Chen *et al. Journal of Wood Science* (2023) 69:17
https://doi.org/10.1186/s10086-023-02092-3

Journal of Wood Science

ORIGINAL ARTICLE

Open Access

Study on preparation of high-performance binderless board from *Broussonetia papyrifera*

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Abstract

Binderless particleboards were prepared without any resin adhesives from *Broussonetia papyrifera* trunk by hot pressing. The effects of particle size, pressing time, pressing temperature and board density on the physical and mechanical properties of binderless boards were investigated. The effects of chemical changes in *Broussonetia papyrifera* binderless particleboards on the board properties were investigated by chemical, spectroscopic and cellulose crystallinity analyses. The bonding performance of the latex of *Broussonetia papyrifera* was discussed. The results showed: (1) the smaller size of the particle, the higher internal bonding (IB) strength and lower thickness swelling (TS) of the binderless board had. The modulus of rupture (MOR), the modulus of elasticity (MOE) and the IB values of the boards increased with the increase of board density. Within a certain range, the binderless boards manufactured at higher temperature and longer pressing time had better properties. Under the optimum board manufacturing condition of 220 ℃/30 min/1.0 g/cm^3, the *Broussonetia papyrifera* binderless board recorded a MOR 28 MPa, MOE 5.3 GPa, IB 2.74 MPa and 24 h TS of 7.4%, which met the performance requirements of Chinese national standard for heavy-duty particleboard. (2) The degradation of cellulose and hemicellulose during hot pressing resulted in decrease of cellulose and hemicellulose content and increase of extractives. Some of the degradation products form new bonding to increase the bonding strength and dimensional stability of the binderless boards. (3) The formation of pseudo-lignin and increased C–O–C and cellulose crystallinity of the boards during hot pressing contributed to high quality of the binderless boards. (4) The poplar veneers bonded with the latex of *Broussonetia papyrifera* had a certain bonding strength (0.6 MPa), indicating the latex played a positive role in self-bonding of the *Broussonetia papyrifera* particleboard.

Keywords *Broussonetia papyrifera*, Manufacturing conditions, Self-bonding, Heavy-duty load-bearing particleboard

Introduction

The annual consumption of wood in China is about 5.57×10^8 m^3, of which nearly 50% of wood depends on imports [1]. To solve the problem of insufficient wood resources, a large area of forest has been planted in China and has reached 79.54 million hectares currently [1]. *Broussonetia papyrifera* (L.) Vent. is a common fast-growing tree and widely distributed in China [2], 68,034 hectares of *Broussonetia papyrifera* had been planted by the end of 2018 [1]. It has strong growth tolerance and excellent fiber quality, and its growth cycle is 3–4 times shorter than poplar [2]. *Broussonetia papyrifera* has been known to be widely used in the fields of feed, paper making and land rocky desertification restoration [3]. Early works proposed that the trunk and branches of *Broussonetia papyrifera* could be used to prepare wood-based panels [4]. Manufacturing wood-based panel products from *Broussonetia papyrifera* would be a valuable alternative, with a potentially large market. Jiang [4] prepared homogeneous particleboard using *Broussonetia papyrifera* wood as raw material, and urea–formaldehyde resins as adhesive,

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finally obtaining particleboard with the properties of the modulus of rupture (MOR) 17.2 MPa, the modulus of elasticity (MOE) 2.5 GPa, the internal bonding (IB) strength 1.24 MPa and 2 h thickness swelling (TS) values less than 8.0%.

Although synthetic adhesives are typically used for wood-based panel production, most of them contain formaldehyde which can affect people's health [5]. The research on binderless boards can solve the problem of formaldehyde release from the root. In the past, binderless boards were mainly manufactured from non-wood lignocellulosic materials (straw, kenaf and bagasse etc.) [6] due to their high hemicelluloses content. Degradation of hemicellulose during hot pressing to produce furan products is believed to play an important role in self-bonding [7]. While only a few of wood materials such as oil palm and oak [8, 9] were used for binderless particleboard manufacturing. In contrast to most of wood species, the whole tree of *Broussonetia papyrifera* contains latex—a white sticky emulsion. The chemical components of latex mainly contain starch grains, polysaccharides, terpenes, phenols, flavonoids, organic acids and fatty acids. The PH value of latex is around 5.8–6.2. It has been found the latex has antibacterial effect and is currently used as Chinese medicine for curing skin disease [2, 10]. Now, we find the latex is sticky and the *Broussonetia papyrifera* wood which contains latex may be a potential material for preparing binderless board.

Binderless board is an environmental-friendly material, whereas its strength and water resistance property are not as good as the board with adhesive at similar manufacturing conditions [11], which reduce their use value. Thus, many studies on steam/heat treatment such as steam-explosion or steam-injection pressing have been reported as methods to improve the self-bonding property [12, 13]. However, these treatments all require special equipment and a great deal of energy consumption, which limits their development and application [14]. Increasing board density, pressing temperature and prolonging pressing time can all improve the properties of binderless boards by conventional hot pressing, this method is simple to operate. However, boards with high densities have high compaction ratios making it difficult for the steam inside the board to escape, thereby producing high steam pressure inside the board. When the steam pressure is higher than the internal bonding strength of the board at downloading, delamination is easy to happen [12]. It was found that cooling the hot-pressing boards with a hydronic cooling system before opening the hot press can effectively solve this problem and has been apply to our previous study to prepare fire-retardant high-density binderless particleboard from bagasse [15].

Table 1 Distribution of particle size of *Broussonetia papyrifera* based on mesh analysis

Particle size	Mesh no. (%)			
	> 20	20–40	40–60	< 60
Coarse	69.3	23.1	4.3	3.3
Fine	40.8	35.3	10.6	13.3

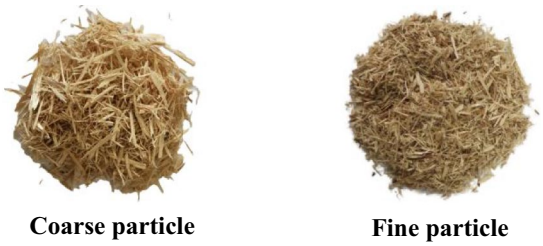

Fig. 1 Broussonetia papyrifera particles

In this study, binderless boards were manufactured from *Broussonetia papyrifera* trunk without any resins by conventional hot pressing. The effects of pressing conditions (particle size, pressing time, pressing temperature and board density) on the board's properties were investigated. In addition, the bonding mechanism of *Broussonetia papyrifera* binderless boards was preliminary studied.

Materials and methods

Particle preparation

Broussonetia papyrifera wood from the Jindong forests farm, Yongzhou City, Hunan Province, China was used as the raw material. The *Broussonetia papyrifera* wood was first cut into chips of about 5 cm long and adjusted the chips to the MC of approximately 60%, then crushed by a knife-ring flaker (PZ8, Pallmann, Germany) for twice to obtain coarse particles. Fine particles were obtained from coarse particles that passed through a 5-mesh screen. The particles were air-dried to a MC of around 10%. Table 1 shows the distribution of particle size of the *Broussonetia papyrifera* based on mesh analysis. Figure 1 shows the geometries of particles. The above two particles were used to prepare binderless particleboard.

