Study on Hot–pressing Technology of Chitosanmodified Starch Adhesive Film

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In order to optimize its production technology, response surface methodology (RSM) was used to optimize the hot-pressing process during the preparation of chitosan-modified starch adhesive film. Ranges of hot-pressing temperature, hot-pressing time, and adhesive consumption were selected based on single-factor tests. A quadratic regression model of the bonding strength was obtained by fitting the response value of the bonding strength of the plywood. The results showed that the adhesive consumption and the hot-pressing temperature had a significant effect on the bonding strength of the adhesive. Also, the interaction between the temperature, time, and adhesive consumption were significant. The optimum hot-pressing process parameters for chitosan-modified starch adhesive film were a hot-pressing temperature of 145.2 °C, a hot-pressing time of 182.7 s, and an adhesive consumption of 239.3 g/m². The predicted values of the quadratic regression model fit well with the actual values of the stability test.

Keywords: Starch; Chitosan; Hot-pressing process; Bond strength

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INTRODUCTION

Starch ($C_6H_{10}O_5$)_n) is a renewable resource widely existing in seeds, roots, stems and tissues of many plants (Mischnick and Momcilovic 2010). It is widely used in textile, paper-making, packaging, and other industrial products because of its wide source, low price, biodegradable, environmental friendly character, and other numerous advantages (Lefang 2002). However, the application of starch adhesive in wood industry is limited by its brittleness, poor water resistance, and storage problems (Wang *et al.* 2012a). Chitosan (CTS, (1,4) -2-deoxy-beta-*d*-glucan) is the product of alkalinized chitin, which is widely available throughout the world as a byproduct of shellfish processing (Pillai *et al.* 2009). The dimer unit of chitosan molecule are N-acetyl-D-glucosamine and D-glucosamine linked by a β -(1-4) glycoside bond. Chitosan is rich in hydroxyl and amino groups, and it easily forms hydrogen bonds with other electronegative molecular groups. The C₂-NH₂, C₃-OH, and C₆-OH in its molecular chain can form intramolecular and intermolecular hydrogen bonds (Rinaudo 2006), so chitosan has good film-forming properties. It also has good antimicrobial, biodegradability, and biocompatibility (Kumar *et al.* 2004). Therefore, adding chitosan to starch adhesive has excellent advantages.

Plywood is one of the most important wood-based panels in the world. It has many advantages and is widely used in various furniture products, including door and window frames, flooring, automobile interior panel, *etc.* (Fang *et al.* 2013). Hot-pressing is the key process in plywood production. In order to optimize hot-pressing conditions, reduce energy

consumption and production costs, it is essential to understand the heat and mass transfer behavior of plywood during hot-pressing (Liu *et al.* 2013).

Researchers have found that if the hot-pressing temperature was high enough, the adhesives can be completely cured. However, further increase of hot-pressing temperature may not be conducive to the bonding of wood and adhesives. In fact, if the hot-pressing temperature is too high, wood and adhesives may degrade to a certain extent, thus reducing the strength (Gu et al. 2013). Short hot-pressing time will make it difficult for the internal groups of the adhesives to complete the reaction, and long hot-pressing time will also easily cause the decomposition of the adhesives and decrease the bonding properties (Chen et al. 2012). Also, long-time hot-pressing will increase the compression ratio of wood and reduce the production efficiency of plywood. It also reduces the moisture content of the panel, and the hot-pressed panel is easy to warp, which is unacceptable in the plywood manufacturing industry (Jing et al. 2015). Under different hot-pressing pressure levels, the bonding strength of the board has obvious differences. Under suitable hot-pressing conditions, the adhesive can penetrate into the wood further to form a nail structure and, mechanical bonding force with veneer to improve the strength of the plywood. However, excessive hot-pressing pressure can easily lead to over-compression of veneers. Therefore, on the premise that the bonding strength of plywood must be up to standard, a lower hot pressing pressure can be chosen to reduce the extent of wood compression (Chen et al. 2012).

