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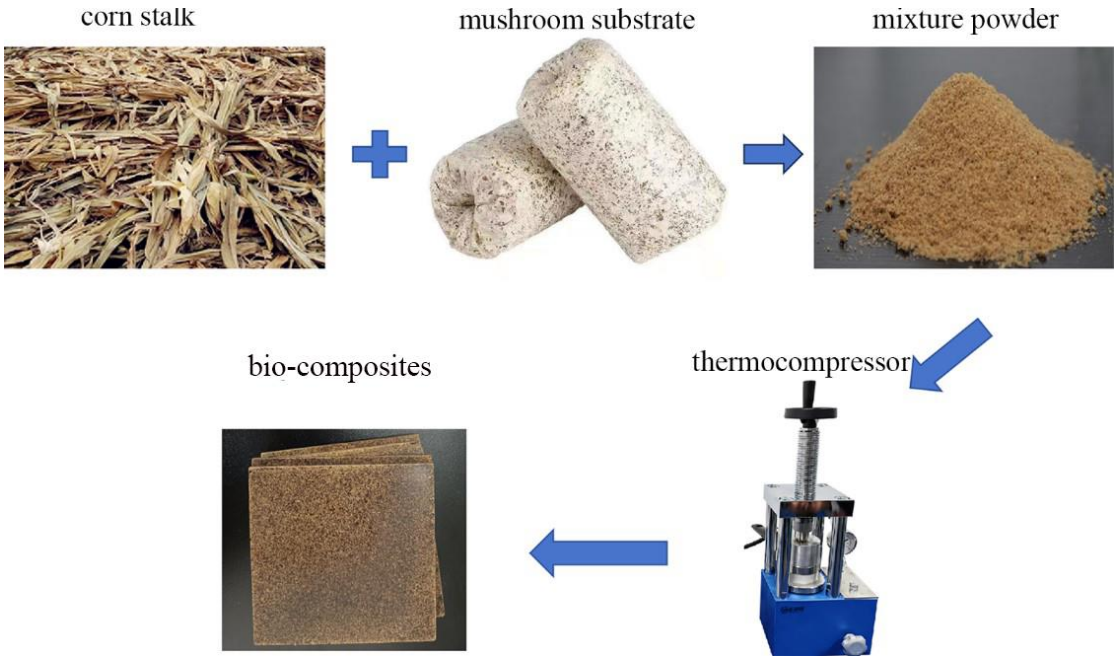
GRAPHICAL ABSTRACT



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Corn straw-based board has great potential for the protection of forest resources, waste recycling, and sustainable economic development. However, corn stalk-based board has poor mechanical properties due to its short fiber length and poor water resistance because of the presence of numerous hydrophilic hydroxyl functional groups in its structure. Natural mycelium originating from waste *Pleurotus ostreatus* substrate is a hydrophobic bio-adhesive. In the present study, formaldehyde-free corn stalk/*P. ostreatus* substrate bio-composites were prepared using the hot-pressing technique without the addition of any chemical adhesive. The mechanical properties and water resistance of the prepared bio-composites were excellent. The highest internal bonding strength (IBS) of 2.16 MPa and the minimum thickness swelling (TS) of 18.3% were observed, which are beyond the national standards for particleboard in China. These bio-composites were prepared using a simple, green, and convenient manufacturing method to promote their popularization and application. The method may, therefore, be used as a novel technical measure to resolve the problem of overuse of forestry resources and waste disposal.

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Keywords: Bio-composites; Corn stalk; Pleurotus ostreatus substrate; Mechanical property; Water resistance

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INTRODUCTION

Corn resources are abundant in China, and the annual yield of agricultural waste corn straw is huge. The geography, climate, economy, science, and technology of the nation lead to most of this corn straw being burned, which causes serious environmental pollution and damage to human health. Therefore, converting this corn straw waste into a value-added resource is a key concern that requires a prompt solution.

According to the 8th National Forest Census of China, while the forest coverage rate in China is as high as 21.66%, the per capita forest area is just 1/7th of the world average. On the other hand, the demand for wood products has increased as the quality of life for people has improved. This gap between the supply and demand of wood resources in the market has to be resolved urgently.

Corn straw, similar to wood, is composed of cellulose, hemicellulose, and lignin mainly (Wang *et al.* 2019). Therefore, it can be considered as a candidate for replacement of wood in the production of artificial boards. Hot-pressing is currently one of the main techniques used for the preparation of artificial boards (Ling *et al.* 2012; Gul *et al.* 2019; Minhas *et al.* 2021). Most corn straw-based bio-boards are prepared through the wet process, which increases the mechanical strength of the bio-board. This is achieved through the increased exposure of active hydroxyl groups *via* the fibrillation of cellulose after the pretreatment of corn straw in water. These exposed hydroxyl groups then combine with water molecules to form hydrogen bonds in corn straw (Kargarfard and Jahan-Latibari 2011). However, the wet process is not environmentally friendly, which seriously limits its industrial manufacturing. In addition, synthetic adhesives have to be essentially added during the preparation of corn straw-based bio-board to increase its mechanical strength (Akgül *et al.* 2010; Kargarfard and Jahan-Latibari 2011; Akinyemi *et al.* 2016; Gűler *et al.* 2016), and the most widely used synthetic adhesives are volatile organic compounds that release formaldehyde into the environment gradually. Chronic exposure to formaldehyde gas is harmful to humans (Demircioglu *et al.* 2011). Moreover, the water resistance of corn stalk-based bio-board is poor due to the presence of numerous hydrophilic hydroxyl functional groups in cell walls, which is considered a drawback when fabricating bio-boards (Kargarfard and Jahan-Latibari 2011; Ye *et al.* 2018). Therefore, preparing environment-friendly bio-boards with high mechanical strength and higher water resistance remains a key concern in this industry.