Binderless particleboard manufacture

The dimensions of the boards were 200 × 200 × 6.5 mm, and the thickness of the particleboard was controlled by 6.5 mm-thick distance bars. The MC of particles were adjusted to 16% by spraying water onto the air-dried particles. The target board densities were set at 0.8, 1.0 and 1.2 g/cm^3. The pressing temperatures were 180 ℃, 200 ℃ and 220 ℃. The pressing times were 15 min, 30 min and

45 min. The pressure was 4–6 MPa. The manufacturing conditions of binderless boards are shown in Table 2. The particles were weighed according to the density of the boards, then particles were hand-formed into a particle mat using a forming box, followed by hot pressing into the particleboard. After the pressing time was over, cooling water was introduced into the hot-pressing system. When the platen temperature dropped below 100 ℃, the binderless boards can be discharged. Figure 2 shows the preparation route of *Broussonetia papyrifera* binderless boards.

Evaluation of physical and mechanical properties of binderless board

Prior to the evaluation of the mechanical and physical properties, the boards were conditioned at ambient conditions (around 18–25 ℃, with the relative humidity of 55–65%) for about 1 week, reaching a MC of 5–8%. Then, tested according to the test methods of evaluating the properties of wood-based panels GB/T 17657-2013 [16]. The MOR and MOE tests were conducted on three $180 \times 50 \times 6.5$ mm specimens cut from each board, by a three-point bending test over an effective

Table 2 Manufacturing conditions and test results of *Broussonetia papyrifera* binderless boards

No.	Manufacturing conditions					Properties			
	Particle size	Temperature (℃)	Time (min)	Pressure(MPa)	Density (g/cm^3)	MOR (MPa)	MOE (MPa)	IB (MPa)	24 h TS (%)
1	F	180	15	4.0–6.0	1.0	15	2475	0.50	88.8
2	F	180	30	4.0–6.0	1.0	20	2897	0.88	42.5
3	F	180	45	4.0–6.0	1.0	26	4329	1.39	37.3
4	F	200	15	4.0–6.0	1.0	20	3605	1.24	27.5
5	F	200	30	4.0–6.0	1.0	26	3735	1.49	26.5
6	F	200	45	4.0–6.0	1.0	27	4527	2.56	20.2
7	F	220	15	4.0–6.0	1.0	25	3580	1.36	15.9
8	F	220	30	4.0–6.0	1.0	28	5316	2.74	7.4
9	F	220	45	4.0–6.0	1.0	23	3827	2.98	5.0
10	F	200	30	4.0–6.0	0.8	12	1806	0.52	31.2
5	F	200	30	4.0–6.0	1.0	26	3735	1.49	26.5
11	F	200	30	4.5–6.5	1.1	27	4865	2.60	14.3
12[a]	F	200	30	4.5–6.5	1.2	–	–	–	–
13[a]	F	220	30	4.5–6.5	1.2	–	–	–	–
14	C	180	30	4.0–6.0	0.8	7	1263	0.21	73.2
15	C	200	30		0.8	9	1240	0.45	45.2
16	C	200	30		1.0	25	3605	0.64	37.5
17	C	200	15		1.0	17	2304	0.28	61.7
18	C	220	15		1.0	23	3537	0.97	30.5

C coarse, *F* fine

[a] The no.12 and no. 13 binderless boards delaminated during hot pressing, its physical and mechanical properties cannot be tested

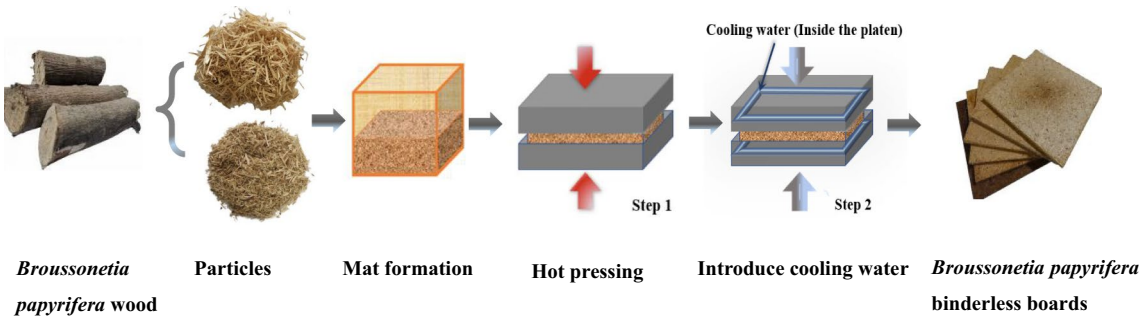

Fig. 2 Preparation route of *Broussonetia papyrifera* binderless board

span of 140 mm at a loading rate of 5 mm/min. IB and TS test specimens were then prepared from MOR samples, three 50 × 50 × 6.5 mm test specimens for the IB test at a loading rate of 5 mm/min. All tests were completed by the testing machine (MWD-10, Naier Testing Machine Co., Ltd., Jinan, China). Three 50 × 50 × 6.5 mm test specimens were for the TS test (24 h immersion in 20 ℃ water). The physical and mechanical properties are evaluated according to the Chinese national standard for particleboard GB/T 4897-2015 [17], as shown in Table 3.

Chemical analysis of *Broussonetia papyrifera* binderless boards

Because no binder is added during binderless board manufacturing, the change of chemical composition is believed to play an important role for self-bonding. To understand the bonding mechanism, the chemical analysis was done on some typical binderless boards. Considering the pressing temperature has a decisive effect on the board performance, we chose 3 binderless boards from different temperatures (180、200、220 ℃) as the samples to carry out chemical analysis. The pressing time was fixed to 30 min, and the board density was 1.0 g/cm^3, which were appropriate for binderless board preparation. The raw material (*Broussonetia papyrifera* particle) was also tested as a control.

Chemical composition analysis of Broussonetia papyrifera particles and boards

The *Broussonetia papyrifera* particles and binderless boards were ground to pass through a 40-mesh screen and retained on a 60-mesh screen for chemical composition analysis. The content of chemical compositions was determined in accordance with the Chinese national standard for analysis methods of forestry biomass raw materials [18, 19] and the Chinese paper industry standard [20]. The content of holocellulose was determined by the wise method [18]; Klason lignin was measured by the Klason method [18]; The sample was extracted with the mixture of benzene and ethanol (2:1, v/v) and refluxed at 90 ℃ for 6 h to determine the alcohol–benzene extractive of the material [19]; in addition,the samples were extracted with distilled water for 3 h at the temperature of 95–100 ℃ to obtain the hot water extractive [19]; The α-cellulose content was determined by nitric acid ethanol method [19]. All the chemical analyses were carried out in duplicate, and the error of the two samples was no more than 2%.

Fourier-transform infrared spectrum (FTIR) analysis of the binderless boards

Broussonetia papyrifera particles and binderless boards were crushed and ground to pass through a 200-mesh screen for the FTIR analysis. The potassium bromide tablet pressing method was used for the test (IRAffinity-1 Fourier infrared spectrometer, Shimadzu Corp., Kyoto, Japan). The scanning range was 500–4000 cm^{-1}, recorded by the means of an average of 32 scans at a resolution of 2 cm^{-1}, and the scanning was repeated 3 times.