It is generally believed that the shear strength of plywood is better with more sizing. This is because the increase in the amount of adhesives increases the bonding area between wood and adhesives. The more uniform the distribution of adhesives, the stronger the adhesion of adhesives (Wang *et al.* 2012b). However, excessive sizing may lead to excessive thickness of the adhesive layer, resulting in a reduction in shear strength (Fang *et al.* 2013). Presently, the main adhesives used for plywood are formaldehyde adhesives, such as phenolic (PF) and urea formaldehyde (UF) resins. Although these adhesives have the advantages of good adhesion and mature technology, the emission of formaldehyde during the production and use of plywood has caused great harm to human health (Sellers 2001). Formaldehyde can damage the respiratory system, eyes, and nervous system, and even lead to cancer and leukemia. The International Agency for Research on Cancer has concluded that formaldehyde is a human carcinogen (Lyon (France) International Agency for Research on Cancer 1980). The California Air Resources Board (CARB) passed a regulation in April 2007 setting a ceiling on formaldehyde emissions from wood products used and sold in California (Gu *et al.* 2013).

Nowadays, plywood factories mainly apply glue by coating adhesive rolls on veneers. However, this process requires high fluidity of adhesives, and it may cause uneven sizing and increase of veneer moisture content in the process of roll coating. Therefore, some researchers use thermoplastic film to solve the problems in the manufacturing of plywood. Fang *et al.* (2013) developed non-formaldehyde plywood with low density polyethylene film. The plywood produced fully meets the requirements of indoor plywood application (Summer 1993; Liang *et al.* 2009; Rowell 2012; Fang *et al.* 2013). However, such resins are not biodegradable, leading to environmental pollution. It would be an interesting attempt to produce a degradable, non-toxic, formaldehyde-free thermosetting film.

In this study, corn starch was selected as the main material of adhesive, and the formaldehyde-free chitosan-modified starch adhesive film was prepared in the laboratory (Lawton 1996; Wang *et al.* 2012b). The hot-pressing process of the film was improved to prepare three-layer *Pinus massoniana* plywood under different conditions. The effects of

hot-pressing temperature, time, pressure, and sizing amount on the bonding strength of plywood were studied. The feasibility of combining starch-based chitosan film with polar wood veneer as a new type of adhesive was explored. This does not only improve the plywood production technologies used in the wood industries, but also make some efforts to replace formaldehyde based plywood.

EXPERIMENTAL

Materials

Pinus massoniana veneer (0.8 to 1 mm thick, 300-mm-long \times 300-mm-wide) with a moisture content of 8 to 10% was collected from the Jingxi plywood factory (Minghou, China). Anhydrous sodium sulfite was obtained from Guoyao Chemical Reagent Co. (Shanghai, China). Specification 1799 polyvinyl alcohol (PVA) was used. Corn starch comes from Guangzhou Guangzuan Chemical Co., Ltd. Chitosan with a deacetylation degree of 80.0 to 95.0 was procured from China Pharmaceutical Group Chemical Reagent Co. (Shanghai, China).

Instrument and Equipment

The specimens were pressed using a model BY302X2/15 test press (Suzhou Machinery Manufacturing Cooperation, Suzhou, China). An electrothermal constant temperature blast dryer (DHG-9223A; Anhui Jingting Indoor Environmental Quality Testing Co., Xuancheng, China), a microcomputer controlled electronic universal testing machine (CWT6104; Shenzhen Xinsisi Material Testing Co., Shenzhen, China), a digital pH meter (pHS-2; Yidian, Shaanxi, China), and a circulating water vacuum pump (SHZ-III; KD, Henan, China) were used to perform specimen preparation and testing.





Analytical Methods

The plywood was placed at 20 ± 2 °C and $65 \pm 5\%$ humidity for 24 h, and its bonding strength was tested according to the standards GB/T 9846.1-2004 (2004) and GB/T 17657-2013 (2013). The specimen was sawn according to the standard depicted in Fig. 1. The bonding strength of the specimen was determined by the arithmetic average of three tests. The two ends of the specimen were clamped in a pair of movable fixtures of a Universal Capability Testing Machine (CMT6014, Shenzhen, China) to form a straight line. The center of the specimen passed through the axis of the movable fixture of the testing machine. The specimen was loaded to failure at the same speed, and the maximum damage load was recorded. The bonding strength of the specimens were calculated using Eq. 1,

$$X_A = \frac{P}{A \times B} \tag{1}$$

where X_A is the bonding strength of the specimen (MPa), P is the maximum failure load (N), A is the width of the shear section of the specimen (mm), and B is the length of the shear section of the specimen (mm).

