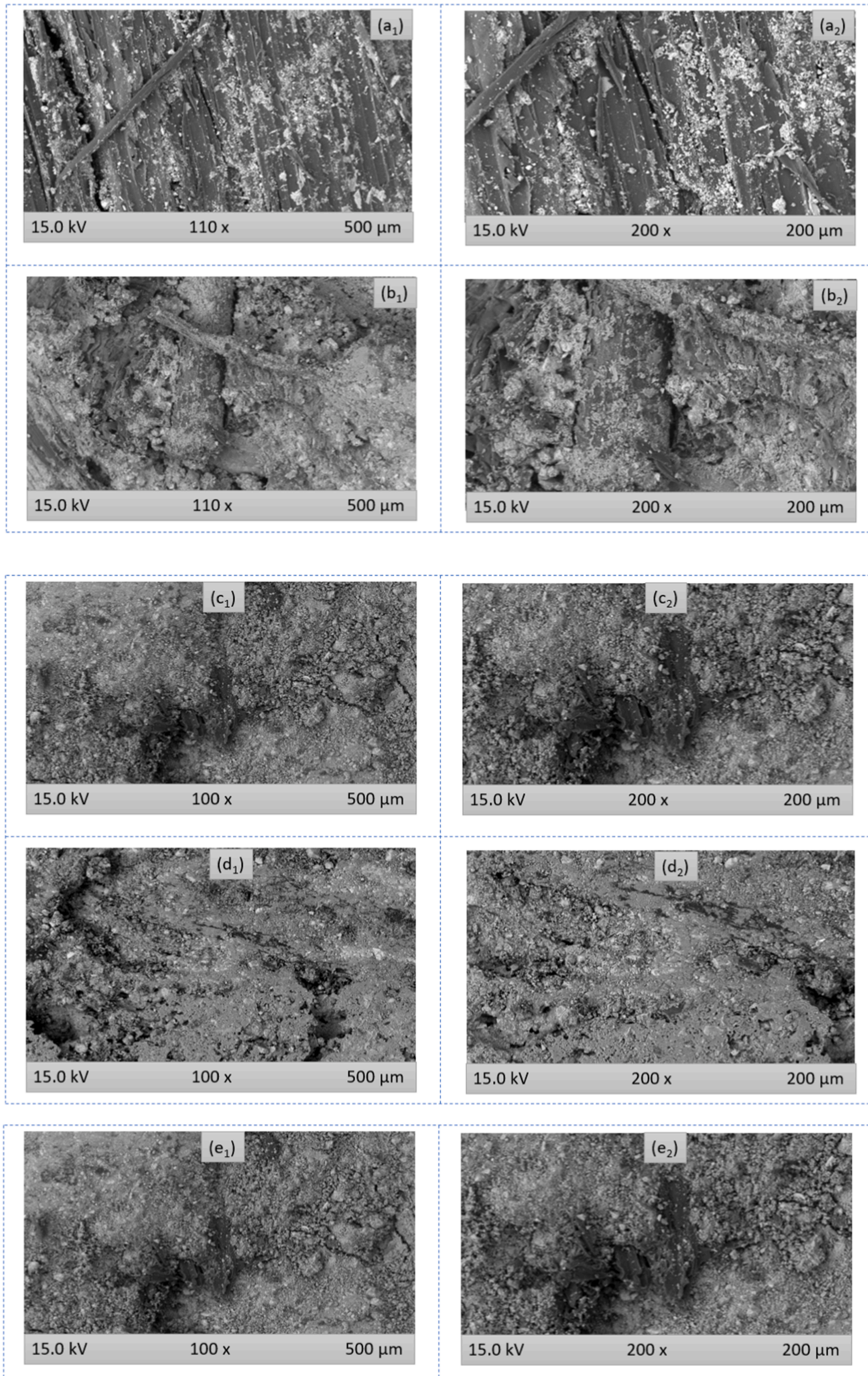

Fig. 5. SEM micrographs of specimens (before fracture) of produced composite panels from coir fiber and Scots pines reinforced with OPC: P@C1 (a_1 and a_2), PC@C2 (b_1 and b_2), PC@C3 (c_1 and c_2), PC@C4 (d_1 and d_2), and PC@5 (e_1 and e_2).