X-ray diffraction (XRD) analysis of the boards and raw materials

X-ray diffraction was used to investigate the effects of hot-pressing temperature on the relative crystallinity of cellulose of *Broussonetia papyrifera* binderless boards. The raw materials and binderless boards were made into powders. Ni-filtered copper radiation (λ = 0.154 nm) generated at a voltage of 40.0 kV and current of 40.0 mA was utilized, and a scan speed of 2°/min from 5 to 50° was used (Rigaku SmartLab SE, Japan). The relative cellulose crystallinity was calculated based on Segal methods.

Analysis of the bonding performance of *Broussonetia papyrifera* latex

FTIR analysis of Broussonetia papyrifera latex

The latex was collected from the twig of *Broussonetia papyrifera* tree. It was cured by drying at 100 ℃ for 2 h and then was ground to pass through a 200-mesh screen for FTIR analysis. The instrument and analysis method and scaning parameters were the same as the FTIR analysis for binderless boards mentioned earlier.

Analysis of Broussonetia papyrifera latex effect on self-bonding of the boards

The poplar veneers (Shandong Province, China) were cut into 80 × 30 mm pieces for testing the bonding strength

Table 3 Standard for physicochemical properties of particleboard of China [17] (6 mm < thickness < 13 mm)

Type	Use (dry conditions)	Physicochemical properties				
		MOR (MPa)	MOE (MPa)	IB (MPa)	2 h TS (%)	24 h TS (%)
P1	General purpose particleboard	10.5	–	0.23	–	–
P2	Furniture grade particleboard	11	1800	0.4	8	–
P3	Load bearing particleboard	15	2200	0.4	–	19
P4	Heavy-duty load bearing particleboard	20	3100	0.6	–	16

using *Broussonetia papyrifera* latex as adhesive. Bonding strength specimens were obtained by applying latex on the surface (1/2 of total area) of two veneer pieces, the specimens were then pressed at 180 ℃ for 10 min. They were subsequently conditioned at ambient conditions for 2 days and the dry bonding strength was tested at a loading rate of 10 mm/min [16]. Specimens without latex were used as the control groups. The preparation route is shown in Fig. 3.

Results and discussion

Physical and mechanical properties

The test results of the physical and mechanical properties of the binderless boards are shown in Table 2. The actual densities of the test specimens were a little different with the target board densities. However, the deviation was usually less than (±0.05) g/cm^3. Because density has a significant effect on the board performance, all of the experimental values obtained were corrected to the target board densities based on the linear regression between board density and properties. The linear regression lines on the relationships between board density and properties were taken first. Each plot was then shifted to the targeted densities according to the slope of the linear regression line, and the average values were calculated.

Effects of size of particles on properties of binderless boards

The effects of particle size on board properties are shown in Fig. 4. It was found that almost all of the properties (except MOE at 200 ℃/30 min and 220 ℃/15 min) of the binderless boards made of fine particles were better than that of coarse particles under the same manufacturing conditions. The smaller size of the particles, the higher value of the MOR and MOE of the binderless board had. Compared with MOR and MOE (Fig. 4a, b), a more significant change was observed between the IB, TS values and particle size. Especially at 200 ℃/15 min, the IB value of the boards made from fine particles was 1.24 MPa, but those made from coarse particles were just 0.28 MPa (Fig. 4c). In addition, the 24 h TS decreased by 55.4% from coarse particle to fine particle (Fig. 4d). The bending strength of binderless particleboard depended on both particle strength and the self-bonding between them. The smaller size of the particle, the larger the specific surface area it had, and the contact area between particles increased, this greatly enhanced their self-bonding which showed high performance and dimension stability of the boards [21]. Therefore, the boards made from fine particles had better performance.

Effects of density on the properties of binderless boards

Mechanical and physical properties of the binderless boards at different board densities are shown in Fig. 5. The values of MOR, MOE and IB increased with the increasing of board density. The MOR, MOE and IB were 12 MPa, 1.8 GPa and 0.52 MPa at a board density of 0.8 g/cm^3, whereas they were 27 MPa, 4.9 GPa and 2.6 MPa at a board density of 1.1 g/cm^3 (Fig. 5a–c), respectively, a significant increase. High mechanical strength of binderless board at higher board density was attributed to the high compaction ratio in the board which made the particles bond intimately and beneficial to board strength. The increase in the heat conductivity of boards with increasing board density caused more degradation of the cellulose and hemicellulose of the particles [14] might also contribute to the increase of bonding strength of the boards.

In general, for conventional particleboards (bonded with adhesive), the TS after long-duration water soaking have a tendency to increase with increasing board density because of the greater springback of pressed particles in boards of higher density [22].

However, in this study, the TS value of binderless particleboard decreased with increasing board density. This might attribute to the following reasons. The TS value, on one hand, depends on the bonding strength among particles. The particles in high density binderless board bond intimately and the water is not easy to penetrate into the board. However, on the other hand, TS depends on the springback of the board. High density board also has a high compaction ratio thus has high springback. The combination of these two factors determines the TS value. High density binderless board exhibit a low TS indicates that the higher bonding strength at high density

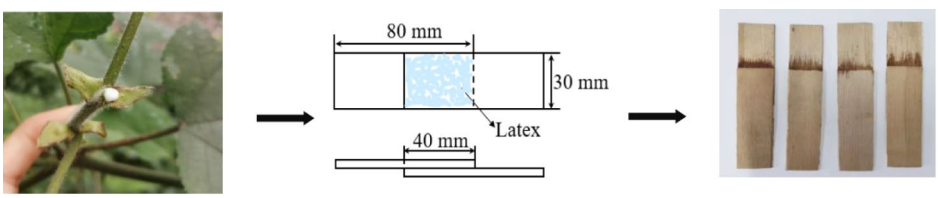

Fig. 3 Preparation of bonding specimens by bonding poplar veneer with *Broussonetia papyrifera* latex