Adhesive Film Preparation

In order to prepare the chitosan-modified starch adhesive film for this experiment, 15 g of starch and 20 g of water were added into the flask with three necks. The aqueous solution was stirred continuously with a regulated temperature up to 50 °C. The pH of the solution was adjusted to 10 by adding 1.5 g of sodium hypochlorite and stirred for 3 h. Then, 0.045 g sodium sulfite was added, and the reaction lasted for 10 min. The reaction was stabilized by adding 0.45 g of PVA, 3.75 g of chitosan, and stirring for 30 min. Chitosan-modified starch adhesive was gelatinized at 70 °C for 15 min. The adhesive was degassed at a vacuum pressure of 0.098 MPa and coated on the steel plate. The adhesive was dried at 50 °C for 1 h, then the film was taken out and stored.

Plywood Sample Preparation

Three pieces of dry *P. massoniana* veneers were used to prepare three layers of plywood using starch-based chitosan adhesive film with the same solid content according to the adhesive consumption designed in the test scheme (Fig. 2). The binder was diluted with 25% water before hot-pressing. The film was then placed on both sides of the veneer core layer and arranged according to the grain direction. After hot-pressing, the plywood was packed and shelved at room temperature for 24 h before sawing and testing.



Fig. 2. Schematic diagram of the adhesive film and film sizing

Experimental Design and Statistical Analysis

In the single factor test, the fixed levels of hot-pressing temperature, hot-pressing pressure, hot-pressing time and sizing amount were defined as $150 \,^{\circ}$ C, $1.2 \,\text{MPa}$, $180 \,\text{s}$, and $300 \,\text{g/m}^2$ (double side), respectively. When one of the factors was a variable, the other three factors were fixed. The influence of hot-pressing pressure on the bonding strength of starch-based chitosan plywood was not sensitive during the single factor test (Fig. 3), so $1.2 \,\text{MPa}$ was used to save energy and reduce the compression of the plywood.

Based on the single factor test, the response surface analysis method of three factors and three levels was determined according to the design principle of the Box-Behnken test (Afshar and Baniasadi 2017). The moisture content of the veneer was 10%. The hotpressing temperature, time, and adhesive consumption, defined as X_1 , X_2 , and X_3 , respectively, were the key factors affecting the bonding strength. The horizontal codes of X_1 , X_2 , and X_3 were expressed as -1, 0, and 1, respectively. Finally, the change of the response value of the bonding strength (Y) was investigated under the three-factor and three-level test designs. Based on the single factor test and considering the curing characteristics and sizing difficulties of the starch-based chitosan adhesives, the suitable range of hot-pressing temperature, time, and adhesive consumption was determined to be 120 to 180 °C, 150 to 210 s, and 150 to 350 g/m² (double side), respectively. The influencing factors and coding level of each test are shown in Table 1.



Fig. 3. (a) Effect of hot-pressing temperature on bonding strength; (b) Effect of hot-pressing pressure on bonding strength; (c) Effect of hot-pressing time on bonding strength; (d) Effect of sizing quantity on bonding strength

Factor				
Level	Hot-Pressing Temperature (<i>X</i> 1)	Hot-Pressing Time (X ₂)	Adhesive Consumption (<i>X</i> ₃)	
	(3°)	(s)	(g/m²)	
-1	120	150	150	
0	150	180	250	
1	180	210	350	

Table 1. Factors and Levels of Design for Response Surface Experiment

RESULTS AND DISCUSSIONS

Diagnostic Checking of the Fitted Model

The hot-pressing process of chitosan-modified starch film was optimized and modeled by 8.0.6 software program design expert (Minneapolis Stat Ease Company, USA). Box-Behnken is used to simulate RSM. Three independent variables were selected in Box-Behnken design, namely; hot-pressing temperature, hot-pressing time and, adhesive consumption (Wu *et al.* 2017).