A mycelium-coated mushroom substrate may be added as a green adhesive during the preparation of an artificial board to enhance its mechanical properties. The solid mycelium is changed during hot-pressing and enters the mushroom substrate, where it forms a tight network of adhesive material, thereby strengthening the mechanical properties of the resulting bio-board (Elsacker *et al.* 2011). According to Jafari *et al.* (2007), mushroom substrate-based bio-board exhibits excellent water resistance due to the rigid impermeable layer of thick mycelium spreading over the mushroom substrate (Khoo *et al.* 2020).

In the present study, corn stalk and *P. ostreatus* substrate were used as raw materials to prepare formaldehyde-free corn stalk/*P. ostreatus* substrate bio-composites using the hot-pressing method and the dry process. The effect of temperature, the mass fraction of raw materials (corn stalk, *P. ostreatus* substrate, and deionized water), the hot-press time and cooling time on the mechanical properties, water resistance, and thermal stability of the prepared bio-composites were investigated. In addition, FT-IR spectroscopy and X-ray diffraction measurements were performed to explore the self-bonding mechanism of these bio-composites. The study provided an effective solution for poor forest resources and waste disposal and reuse.

EXPERIMENTS

Materials

Corn stalk (Suyu 28) and *P. ostreatus* substrate were collected from the rural regions around Changchun, China. The stems and leaves of corn stalk and the *P. ostreatus* substrate were crushed to the mesh level of 20–60 and 20–100, respectively. The resulting powders of corn stalk and *P. ostreatus* substrate were dried in an oven at 100 °C until a constant weight was reached.

Preparation of Bio-composites

Approximately 70 g of the *P. ostreatus* substrate, corn stalk, and deionized water were mixed evenly and then sealed into a plastic bag to leave undisturbed at room temperature for 24 h. Next, the powder mixture was placed inside the mold and pressed using a thermo-compressor (Xinnuo Machinery Equipment Co. Ltd) with a pressure of 18 MPa at 190 °C for 2 h to prepare bio-composites with the dimensions of 100×100×5.0 mm. Afterward, the mold was allowed to cool for 3 h at room temperature, followed by removing the prepared bio-composites from the mold. Note that the conditions of temperature and time were intentionally more severe in comparison to present industrial practices for this kind of product. This difference is because no synthetic adhesive, such as a formaldehyde-based resin system, was used in this work.

Table 1. Orthogonal Experimental Design for the Preparation of Bio-composites

Sample	Mass ratio of raw materials (*P. ostreatus* substrate: corn stalk: deionized water)	Hot-press temperature (°C)	Hot-press time (h)	Cooling time (h)
f1	9:0:1	190	2	3
f2	8:1:1	190	2	3
f3	7:2:1	190	2	3
f4	6:3:1	190	2	3
f5	5:4:1	190	2	3
f6	4:5:1	190	2	3
f7	3:6:1	190	2	3
f8	2:7:1	190	2	3
f9	1:8:1	190	2	3
f10	0:9:1	190	2	3
f11	8:0:2	190	2	3
f12	7:1:2	190	2	3
f13	6:2:2	190	2	3
f14	5:3:2	190	2	3
f15	4:4:2	190	2	3
f16	3:5:2	190	2	3
f17	2:6:2	190	2	3
f18	1:7:2	190	2	3
f19	0:8:2	190	2	3
f20	3:6:1	180	2	3
f21	3:6:1	170	2	3
f22	3:6:1	160	2	3
f23	3:6:1	150	2	3
f24	3:6:1	140	2	3
f25	3:6:1	130	2	3
f26	3:6:1	120	2	3
f27	3:6:1	110	2	3
f28	3:6:1	100	2	3
f29	3:6:1	90	2	3
f30	3:6:1	80	2	3
f31	3:6:1	70	2	3
f32	3:6:1	190	1.5	3
f33	3:6:1	190	2	2.5

The bio-composites were evaluated for the effects of water content, mass ratio of raw materials, hot-press temperature, hot-press time and cooling time on their mechanical and physical properties. The study was designed as an orthogonal experiment (refer to Table 1).