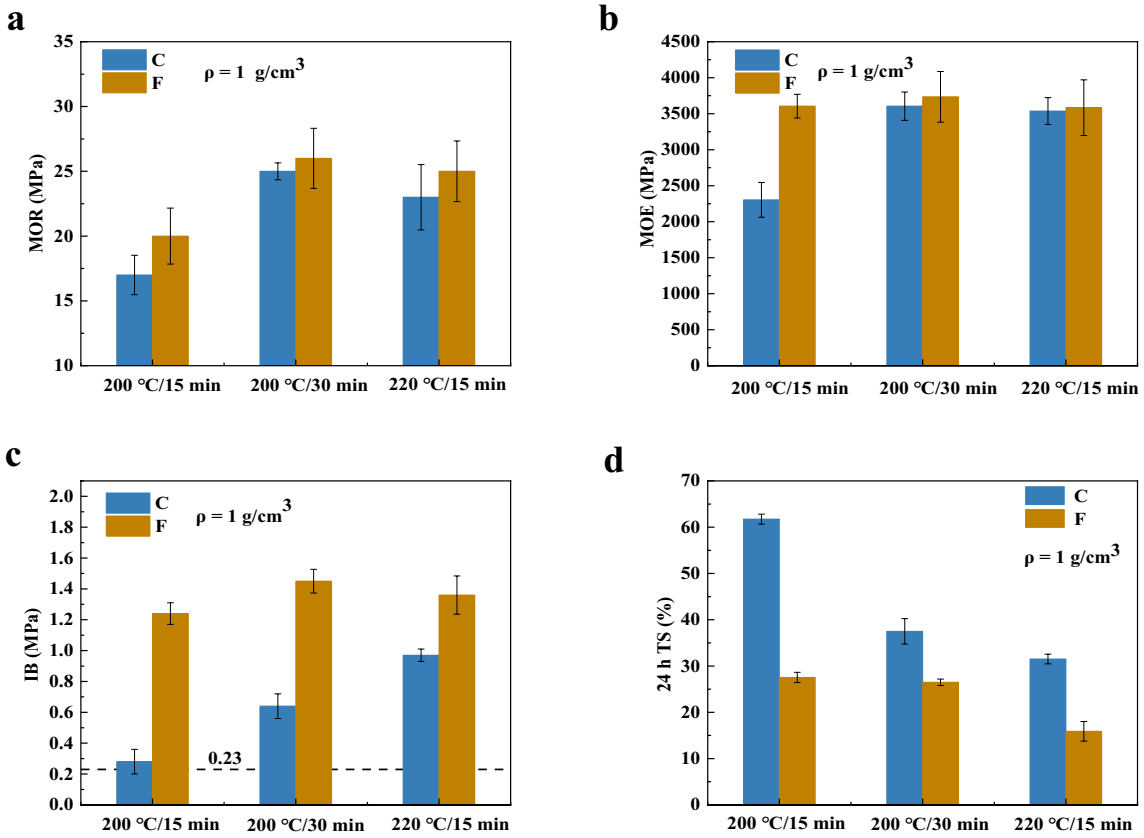

Fig. 4 Effects of particle size on physical and mechanical properties of binderless boards. **a** Effects of particle size on the MOR of binderless board. **b** Effects of particle size on the MOE of binderless board. **c** Effects of particle size on the IB of binderless board. **d** Effects of particle size on 24 h TS of binderless board. Error bars indicate standard deviations

played a more dominant role than the compression ratio of particles [14]. The TS value was 14.3% with a density of 1.1 g/cm^3 (Fig. 5d).

Effects of pressing temperature and time on properties of binderless boards

The effects of pressing temperature and time on the properties of binderless boards are shown in Fig. 6. Within a certain range, the binderless boards manufactured at higher temperature and longer pressing time had better properties. At 180 ℃/15 min, the MOR and MOE values were 15 MPa and 2.5 GPa (Fig. 6a, b), respectively. IB and TS also recorded relatively poor values of 0.5 MPa and 88.8% (Fig. 6c, d), respectively. Some research found that to acquire high quality binderless board, high temperature, a certain pressing time and high compactness were needed [8, 12, 23, 24], thus the temperature and pressing time were the decisive parameters of the outcome [24]. The cellulose, hemicellulose and lignin components in particles did not degrade significantly at low temperature and short pressing time, which resulted in a poor self-bonding between particles and gave poor strength to the boards.

The MOR and MOE values of the board with the condition of 220 ℃/30 min were 28 MPa and 5.3 GPa, respectively, which were 13 MPa and 2.8 GPa more than tthe board manufactured at 180 ℃/15 min (Fig. 6a, b), indicating that higher pressing temperature and longer pressing time contributed to improved board property. However, the MOR and MOE decreased when the hot pressing time further increased from 30 to 45 min at 220 ℃. The reason might be due to the MOR strength not only depended on the bonding strength between particles but also related to the own strength of the material [25]. Too high of pressing temperature and too long of pressing time resulted in severe degradation of chemical components, which destroyed the support structure of the materials thus deteriorated the self-strength of the particles and showed a lower value of MOR and MOE. The IB values kept increasing for a longer time which recorded a value of 1.36 MPa at 30 min and 2.98 MPa at 45 min (Fig. 6c). Similar to the IB, the TS value was only 5.0% at 45 min (Fig. 6d). A long pressing time contributed

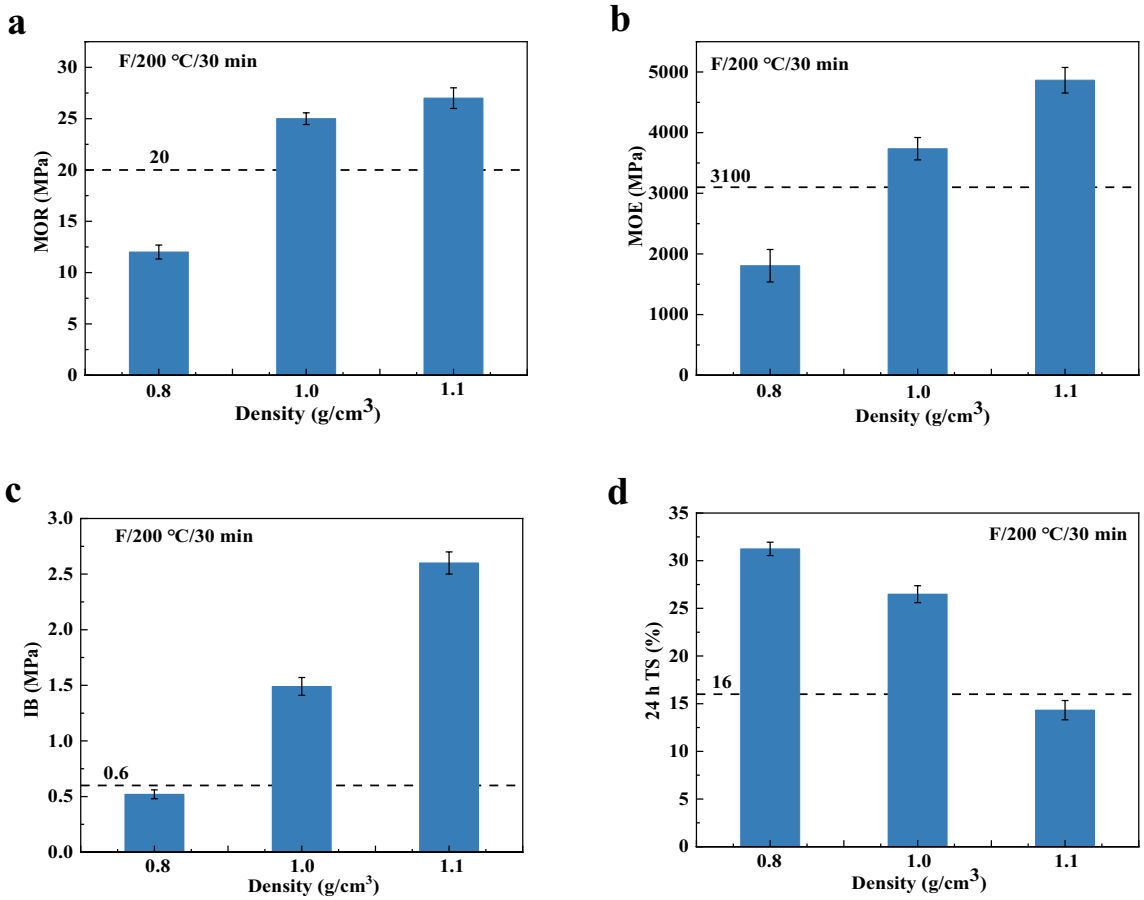

Fig. 5 Effects of board density on physical and mechanical properties of binderless boards. **a** Effects of board density on the MOR of binderless board. **b** Effects of board density on the MOE of binderless board. **c** Effects of board density on the IB of binderless board. **d** Effects of board density on the 24 h TS of binderless board. Error bars indicate standard deviations

to low TS and thus better dimensional stability, there had been a similar report before [25]. However, compared with pressing time, higher pressing temperature showed more advantages for degradation, crosslinking and polymerization of elements [26]. Based on Fig. 6, the optimum conditions for manufacturing high-density binderless particleboard were at 220 ℃/30 min/1 g/cm^3, the MOR, MOE, IB and 24 h TS values were 28 MPa, 5.3 GPa, 2.74 MPa and 7.4%, respectively, all properties were far surpass the requirements of type-P4 particleboard [17] (20 MPa, 3.1 GPa, 0.6 MPa and 16%).