According to Fig. 3, the corresponding relationship between the response value Y (bonding strength) and variable *X* were designed. The results are shown in Table 2.

	Factor			
Number	Hot-Pressing Temperature (<i>X</i> ₁)	Hot-Pressing Time (X ₂)	Adhesive Consumption (X_3)	Bonding Strength (Y)
	(°C)	(s)	(g/m²)	(MPa)
1	180	180	350	1.2410
2	150	180	250	2.0350
3	150	180	250	2.2715
4	120	180	350	1.3410
5	150	180	250	2.2150
6	150	210	150	1.6065
7	120	180	150	1.6095
8	150	150	150	1.4360
9	180	210	250	1.6540
10	150	150	350	1.2450
11	150	180	250	2.2230
12	150	210	350	1.3320
13	180	180	150	1.4055
14	180	150	250	1.6575
15	150	180	250	2.1290
16	120	150	250	1.8140
17	120	210	250	1.8380

Table 2. Experimental Designs and Results

A quadratic regression model of plywood bonding strength (Y) and hot-pressing temperature (X_1), hot-pressing time (X_2), and adhesive consumption (X_3) was obtained by regression fitting the results of 17 test schemes established by three factors and three levels (Table 2). The quadratic regression model is shown in Eq. 2.

$$Y = 2.17 - 0.081X_1 + 0.035X_2 - 0.11X_3 - 0.006875X_1X_2 + 0.026X_1X_3 - 0.021X_2X_3 - 0.22X_1^2 - 0.21X_2^2 - 0.56X_3^2$$
(2)

The model P < 0.0001 indicates that the quadratic model was highly significant, while the missing item P = 0.8385 (>0.05) was not significant (Table 3). This shows that the quadratic equation fit the test data well, the error of the test was small, and the response between variables and response values could be better. Therefore, Eq. 2 could be used to analyze and predict the bonding strength of starch-based chitosan plywood.

Several conclusions can be drawn from the P values of the variables (X) in Table 4. The hot-pressing temperature (X_1) has a significant effect on the bonding strength of starchbased chitosan plywood (p < 0.05), and adhesive consumption (X_3) has a significant effect on the bonding strength of starch-based chitosan plywood (p < 0.01). Therefore, when pressing starch-based chitosan plywood, the influence of hot-pressing temperature and adhesive consumption should be considered. The hot-pressing time can be flexibly selected according to the actual situation.

Source of Variation	Sum of Square	Degree of Freedom	Mean Square	F Value	P Value
Model	2.01	9	0.22	36.99	< 0.0001*
Residual	0.042	7	6.036E-003		
Lack of Fit	7.320E-003	3	2.440E-003	0.28	0.8385
Pure Error	0.035	4	8.733E-003		
Sum	2.05	16			

Table 3. Analysis of Variance for the Established Regression Model

Note:* indicates a significant difference of 0.05,** indicates an extremely significant difference of 0.01.

Source of Variation	Sum of Square	Degree of Freedom	Mean Square	F Value	P Value
X 1	0.052	1	0.052	8.60	0.0219*
X2	9.661E-003	1	9.661E-003	1.60	0.2463
X3	0.10	1	0.10	16.72	0.0046**
X ₁ X ₂	1.891E-004	1	1.891E-004	0.031	0.8645
<i>X</i> ₁ <i>X</i> ₃	2.704E-003	1	2.704E-003	0.45	0.5247
X ₂ X ₃	1.743E-003	1	1.743E-003	0.29	0.6077
X ₁ ²	0.20	1	0.20	33.68	0.0007
X2 ²	0.19	1	0.19	31.98	0.0008
X3 ²	1.30	1	1.30	215.43	< 0.0001

Table 4. Significance Test of Bonding Strength Regression Coefficients

Note: * indicates a significant difference of 0.05, ** indicates an extremely significant difference of 0.01.