Characterization of the Prepared Bio-composites

Evaluation of mechanical properties

The IBS, the modulus of elasticity (MOE), and the modulus of rupture (MOR) of the prepared bio-composites were measured using a computer-controlled electronic universal tensile machine (Model WDW-1, Jilin Guanteng Automation Technology Co. Ltd., China) and compared to the China National Standard GB/T 17657-2022. In order to measure the IBS of bio-composites, board samples of size of 50×50×5 mm (thickness) were attached to a loading block using hot-melt glue. After curing, the block was set into the grip and the IBS test was conducted using a crosshead speed of 2 mm/min. The MOE and MOR of the prepared bio-composites were measured using the three-point bending test. In this test, samples of size 100 mm × 25 mm × 5 mm were placed on the support shaft, and the span between the two supports was adjusted to 50 mm. A central loading roller was then utilized to apply pressure to the specimen at a speed of 5 mm/min. Ten samples were tested for each group, and standard deviations were calculated.

Microscopic observation

The internal and surface structures of the prepared bio-composites were examined using a scanning electron microscope (SEM; JFC-1600, JEOL Ltd., Tokyo, Japan). First, a thin layer of gold was sprayed on the surface of the bio-composite specimen using an auto-fine coater (JFC-1600, JEOL Ltd., Tokyo, Japan), followed by examining the gold-coated specimens under SEM and obtaining the desired morphological images.

Water immersion test

A water immersion test was conducted to investigate the water resistance properties of the prepared bio-composites. The thickness of the bio-composites prior to and after water immersion was measured for different cumulative durations of 2, 4, 6, 8, 12, 24, and 48 h. The measurements were performed using a digital caliper gauze (Model AY0050152, Shanggong, China). The thickness of swelling rate (TSR) value was calculated using the following equation,

$$TSR = (T_f - T_i)/T_i \times 100\% \quad (1)$$

where T_f denotes the final value of thickness after water immersion and T_i denotes the initial thickness prior to water immersion. Ten samples were tested for each group, and standard deviations were calculated.

Thermogravimetric analysis (TGA)

The thermal stability, stage of decomposition, and the kinetics reaction of the prepared bio-composites f3-f7 were determined through TGA using a Thermal Gravimetric Analyzer (Discovery TGA 550, TA Instruments, USA). Nitrogen gas was used as the carrier gas in this analysis. Approximately 20 mg of the bio-composite powder was placed inside the TGA analyzer, and the gas flow rate was set to 25 mL/min. The heating rate was set to 10 °C/min. The temperature applied to the samples in the TGA analysis ranged from 20 to 900 °C.

Fourier transform infrared spectroscopy (FTIR) analysis

Small portions of the bio-composites to be evaluated were ground to a powder form and subjected to the FTIR analysis using the PerkinElmer Spectrum Two FT-IR spectrometer.

X-ray diffraction analysis

X-ray diffraction was studied to examine the crystallinity of the bio-composites. The Rigaku Smart Lab X-ray diffractometer (Smartlab SE, Rigaku Company, Japan) with Cu Ka radiation (λ = 1.5406 Å) was used for this analysis. The crystallinity of the bio-composites was calculated using the formula Cr = (A_c/A_a × 100%), where Cr denotes the relative percentage of crystallinity and A_c and A_a denote the area of crystalline peaks and the area of all peaks (including both crystalline and amorphous regions), respectively.

Statistical analysis

The credibility of all experimental data (IBS, MOR, MOE, TSR, and MC) was determined to identify the most appropriate process parameters for the prepared bio-composites. Analysis of Variance (ANOVA) was conducted using the statistical package for social sciences (SPSS) software, and p ⩽ 5% was used as the significance threshold. Ten duplicates were used for each condition.

RESULTS AND DISCUSSION

Internal Bonding Strength

The IBS results for the corn stalk/*P. ostreatus* substrate bio-composites with different ratios of corn stalk, *P. ostreatus* substrate, and deionized water are presented in Fig. 1. All IBS data were compared with the GB/T 4897 (2015) standard. As presented in Fig. 1a and 1b, the IBS of bio-composites with a moisture content of 10% of all raw materials (corn stalk, *P. ostreatus* substrate, and deionized water) was significantly better than that of the bio-composite with a moisture content of 20% of the raw materials. Therefore, a certain moisture content enhanced the strength of bio-composites. This is attributed to the partial degradation of hemicellulose in corn stalk and the *P. ostreatus* substrate under the synergistic effect of water and high temperature, which leads to the increase in the number of hydroxyl groups in the cellulose microfibrils. Hydrogen bonds are then formed among these hydroxyl groups, improving the mechanical properties of the bio-composites. However, excessive moisture results in a powerful steam pressure during hot-pressing, which destroys the polymer bonding that has already formed and leads to the delamination of bio-composites after hot-pressing.

As depicted in Fig. 1a and 1b, the mass ratio of raw materials exerted a significant impact on the mechanical strength of bio-composites, and the IBS of the bio-composites f3, f4, f5, f6, and f7 surpassed the IBS standard requirement for the loading-bearing particleboard used in a dry condition in China (GB/T 4897-2015, IBS, 0.45 MPa). In comparison to corn stalk-based bio-board and *P. ostreatus* substrate-based bio-board, the bio-board with a mixture of corn stalk and *P. ostreatus* substrate exhibited improved mechanical properties. Several loose and porous structures were visible inside corn stalk under ambient conditions (Fig. 2a). The *P. ostreatus* substrate was composed of several non-compact net structures (Fig. 2b). After hot-pressing, the *P. ostreatus* substrate filled the space of corn stalk and acted as an adhesive that bound the corn stalk together (Fig.