Effects of the change of chemical composition of binderless boards on boards properties

The change in the chemical composition of raw materials and binderless boards is shown in Table 4. The cellulose and hemicellulose contents decreased, while the amount of the extractives and lignin content increased with increasing pressing temperature. Sugars (cellulose and hemicellulose) in particles degraded under high temperature and generated low molecular unsaturated compounds and carbonyl compounds [27, 28], which resulted in a decrease in sugar while increased the extractive content. Lignin content in the binderless board increased compared with the raw material. As some of the repolymerization substance of sugar degradation products (such as furfural) can be measured as "lignin" (also called pseudo-lignin) [29–32], and hemicellulose cleavage products can contribute to polymerization reactions of lignin during curing, which might also result in high values for lignin [33]. Because resin was not used in the binderless boards, self-bonding strength was significantly affected by the changes in the chemical composition, especially on IB and TS.

Correlation between the change in chemical composition and IB strength of the binderless board

The correlation between the chemical composition and IB strength of the binderless boards is shown in Fig. 7. It was found that IB strength increased with an increased

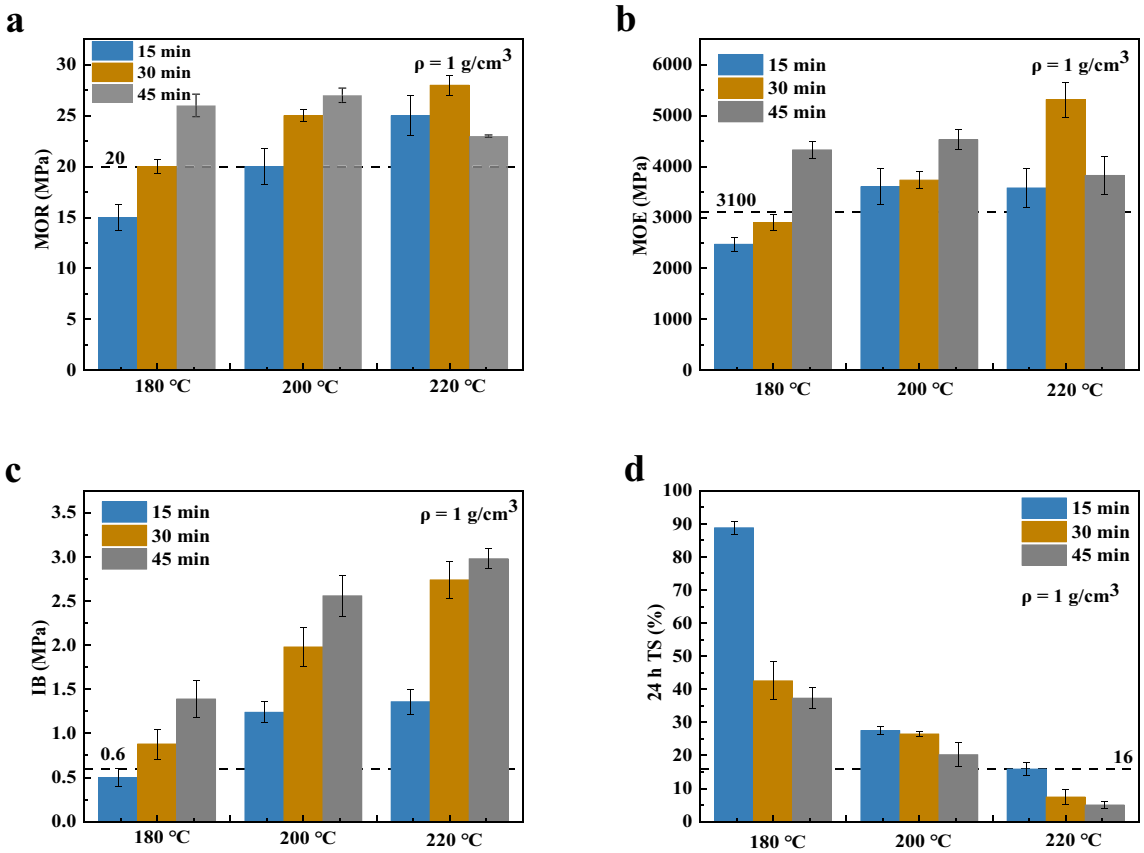

Fig. 6 Effects of pressing temperature and pressing time on physical and mechanical properties of binderless board. **a** Effects of pressing temperature and time on the MOR of binderless board. **b** Effects of pressing temperature and time on the MOE of binderless board. **c** Effects of pressing temperature and time on the IB of binderless board. **d** Effects of pressing temperature and time on 24 h TS of binderless board. Error bars indicate standard deviations

Table 4 Chemical composition of raw materials and binderless boards at different temperatures of *Broussonetia papyrifera*

	Chemical components (%)				Extractive (%)	
	Holocellulose	α-Celluose	Hemicellulose	Lignin	Hot water	Alcohol–benzene
Raw material	81.20	50.45	30.75	19.28	3.34	1.47
180 ℃ board	76.84	47.21	29.63	21.88	5.87	2.44
200 ℃ board	73.87	44.91	28.96	23.16	9.29	3.52
220 ℃ board	67.50	42.67	24.83	28.38	10.63	6.13

amount of hot water and alcohol–benzene extractive (Fig. 7a). A similar correlation was found between the IB strength and the degree of hemicelluloses and cellulose degradation (Fig. 7b, c). Due to the degradation of hemicellulose, furan intermediates were formed and polymerized during hot pressing [34, 35], thus forming binders to bond the particles together. Besides, the degradation of polysaccharides and softening of lignin destroyed the structure of particles [36] which gave a higher IB value to the boards (Fig. 7d).

Correlation between the change in chemical composition and TS of binderless boards

Figure 8 shows the correlation between the TS value and the change in chemical composition. Because no binder was used in the boards, the TS value was largely