the thermal conductivity values started to decline. That is why our study reveals the 100% coir fiber-reinforced cementitious composite to be the best insulation material, whereas 100% Scots pine displayed the lowest thermal conductivity values according to the performance perspectives. Another highly impressive finding of this research is that the cementitious composite panels also provided competitive thermal conductivity with MUF polymeric composites with coir fiber and fibrous chips, which in our previous study ranged within 0.09302 (0.00999) to 0.1078 (0.0072) W/(m.K) [29]. In another study by He et al. [36], the lowest thermal conductivity value was obtained for wood/magnesium-oxychloride cement composite by 0.97 W/(m.K), whereas the results explained in this current study are within 0.17045 (0.00352) to 0.10547 (0.00657) W/(m.K), demonstrating better insulation capability of the panels. In addition, we also compared our obtained results for thermal conductivity with other studies for different natural fiber reinforced composites in terms of various standards (ISO 10456, ISO 8032, and JIS

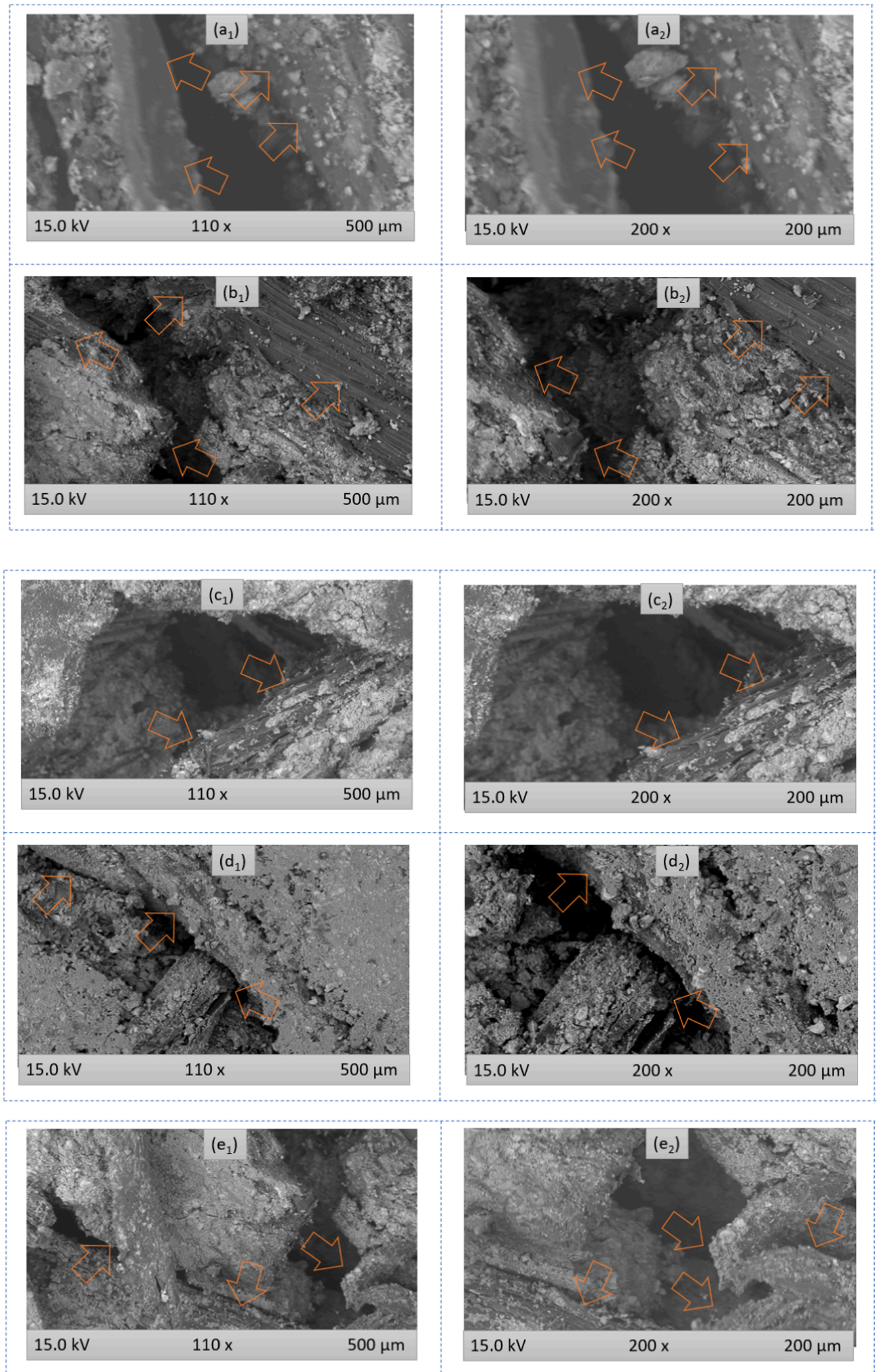

Fig. 6. SEM micrographs of specimens (after fracture) of produced composite panels from coir fiber and Scots pines reinforced with OPC: P@C1 (a_1 and a_2), PC@C2 (b_1 and b_2), PC@C3 (c_1 and c_2), and PC@C4 (d_1 and d_2), and PC@C5 (e_1 and e_2).

R 2618–1992) (Table 5) and found the perceived values did not make any extensive changes which also confirm about the validity of our current research.

3.6. FTIR analysis

The FTIR analysis of the developed composite panels from coir fiber and Scots pines reinforced with OPC matrix is shown within 4000 to 600 cm^{-1} wavelengths (Fig. 9). The peaks within 3200 to 3400 cm^{-1} represents the stretching vibrations of O–H and C–H bands of hemicellulose [58,59]. The peaks around 1000 cm^{-1} is associated with the stretching vibrations of Si-O [59] and C-O bands (cellulose and hemicellulose). The spectrum within 873 to 1418 cm^{-1} are related with the carbonations of different hydrates in the composite systems [59,60]. Furthermore, the absorption band at 1418 cm^{-1} is attributed to the CH bending of aromatic rings of lignin polymers [61], which is broader with coir-based