2c), thereby improving the mechanical properties of the composite.

The MOR and MOE of bio-board prepared with different mass ratios of corn stalk, *P. ostreatus* substrate, and 10% deionized water are presented in Fig. 1c. Similar to the IBS results, the MOR and MOE values of the bio-composites f3 to f7 met the national standards for the loading-bearing particleboard established in China (GB/T 4897-2015, MOR: 15 MPa, MOE: 2200 MPa). This further confirmed that the addition of *P. ostreatus* substrate to corn stalk effectively improved the mechanical properties of the prepared bio-board.

Subsequently, bio-composites with the f7 mass ratio of raw materials were prepared at different temperatures (70 to 190 °C) to study the effect of temperature on the mechanical strength of the boards. The results are presented in Fig. 1d. As the temperature increased from 70 to 180 °C, the IBS of the bio-composites varied within the range of 0.01 to 0.2 MPa. The IBS of these bio-composites reached its maximum value of 1.2 MPa at the hot-pressing temperature of 190 °C. This was attributable to the depolymerization degree of lignin (Widsten and Kandelbauer 2008) and hemicellulose (Su *et al.* 2022), which increases with the increase in the hot-pressing temperature, providing greater opportunities for the cross-linking of the depolymerization products of lignocellulose. At a high temperature, condensation reactions occur between the liberated xylose from hemicellulose and the phenoxy radicals from lignin (Le *et al.* 2016). In addition, self-crosslinking may occur among the phenoxy radicals (Yang *et al.* 2023). These higher opportunities for depolymerization and cross-linking at 190 °C could have contributed to the abrupt increase in the IBS of the bio-composites.

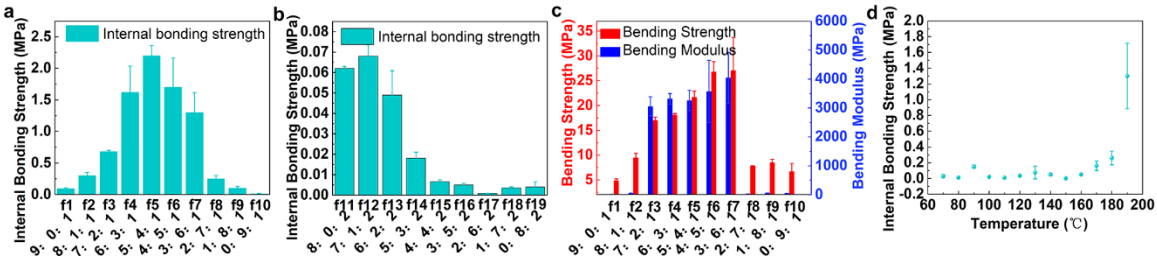

Fig. 1. IBS of the bio-board prepared with different mass ratios of raw materials: corn stalk, *P. ostreatus* substrate, and deionized water (a) 10% (b) 20%. The mass percentages of all raw materials (*P. ostreatus* substrate: corn stalk: deionized water) used for preparing the bio-board are listed under the bio-board number. (c) The MOR and MOE of the bio-boards prepared with different mass ratios of raw materials (corn stalk, *P. ostreatus* substrate, and 10% deionized water). The hot-pressing temperature-dependent (d) IBS of the bio-composites prepared with the f7 mass ratio of raw materials for the bio-composites

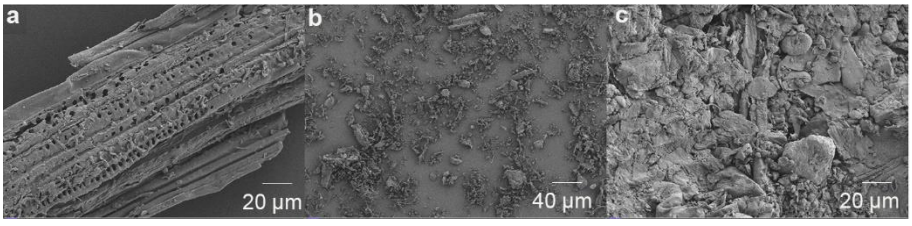

Fig. 2. SEM micrographs of the cross-sections of (a) corn stalk powder at a magnification of 500x, (b) *P. ostreatus* substrate powder at a magnification of 200x, and (c) f7 bio-composite at a magnification of 500x

Next, in order to obtain the optimal technological parameters for hot-pressing time and cooling time, bio-composites with the f7 mass ratio of raw materials were prepared at 190 ℃ with a decreased hot-pressing time (1.5 h, f32) or cooling time (2.5 h, f33). As shown in Table 2, the MOR and MOE of bio-composites f32 and f33 were far lower than the national standards for particleboard in China. This indicates that, in order to obtain bio-composites with superior mechanical properties, the hot-pressing time and cooling time should not be lower than 2 h and 3 h, respectively.