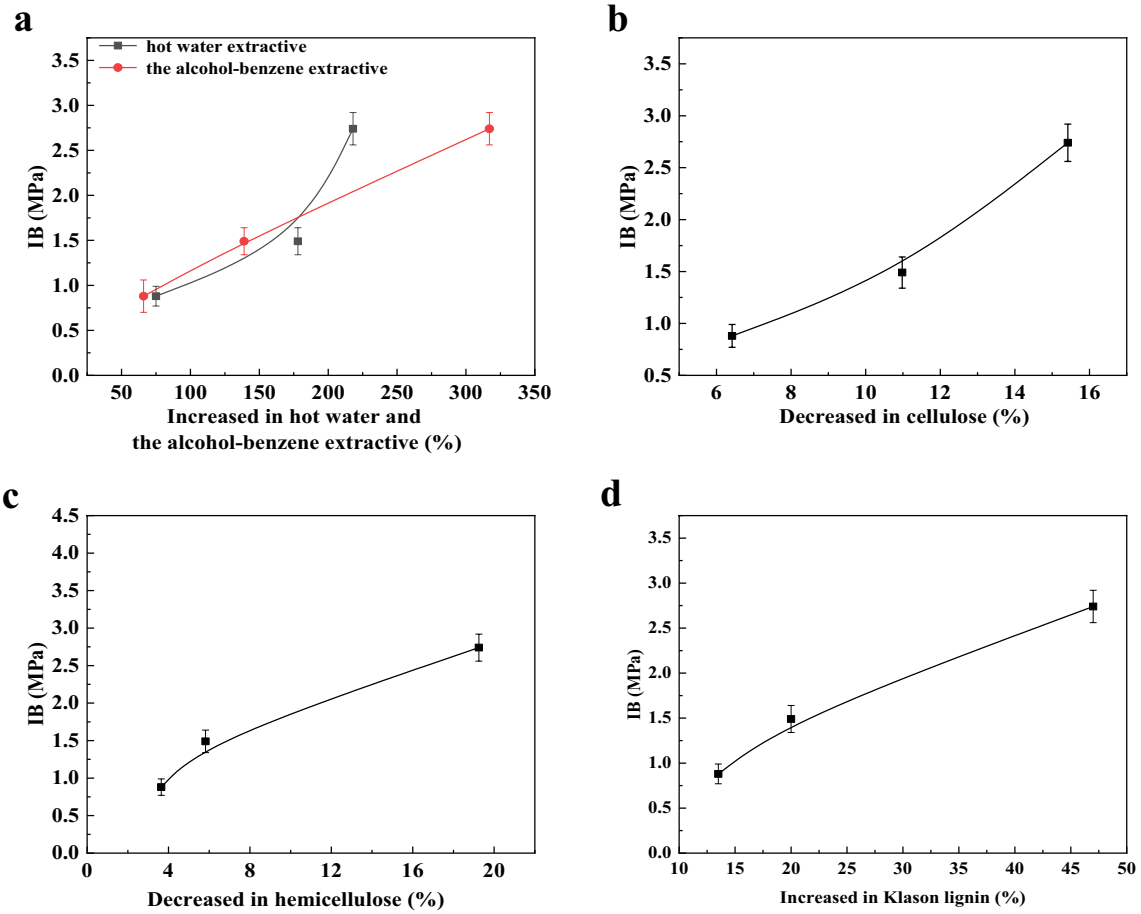

Fig. 7 Correlation between the change in chemical composition and IB strength of binderless board. **a** Effect of an increase in extractives on IB strength of binderless board. **b** Effect of a decrease in cellulose on IB strength of binderless board. **c** Effect of a decrease in hemicellulose on IB strength of binderless board. **d** Effect of an increase in Klason lignin on IB strength of binderless board. Error bars indicate standard deviations

dependent on the chemical behaviors of particles during hot pressing [24]. The change in TS was similar to the correlation between IB strength and the change in chemical composition (Fig. 8a–d). The TS value at 180 ℃ was 42.6%, and it decreased to 7.4% at 220 ℃. The degradation of cellulose and hemicellulose increased the compressibility of the boards, especially the decrease of moisture absorption caused by the change of hemicellulose, which was one of the main reasons for the improvement of dimensional stability [37]. Increasing temperature and time during hot pressing not only improved the dimensional stability of the binderless boards but also greatly improved the mechanical properties.

FTIR analysis of *Broussonetia papyrifera* raw materials and binderless boards

Figure 9 shows the infrared spectra of the raw materials and binderless boards manufactured at 180 ℃, 200 ℃ and 220 ℃. However, no remarkable peak change was observed. The 3348 cm^{-1} bands, were derived from O–H in cellulose, hemicellulose and lignin [27]. The peaks at 1622 cm^{-1} were from the C=C of aromatic ring [38], while the peaks at 1320 cm^{-1} were from guaiacyl and syringyl nuclei of lignin [27]. The peak intensity of binderless boards at both of 1622 cm^{-1} and 1320 cm^{-1} were higher than that of raw material. During hot pressing, cellulose and hemicelluloses hydrolyzed and produced some sugar degradation products which might undergo further reaction to produce aromatic substance (intermediate product of pseudo-lignin) [30]. As a result, the peak intensity at 1622 cm^{-1} of binderless boards strengthen. Newly formed aromatic substance containing active functional groups might further react with degradation substances to form macromolecule. This substance, which has three-dimensional structure and has functional groups similar to lignin, is normally termed pseudo-lignin [30, 31]. The pseudo-lignin contains

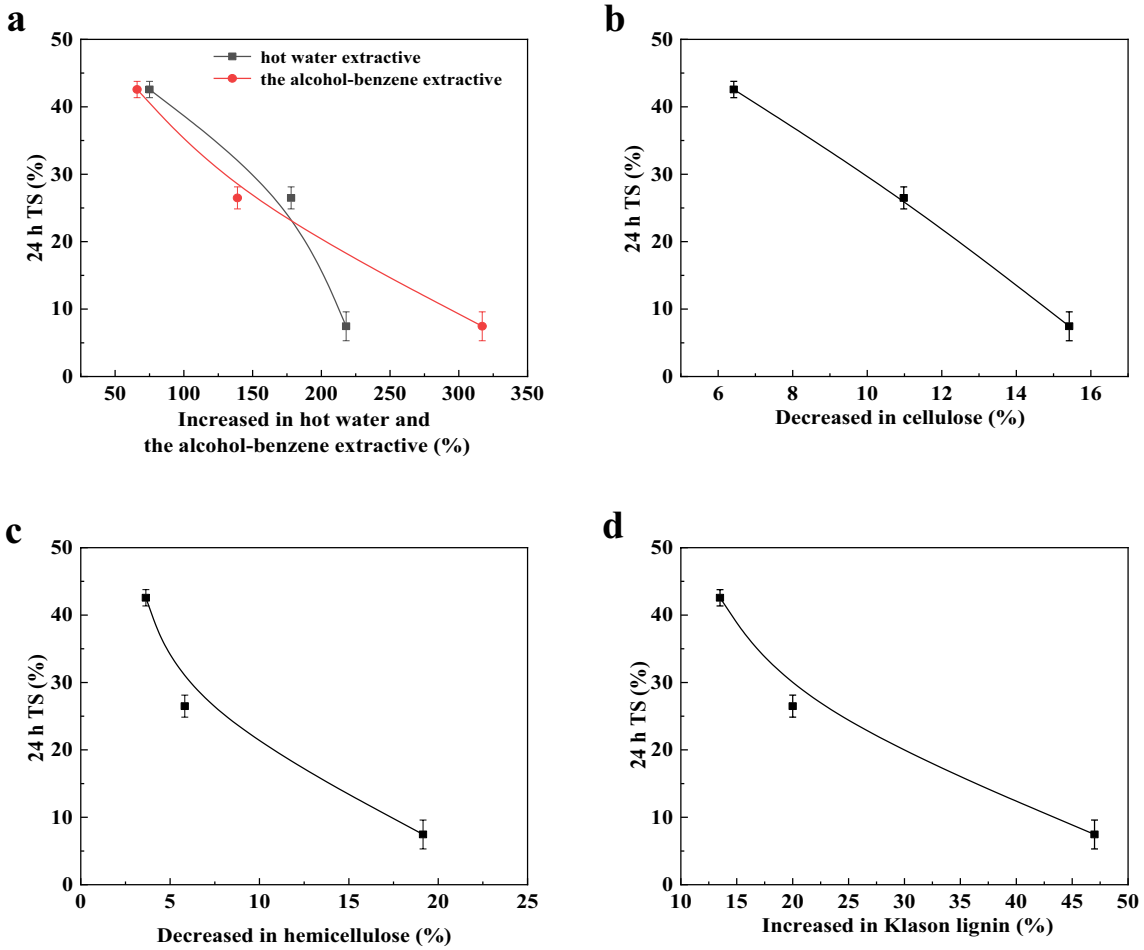

Fig. 8 Correlation between the change of chemical composition and TS of binderless board. **a** Effect of an increase in extractive on 24 h TS of binderless board. **b** Effect of a decrease in cellulose on 24 h TS of binderless board. **c** Effect of a decrease in hemicellulose on 24 h TS of binderless board. **d** Effect of an increase in Klason lignin on 24 h TS of binderless board. Error bars indicate standard deviations