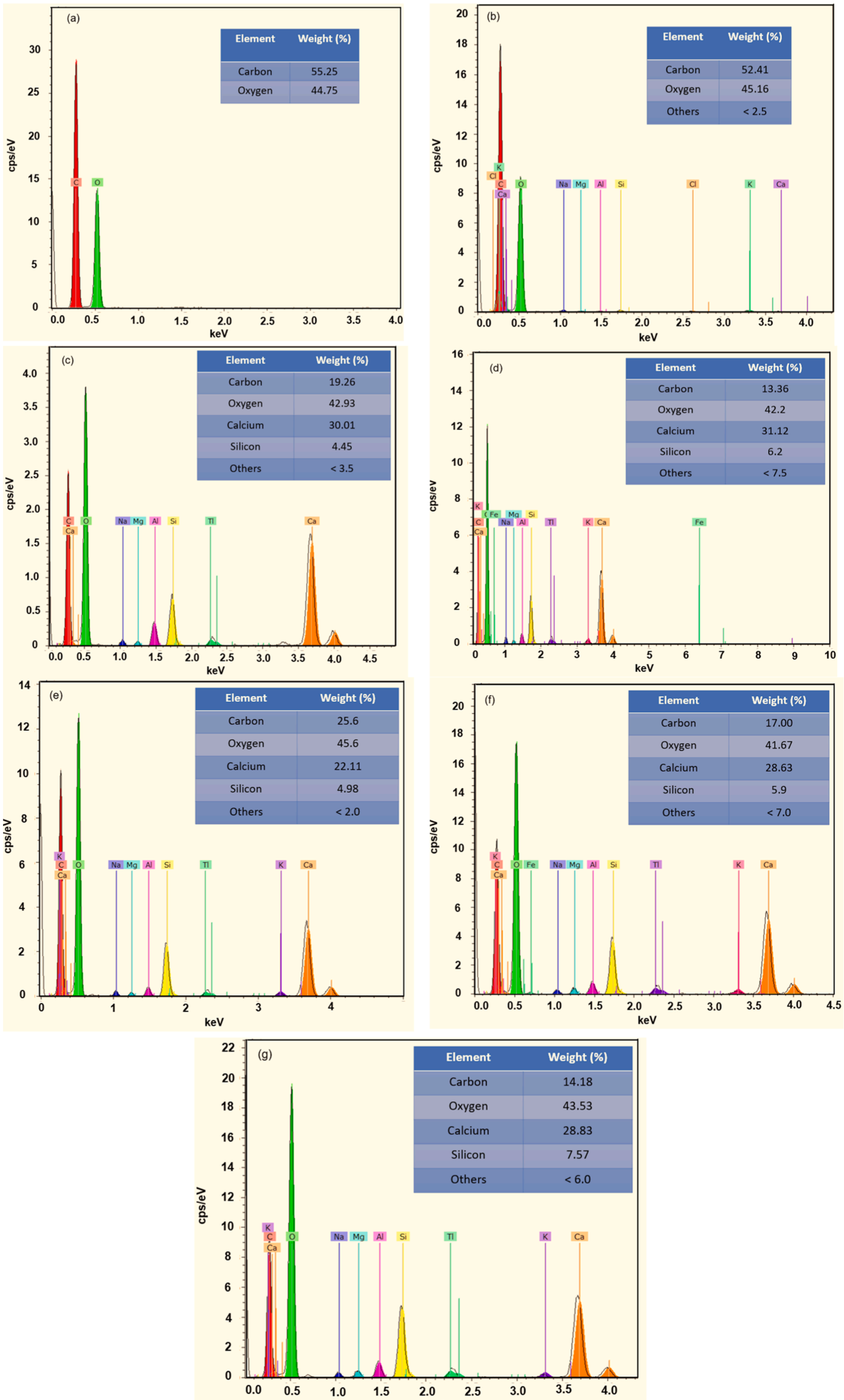

Fig. 7. EDX spectrum of produced composite panels from coir fiber and Scots pines reinforced with OPC: (a) 100% Scots pine, (b) 100% coir fiber, (c) P@C1, (d) PC@2, © PC@3, (f) PC@4, and (g) C@5.

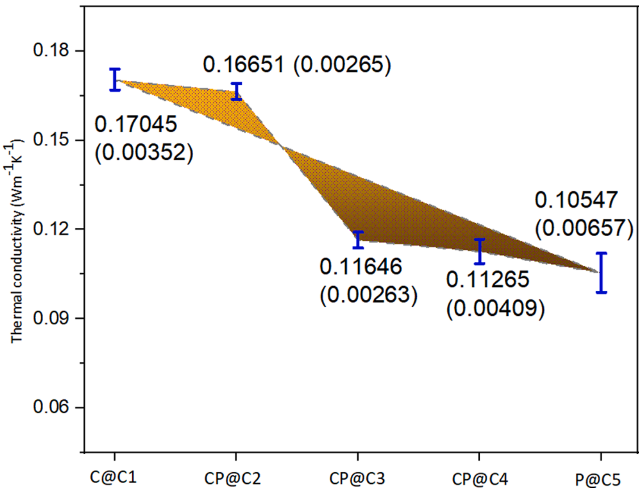

Fig. 8. Thermal conductivity of coir fiber and Scots pines reinforced with OPC composites.