Table 2. MOR and MOE of the Control Group (f32 and f33) with Shorter Hot-press Time and Cooling Time

Sample	Hot-pressing time	Cooling time	MOR (MPa)	MOE (MPa)
f32	1.5	3	8.06±1.97	1684.02±354.2
f33	2	2.5	8.24±0.81	1475.43±310.3

The degree of water resistance of the *P. ostreatus* substrate-based bio-board (f1), bio-composites f3–f7, and corn stalk-based bio-board (f10) was determined through thickness swelling (TS) measurements. As presented in Fig. 3, the TSR value of f1 and that of the bio-composites f3 to f7 were evidently lower than that of bio-board f10. The addition of *P. ostreatus* substrate improved the water resistance of the bio-board owing to the thick waterproof mycelium coated over the *P. ostreatus* substrate (Khoo *et al.* 2020). However, despite the thick waterproof mycelium layer, the TSR of bio-board f1 was evidently higher than that of the bio-composites f3 to f7 (Fig. 3). At a high temperature, the abundant protein in the *P. ostreatus* substrate (Buendía *et al.* 2016) reacted with the abundant cellulose in the *P. ostreatus* substrate and corn stalk to form a hydrophobic ester, as evidenced by the FT-IR spectra of the bio-composites f3 to f7 (Fig. 5b). In addition, the composites f5, f6, and f7 presented the TSR values of 19.9%, 21.2%, and 18.3%, respectively, after 24 h, and each of these values exceeded the requirements (22.0%) of load-bearing particleboard for use in dry conditions according to the current national standard used in China (GB/T 4897 2015). The evident differences in the waterproof performance among the bio-composites f3 to f7 were attributed to the content of waterproof ester formed due to the self-bonding of bio-composites at high temperature and pressure depending on the mass ratio of raw materials. The water resistance of the bio-composite f7 was the best among all evaluated boards, indicating its highest content of ester formed through the cross-linking polymerization reaction of raw materials at high temperature and high pressure, leading to the strongest adhesion and water stability in this bio-composite. The highest content of ester in the bio-composite f7 was evidenced by the FT-IR spectroscopy results, as depicted in Fig. 5b.

The densities of the *P. ostreatus* substrate-based bio-board f1, bio-composites f3 to f7, and corn stalk-based bio-board f10 ranged from 1.15 to 1.34 g/cm^3 (Fig. 3b), indicating that these were high-density fiberboards. The density of the bio-composite increased continuously from f3 to f7, indicating a gradual decrease in the void spaces within the structure of the board, which led to improved water resistance in the bio-composites f3 to f7. Similar results were reported by Akgül *et al.* (2010).

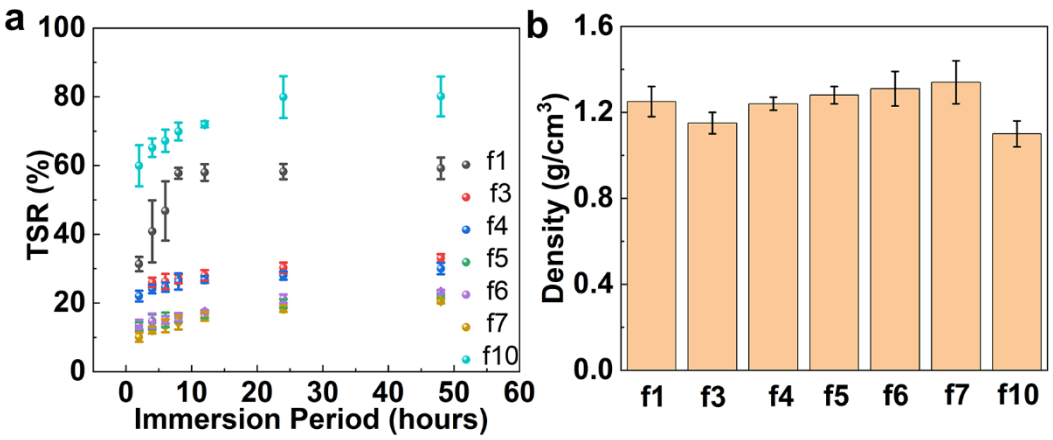

Fig. 3. (a) TSR values of the bio-board f1, bio-composites f3–f7, and bio-board f10 as a function of immersion period. (b) Densities of bio-board f1, bio-composites f3–f7, and bio-board f10 after hot-pressing.

The moisture contents (MC) of the bio-board f1, bio-composites f3 to f7, and bio-board f10 prior to and after hot-pressing are listed in Table 3. The MC of all the bio-boards prior to hot-pressing varied within the range of 5.55% to 8.16%. However, the MC was reduced to below 0.69% after hot-pressing owing to the high hot-pressing temperature.