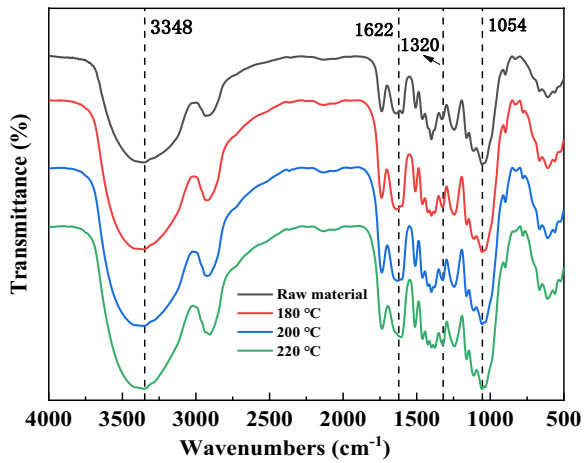

Fig. 9 FTIR of raw materials and binderless boards at different temperatures of *Broussonetia papyrifera*

structures that resemble Guaiacyl and syringyl units of lignin and thus resulted in relatively strong absorption peak at 1320 cm^{-1} for binderless boards, especially the board manufactured at 220 ℃. The pseudo-lignin is a complex polymer formed by polymerization reaction. The formation of pseudo-lignin during hot pressing might contribute to the adhesion among particles and thus improving the properties of binderless board. The peaks at 1054 cm^{-1} were from C–O–C [27]. The band intensity for binderless boards were higher than that of raw material which indicated more C–O–C was formed due to the dehydration of hydroxyl groups, and improving the bonding of binderless board.

XRD analysis of cellulose crystallinity of Broussonetia papyrifera and binderless boards

The X-ray diffraction pattern of raw material and binderless particleboards at different pressing temperatures is

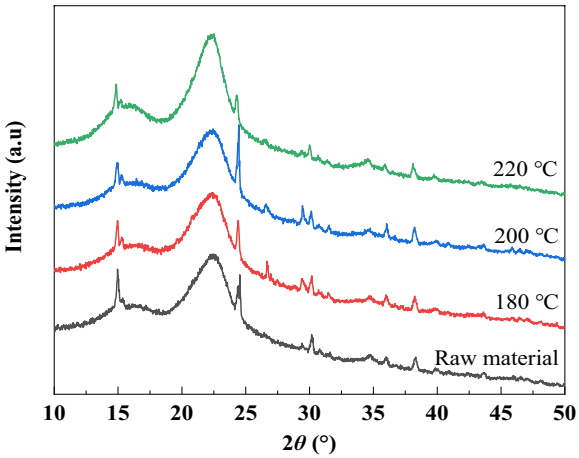

Fig. 10 XRD of raw material and binderless boards of *Broussonetia papyrifera* at different temperatures

Table 5 Crystallinity change of cellulose from different samples

Sample	Crystallinity (%)
Raw material	37.89
180 ℃ board	40.02
200 ℃ board	41.32
220 ℃ board	44.35

shown in Fig. 10. The diffraction pattern exhibited a sharp high peak at 23° and two weaker peaks, respectively, at 16° and 24°. The peak positions of the raw materials and boards were basically similar, indicating hot pressing had little effect on the crystal layer distance. However, the diffracted intensity of all samples changed significantly. The diffracted intensity increased with increasing pressing temperature, which illustrated that temperature had a great influence on the non-crystalline region of *Broussonetia papyrifera* cellulose.

The crystallinity of raw material and binderless boards at different pressing temperatures are shown in Table 5, the relative crystallinity increased with increasing temperature. Compared with *Broussonetia papyrifera* raw material (37.89%), the relative crystallinity of binderless boards at 180 ℃ and 200 ℃ increased to 40.02% and 41.32%, respectively, and it continually rose to 44.35% at 220 ℃. When the temperature increased, the water molecules entered the amorphous region which made hydroxyl groups of cellulose get closer, resulting in the rearrangement of cellulose molecular chains. The crystalline region, as well as the relative crystallinity of cellulose thus increased [27, 39]. In general, the strength and dimensional stability of wood-based material increased with increasing crystallinity. Figure 11 shows a correlation between the change in the relative crystallinity and the performance of binderless board. With the increase of cellulose crystallinity, the MOR, MOE, IB values of binderless boards were increased by 40%, 83% and 211%, respectively, and 24 h TS were decreased by 82%. Higher cellulose relative crystallinity of the binderless boards contributed to better board properties.

Effects of *Broussonetia papyrifera* latex on bonding performance

FTIR analysis of Broussonetia papyrifera latex

The components in *Broussonetia papyrifera* latex mainly contain starch grains, polysaccharides, terpenes, phenols, flavonoids, organic acids and fatty acids [10].

Figure 12 shows the infrared spectra of the *Broussonetia papyrifera* latex sample. The 3393 cm^{-1} bands, might derived from the O–H in polysaccharides, starch grains, flavonoids and phenolic compounds [10, 27], the peaks were large and wide, which indicated there were a large number of O–H on that components. Polysaccharides had a multi-hydroxyl structure and showed hydrophilicity and high sticky. The peaks near 2920 cm^{-1} were mainly the C–H from organic acids and fatty acids. The peaks at 1641 cm^{-1} from C=C were aromatic compounds in terpenes. The high-intensity peak at 1382 cm^{-1} was an isolated methyl group in flavonoids. Sugars in wood can react with hemiacetal hydroxyl groups in terpenes to form glycosides. The bonding was thus strengthened. Polysaccharides and starch grains were beneficial to the self-bonding of particles [10, 40]. *Broussonetia papyrifera* latex contains organic acids, which can promote the degradation of cellulose and hemicellulose in raw material to generate more aldehyde hydrolysates and make the particles more compact in bonding [39, 40].

The bonding strength of Broussonetia papyrifera latex

Poplar veneers were bonded together using *Broussonetia papyrifera* latex as the adhesive and the bonding strength was tested. The maximum and average values of the bonding strength were 0.6 MPa and 0.4 MPa. However, the control test specimens without *Broussonetia papyrifera* latex were delaminated after being placed for a period of time. The *Broussonetia papyrifera* latex was not only sticky but also could bond the veneers together as well and showed a bonding strength. The cured *Broussonetia papyrifera* latex did not dissolve after being soaked in water for 24 h. The latex contained starch, polysaccharide and organic acid, which were conducive to bonding the particles during hot pressing [40]. Therefore, it can be concluded that *Broussonetia papyrifera* latex was favorable for the self-bonding of binderless boards.