Table 5
Comparative thermal conductivity studies for different natural fiber reinforced composites.

Composite description	Thermal conductivity W/(m.K)	Density (kg/m^3)	Standard adopted	References
Scots pine/coir reinforced OPC composite	0.17045 (0.00352) to 0.10547 (0.00657)	1021.03 (38.95) to 1337.47 (65.64)	ISO 10456	Current study
3% plant aggregates reinforced composite	0.35 ± 0.02	1671 ± 21	ISO 8302	[46]
6% plant aggregates reinforced composite	0.2 ± 0.01	1271 ± 16	ISO 8302	[46]
Rice husk/OPC (type II)	0.1226 (±0.0157)	1091.2 (±91.5)	ISO 8302	[47]
Rice husk/OPC (type II)	0.3554 (±0.0210)	1311.4 (±14.6)	ISO 8302	[47]
Mycelium and Miscanthus composites	0.0882 to 0.121	122	ISO 8302	[48]
Coir fiber/OPC lightweight composite	0.2518 ± 0.009	1125 ± 18.71	JIS R 2618-1992	[49]
Hemp fiber reinforced composite	0.139	1080	ISO 10456	[50]

materials as coir possesses higher lignin contents when compared to Scots pine. Moreover, the peaks at 573 cm^{-1} is attributed to the cementitious matrix that is absent from the coir fiber and Scots pine controls, but is present in the cementitious materials [12]. The overall discussions demonstrate a successful bonding of coir fiber and Scots pines with the OPC matrix, which adds better thermal and mechanical properties to the composite panels.

3.7. Physical properties investigation

The physical effects of different lignocellulosic materials on cementitious materials are shown in Fig. 10 in order to investigate the dimensional stabilities for 2 h and 24 h periods. It is evident that the incorporation of coir fiber leads to decreased moisture content as well as decreased water uptake. This is because fiber materials could achieve closer contact with the cements, thereby reducing the possibility of void creations. This, in turn, led to less water absorption [36] than in the porous Scots pine particle reinforcements. Moreover, plant materials are hydrophilic in nature due to the presence of –OH groups in their polymeric structures; hence, a hydrogen bond is created when they come in contact with water. This phenomenon is also responsible for moisture contents and water absorbency as well as thickness swelling (Fig. 10 b). Coir fiber also possesses high resistance against moisture, which helps it to withstand water [62]. As in the case with other thermal and mechanical properties, 100% coir fiber-reinforced cementitious materials also exhibited better stability against water compared to 100% Scots pine reinforced cementitious composite panels. The moisture content for composite 1 was 5.3 (0.4)% and 3.9 (0.9)% for composite 5. The sequence of moisture contents, water absorbency, and thickness swelling in terms of better stability is as follows: C@C5 > PC@C4 > PC@C3 > PC@C2 > P@C1. A similar trend was also observed for water absorption, where the values varied from 19.1 (0.9)% for composite panel 1 to 14.6 (0.9)% for composite panel 5 after 2 h of water immersion. After 24 h of immersion, the values increased slightly by 30.3 (0.4)% for composite 1 and 25.1 (0.3)% for composite 5. The other panels showed water absorption values between composite panel 1 and 5 as they are made of different proportions of Scots pine and coir. Composite panel 1 displayed a thickness swelling value of 1.9 (0.02) and composite 5 displayed a value of 1.1 (0.07)% after 2 h. After 24 h, the values increased slightly to 2.6 (0.06) and 1.9 (0.06)%, respectively. The other composites demonstrated thickness swelling values that ranged between composite 1 and 5. A similar phenomenon of physical properties was also reported by other researchers for lignocellulosic fiber-reinforced cementitious composites [38,41,63]. However, the developed composite panels could also be used for indoor applications due to their improved physical properties.