Table 3. The MCs of Bio-board f1, Bio-composites f3 to f7, and Bio-board f10 Prior to and after Hot-pressing

Sample	MC before hot-press	MC after hot-press
f1	6.53%±0.05%	0.51%±0.05%
f3	5.55%±0.07%	0.52%±0.06%
f4	6.39%±0.04%	0.68%±0.03%
f5	8.16%±0.06%	0.66%±0.04%
f6	5.69%±0.05%	0.69%±0.03%
f7	7.89%±0.04%	0.67%±0.04%
f10	5.83%±0.06%	0.49%±0.05%

The bio-composites f3 to f7 were subjected to thermogravimetric analysis (TGA). The results are presented in Fig. 4a. The thermal stability evolution from f3 to f7 was almost consistent and could be divided into four stages. In the temperature range of 20 to 200 °C, the mass loss of the bio-composites f3 to f7 fluctuated between 4.4% and 6%, which was attributable to the evaporation of water and light volatiles (Hu *et al.* 2016). In the temperature range of 200 to 420 °C, the mass loss of bio-composites f3 to f7 fluctuated from 55% to 62%. This stage corresponded to the thermal degradation of cellulose and lignin, which specifically involved the rupture of intermolecular and intramolecular hydrogen bonds and the generation of water in cellulose, with the polymerization degree of cellulose decreasing from 10,000 to 200, along with the breaking of the β-O-4 linkages in lignin (Yeo *et al.* 2019). In the temperature range of 420 to 600 °C, the mass loss of bio-composites f3 to f7 fluctuated between 7% and 8%, which was consistent with the thermal decomposition of lignin after 400 °C (Yeo *et al.* 2019). In the temperature range of 600 to 900 °C, the mass loss of bio-composites f3 to f7 fluctuated between 9% to 12%, which corresponded to the further pyrolysis reaction of lignin, with the simultaneous breaking of various types of bonds (Yeo *et al.* 2019).

The DTG results reflected the mass loss rates, and the DTG curves of bio-composites f3 to f7 are presented in Fig. 4b. In the present study, the temperature corresponding to the maximum thermal degradation rate was denoted as "T_{DTGmax}". The T_{DTGmax} values determined for f3, f4, f5, f6, and f7 were 335.9, 331.9, 339.22, 330.3, and 330.3 °C, respectively, indicating the excellent thermal stability of f5 among bio-composites f3 to f7.

In summary, the thermal stability of the bio-composites prepared in the present study was superior to that of conventional wood materials, with the latter exhibiting deteriorated thermal stability at a much lower temperature of 120 °C. However, to meet the fire protection standards, further treatment of these bio-composites is necessary.

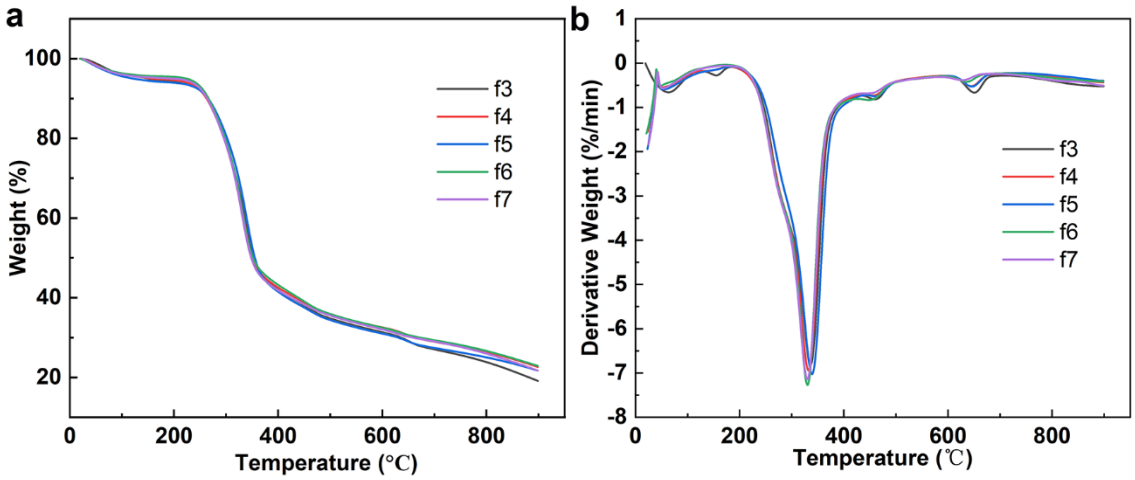

Fig. 4. (a) TGA results of bio-composites f3–f7 for the temperature range of 20 to 900 °C. (b) DTG results for bio-composites f3–f7 for the temperature range of 20–900 °C

The FT-IR spectra of the raw materials, namely, the *P. ostreatus* substrate and corn stalk, and the bio-composite f7 prior to and after compression, were recorded and analyzed to decipher the reaction mechanism underlying the self-bonding of bio-composites after heat-compression. The comparison of the IR vibrations of the *P. ostreatus* substrate, corn stalk, and the bio-composite f7 prior to and after heat compression is illustrated in Fig. 5a, and the numerical results are presented in Table 1. The IR peaks of the four specimens from 3300 to 3350 cm^{-1} corresponded to the O–H symmetric stretching vibrations of the free and intermolecular bonded hydroxyl groups (Kotilainen *et al.* 2000). The IR peaks of the four specimens from 2910 to 2930 cm^{-1} corresponded to the C–H asymmetric stretching vibrations (Kotilainen *et al.* 2000; Khoo *et al.* 2020). The IR peaks from 1720 to 1735 cm^{-1} were assigned to C=O stretching vibration. This C=O band vibration remained almost undetected in the *P. ostreatus* sample spectra while appearing with weak intensity absorbance counts in the spectra of corn stalk. It was, therefore, attributed to the acetyl groups and other carbonyl groups of carboxylic acids in hemicelluloses (Delmotte *et al.* 2008). This C=O band vibration appeared with higher intensity absorbance counts in the bio-composite f7 after hot-pressing compared to that in the corn stalk and bio-composite f7 prior to hot-pressing. This finding indicated the thermal degradation of lignocellulose and the self-bonding of corn stalk and the *P. ostreatus* substrate during hot-pressing. The peaks that appeared at 1151 cm^{-1} and between 1230 and 1240 cm^{-1} were attributed to the C–O stretching vibration (Aboulkas *et al.* 2017; Khoo *et al.* 2020). Considering the increasing intensity absorbance counts of the C=O band vibration in the bio-composite f7