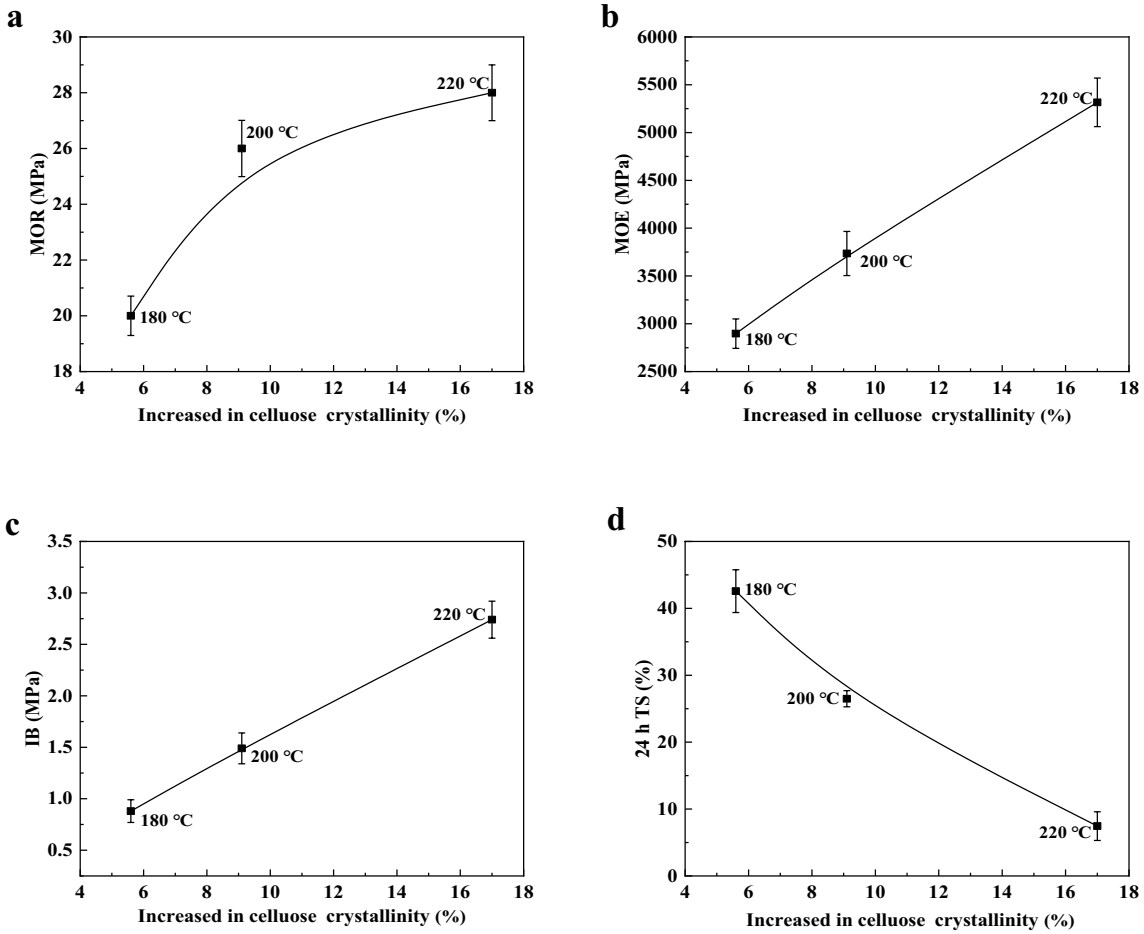

Fig. 11 Correlation between the change in the cellulose crystallinity and properties of binderless board. **a** Effect of an increase in the cellulose crystallinity on MOR of binderless board. **b** Effect of an increase in the cellulose crystallinity on MOE of binderless board. **c** Effect of an increase in the cellulose crystallinity on IB strength of binderless board. **d** Effect of an increase in the cellulose crystallinity on 24 h TS of binderless board. Error bars indicate standard deviations

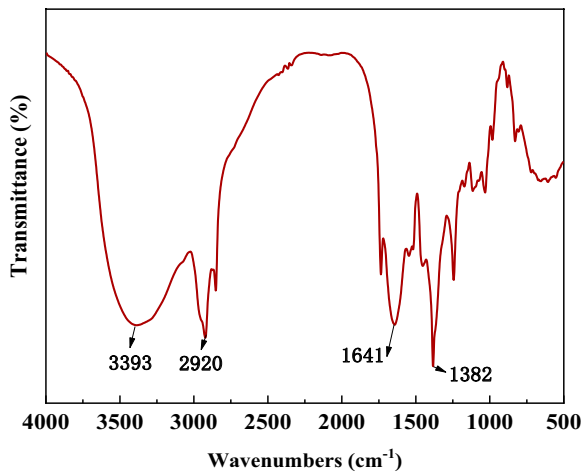

Fig. 12 FTIR of the *Broussonetia papyrifera* latex

Conclusions

High-performance binderless particleboards were successfully prepared by hot pressing from *Broussonetia papyrifera* trunk. The mechanical properties (MOR, MOE, IB) and dimensional stability (TS) of the binderless board can be significantly improved by optimization of the manufacturing conditions, including reducing the size of particles, prolonging pressing time, rising temperature and increasing board density. However, the MOR and MOE of the binderless board obtained at too high temperature for too long pressing time (220 ℃/45 min) deteriorated. The optimal board met the performance requirements of heavy-duty type-P4 particleboard of Chinese national standard GB/T 4897-2015 [17]. The cellulose and hemicellulose content in *Broussonetia papyrifera* decreased during board manufacturing. During hot pressing, the formation of pseudo-lignin and the increased C–O–C and cellulose crystallinity of the

boards contributed to high quality of the binderless boards. It was also found that *Broussonetia papyrifera* latex had a certain bonding ability and played a positive role in promoting the self-bonding of the *Broussonetia papyrifera* particle. The *Broussonetia papyrifera* binderless particleboard recorded an IB value of as high as 2.98 MPa value and a low 24 h TS value of only 5.0%.

Abbreviations

MOR	Modulus of rupture
MOE	Modulus of elasticity
IB	Internal bonding
TS	Thickness swelling
FTIR	Fourier-transform infrared spectrum
XRD	X-ray diffraction

Acknowledgements
Thanks are due to Prof. Tiehua Li (College of Forestry, CSUFT) for providing *Broussonetia papyrifera* wood and to Dr. Min Zhang (College of Materials Science and Engineering, CSUFT) for valuable discussion. This work was supported by Hunan Provincial Innovation Foundation for Postgraduate Grant NO. CX20220731.

Author contributions
MC: carried out the tests, data analysis and figure preparation and was a major contributor in writing the manuscript. SZ and JW: supported numerical analysis, board preparation and properties test. JX: designed the research, draft the manuscript, prepared figures and supervision. All authors read and approved the final manuscript.

Funding
This work was supported by Hunan Provincial Innovation Foundation for Postgraduate Grant NO. CX20220731.

Availability of data and materials
The data sets used or analyzed during the current study are available from the corresponding author upon reasonable request.

Declarations

Competing interests
The authors declare that they have no competing interests.

Received: 26 November 2022 Accepted: 20 March 2023
Published online: 30 March 2023

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