4. Conclusions

Interest in green, environmentally friendly building products is increasing around the world. In this regard, naturally originated lignocellulosic materials are revealing new sustainable building material routes for the construction and building sector. The developed composites with variable densities were 1176.1 (35.76), 1021.03 (38.95), 1337.47 (65.64), 1118.4 (6.94), and 1150.17 (18.81) kg/m^3 for composite s 1, 2, 3, 4, and 5, respectively. These composites were made from lignocellulosic materials (coir fiber and Scots pine particle) bonded with OPC, and they provided satisfactory performances (mechanical, thermal, and physical). The thermo-mechanical properties of Scots pine improved with the loading of coir fibers in the composite systems. All the composite materials provided internal bonding strengths higher than 0.5 MPa, indicating strong materials for structural applications. The SEM analysis showed the explicit presence of coir fiber and Scots pine in the fractured composite panels, whereas the presence of lignocellulosic materials were further justified in terms of FTIR analysis. The EDX spectra of the composites also provides successful bonding of coir fiber and Scots pine with OPC in terms of chemical elements presence. The thermal conductivity values of the composites were 0.17045 (0.00352) W/(m.K) for P@C1, 0.16651 (0.00265) for CP@C2, 0.11646 (0.00263) for CP@C3, 0.11265 (0.00409) for CP@C4, and 0.10547 (0.00657) W/m.K) for C@C5, which provides explicit proof of the developed panels as potential insulation materials. Overall, the current research is going to explore and open new routes toward producing green and sustainable products in building and construction sector.

CRediT authorship contribution statement

K.M. Faridul Hasan: Conceptualization, Methodology, Investigation, Data curation. **Péter György Horváth:** Conceptualization, Supervision. **Zsófia Kóczán:** Investigation, Data curation. **Miklós Bak:** Investigation, Data curation. **Tibor Alpár:** Conceptualization, Data curation, Investigation, Supervision.

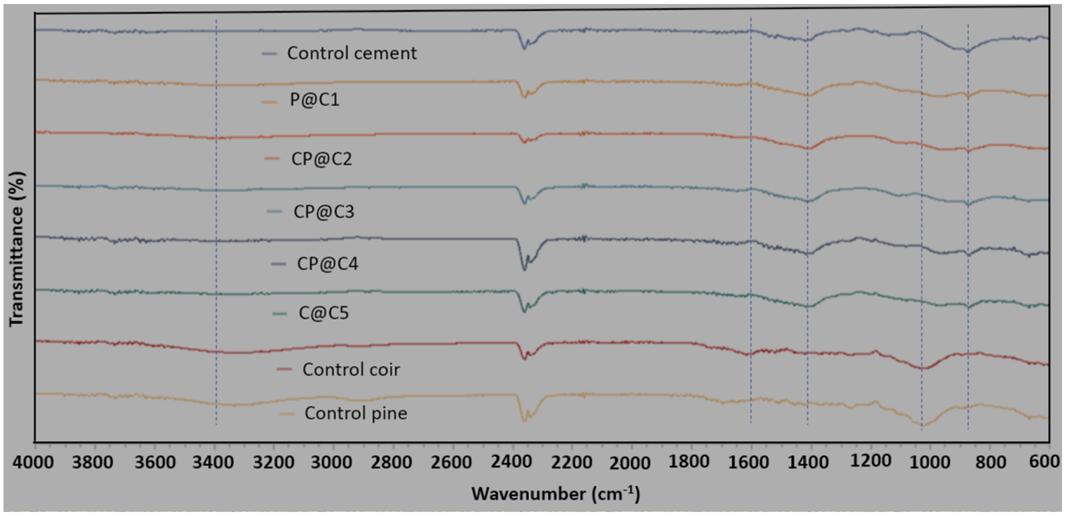

Fig. 9. FTIR analysis of produced control lignocellulosic materials and associated composite panels from reinforced with OPC.