after compression, it was concluded that increased ester linkages were formed during the process of hot-pressing. According to previous reports (Mussatto *et al.* 2004; Le *et al.* 2016), the thermal degradation products of hemicellulose react with lignin to form ester linkages, which is of great significance for the self-bonding of bio-composites. In addition, the abundant protein in mycelium reacts with lignocellulose in both corn stalk and the *P. ostreatus* substrate, resulting in increased ester linkages in the bio-composite. The IR peaks that appeared from 1630 to 1640 cm^{-1} were responsible for the C=C stretching vibration corresponding to lignin aromatic groups. As depicted in Fig. 4, the intensity absorbance counts of C=C stretching in the bio-composite f7 remained almost constant after compression relative to that prior to compression, suggesting that lignin was hardly degraded during hot-pressing. The IR peak that appeared between 1020 and 1030 cm^{-1} corresponded to the C-O stretching vibration, which could have originated from lignin and saccharides. It is noteworthy that this C-O stretching vibration exhibited evidently an increase in the intensity absorbance counts in the bio-composite f7 after compression relative to *P. ostreatus*, corn stalk, and bio-composite f7 prior to compression, indicating the thermal degradation and cross-linking of raw materials in the bio-composites during hot-pressing.

As discussed above, the mass ratio of the raw materials, *i.e.*, *P. ostreatus* substrate and corn stalk, significantly influences the waterproof performance of bio-composites. The FTIR spectra of bio-boards f1, f3 to f7, and f10 after heat compression were compared (Fig. 5b). It was observed that the intensity absorbance counts of the C=O band vibration in bio-composite f7 were evidently stronger than those of the other bio-boards, indicating that the content of waterproof ester was higher in f7 compared to the other bio-boards, which led to a superior waterproof performance of this bio-composite.

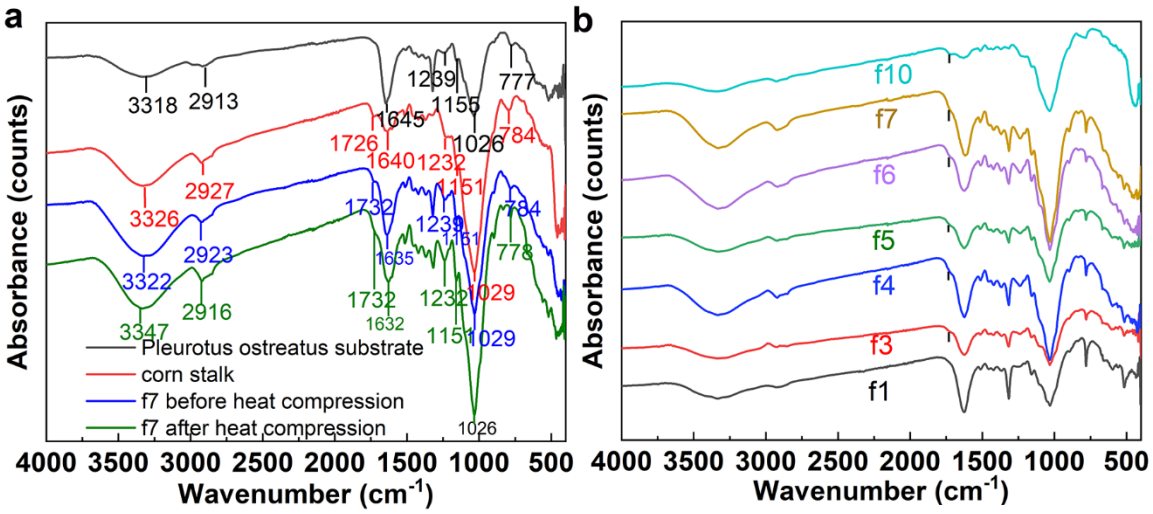

Fig. 5. FTIR spectra of (a) the *P. ostreatus* substrate and corn stalk, the raw material of bio-composite f7 prior to and after heat compression, and (b) bio-boards f1, f3 to f7, and f10 after heat compression. The black vertical line in the FTIR spectra of bio-composites f3 to f7 and f10 at around 1730 cm^{-1} corresponds to the C=O band vibration.

Relative crystallinity is an effective standard for assessing the physical and mechanical properties of bio-composites. A neater-arranged cellulose chain indicates higher mechanical strength, density, and dimensional stability (Agarwal *et al.* 2010). In the present study, X-ray diffraction measurements were performed to determine the relative

crystallinity of the *P. ostreatus* substrate-based bio-board f1, corn stalk-based bio-board f10, and *P. ostreatus* substrate/corn stalk bio-composites f3 to f7 after heat compression. As depicted in Fig. 6b, sharp peaks appeared at 14.3°, 14.9°, and 22.3° in bio-composites f3 to f7, indicating the existence of Type I cellulose (Pickering *et al.* 2007), which contributes to the high crystal stability of the composite. The XRD results of bio-composite f7 prior to and after compression are presented in Fig. 6a. As depicted in the inset of Fig. 6a, the relative crystallinity of bio-composite f7 increased after hot-pressing, owing to the degradation of hemicellulose and a small portion of amorphous cellulose at a high temperature. Theoretically, the improved relative crystallinity after hot-pressing is directly related to the higher kinetic energy that stretches the polymer within the lignocellulose matrix at a higher temperature (Khoo *et al.* 2020). The relative crystallinity results for bio-boards f1, f3 to f7, and f10 are presented in Fig. 6c. The extremely low relative crystallinity of bio-board f1 could be associated with the degradation of cellulose by white rot fungi in the *P. ostreatus* substrate (Baker *et al.* 2016). In addition, the relative crystallinity of f10 was lower than that of the bio-composites f3 to f7, indicating that, compared to bio-boards f1 and f10, bio-composites f3 to f7 had stronger intermolecular bonds, which resulted in a denser structure (Miyata and Masuko 1988), superior mechanical properties, and greater water resistance in bio-composites f3 to f7 relative to the *P. ostreatus* substrate-based and corn stalk-based bio-boards.

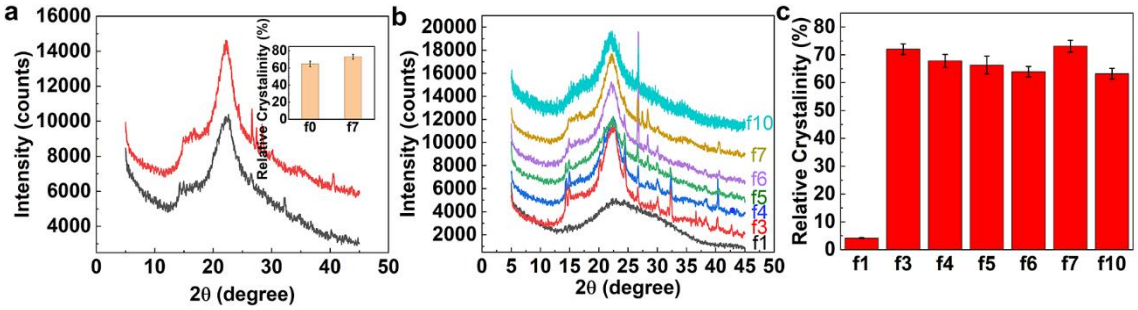

Fig. 6. (a) The XRD results of bio-composite f7 prior to (black curve) and after (red curve) hot-pressing. The inset depicts the relative crystallinity of bio-composite f7 prior to (f0) and after heat compression (f7). The condition f0 denotes the raw material for the preparation of bio-composite f7 prior to hot-pressing. (b) The XRD results of bio-boards f1, f3 to f7, and f10 after heat compression. (c) The relative crystallinity of bio-boards f1, f3 to f7, and f10 after heat compression

As discussed above, the addition of *P. ostreatus* substrate powder into corn stalk powder effectively improved the mechanical properties and water resistance of bio-boards. Moreover, both the mechanical properties and water resistance of bio-composites f5 to f7 have far surpassed the China national standards for loading-bearing particleboard, suggesting a novel idea to dispose of the waste bio-resources effectively. In summary, among bio-composites f5 to f7, f7 possesses superior IBS, MOR, MOE, and water resistance in comparison to other bio-composites. That is, the technological parameters of f7 (Table 1) should be used to obtain bio-composites with optimal mechanical properties and water resistance.

CONCLUSIONS

1. The mechanical properties and water resistance of the bio-composites prepared in the present study were evidently superior to the corn stalk-based and *P. ostreatus* substrate-based bio-boards and much beyond the requirements for particleboard according to the national standards used in China. This was attributable to the increasing ester linkages and stronger intermolecular bonding in the bio-composites after hot-pressing.
2. In order to prepare bio-composites with mechanical properties and water resistance much better than the national standards for particleboard used in China, the optimal mass ratio of *P. ostreatus* substrate/corn stalk/deionized water was 3:6:1, the optimal hot-press temperature was 190 °C, and the optimal hot-press duration and cooling time were 2 h and 3 h, respectively.
3. The bio-composites were prepared using the dry process, which is more environment-friendly compared to the wet process. In addition, the technological process is simple, green, and convenient for popularization and application, and it has, therefore, several applications, such as children's toys and furniture, that result in minimum physical injuries.
4. The development of bio-composites is an effective solution for the shortage of forestry resources and waste disposal and reuse, and should, therefore, be considered to be of great significance in terms of greater ecological and economic benefits.

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