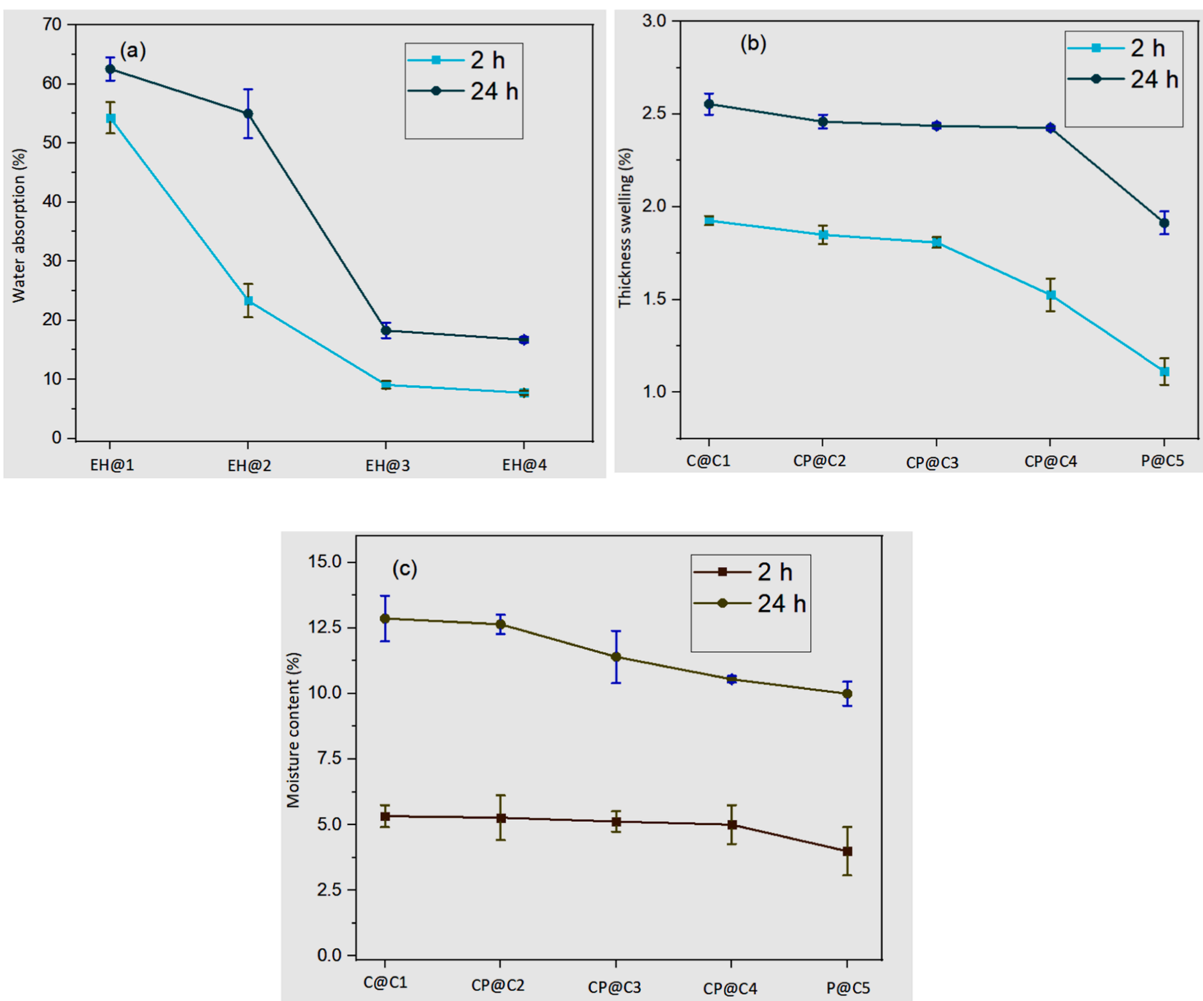

Fig. 10. Physical properties of produced composite panels from coir fiber and Scots pines reinforced with OPC: (a) water absorbency, (b) thickness swelling, and (c) moisture content.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.conbuildmat.2021.124816.

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