Optimization of Bonding Strength

The effects of hot-pressing temperature (X_1) , hot-pressing time (X_2) , and adhesive consumption (X_3) on the bonding strength (Y) are shown in Figs. 4 through 6. The threedimensional (3D) response surface graph and contour map directly reflect the interaction among hot-pressing temperature, hot-pressing time, and adhesive consumption, and the influence of these factors on the response value.

The response surface and contour plot of the hot-pressing temperature and time when the adhesive consumption was fixed at 250 g/m^2 is shown in Fig. 4. Figure 4 revealed the effect of the hot-pressing temperature and time and their interaction on the bonding strength. Precisely, Fig. 4a shows that when the hot-pressing temperature was constant, the bonding strength increased first and then decreased as the hot-pressing time increased. The highest bonding strength occurred at approximately 182 s. It takes a certain time to transfer heat from the surface to the core board, but prolonged hot-pressing time will not only affect the efficiency of hot-pressing, but also affect the effect of curing. The long hot-pressing time led to brittle plywood, which reduced the bonding strength of the plywood. When the hot-pressing time was fixed, the bonding strength initially increased and then decreased with as the hot-pressing temperature increased. The maximum value was obtained at approximately 145 °C. This may have been a result of the acceleration of molecular thermal movement and water evaporation during hot-pressing, which promoted the formation of hydrogen bonding forces. However, excessive temperature may lead to degradation of hemicellulose (slight degradation at 120 to 160 °C) and lignin in wood, resulting in a decrease in bonding strength (Leppänen et al. 2011; Kim et al. 2016). The higher the temperature, the more the degradation. Lignin is degraded continuously in the range of 100 to 700 °C, and the highest degradation rate will occur at 340 °C.

Therefore, it is particularly important to select the appropriate temperature during hot-pressing. It was found that the bonding strength was the highest and the quality of plywood was the best when the hot-pressing temperature was fixed at 145 °C and the hot-pressing time was 182 s. From the ellipse of the medium height line in Fig. 4b, the interaction between the hot-pressing temperature and the hot-pressing time is significant.

The response surface diagram and contour diagram of temperature and adhesive consumption when the hot-pressing time was fixed at the level of 180 s are shown in Fig. 5. When the hot-pressing temperature was constant, the bonding strength increased first and then decreased with the increased adhesive consumption. Furthermore, Fig. 5a showed that when the adhesive consumption was less than 240 g/m^2 , a complete and continuous adhesive layer could not be formed on the slab plane due to the small adhesive consumption, which resulted in low bonding strength. When the adhesive consumption exceeded 240 g/m², the bonding strength decreased as the adhesive consumption increased, which was as a result of the inability of the excessive adhesive consumption to penetrate the plywood. This thickened the adhesive layer, increasing the stress produced during the curing of the adhesive layer. Like the previous observation in the use of small adhesive, the bonding strength also decreased. In addition, the increased adhesive consumption will also result in wasted resources. The highest bonding strength was observed when the hotpressing temperature and adhesive consumption was about 145 °C and 239 g/m², respectively. As seen from the ellipse of the contour in Fig. 5b, the interaction between hot-pressing temperature and adhesive consumption is significant.

The response surface and contour map of adhesive consumption and hot-pressing time when the hot-pressing temperature is at zero level and 150 $^{\circ}$ C are shown in Fig. 6. Figure 6a showed that the bonding strength increased first and then decreased with the

increase in the adhesive consumption when the hot-pressing time was fixed. The bonding strength increased first and then decreased with the increase in hot-pressing time when the adhesive consumption was constant. This indicates that the excessive hot-pressing time and excessive adhesive consumption had a negative effect on the bonding strength, which is determined by the properties of the starch-based chitosan adhesive itself. Excessive hot-pressing time and adhesive consumption will lead to excessive curing of the adhesive layer. The bonding between veneers could not be tightly bonded, leading to an increase in the stress caused by curing of the adhesive layer. The curing of the adhesive layer was incomplete, and the amount of the adhesive was insufficient when the hot-pressing time was too short and at small adhesive consumption, which led to the discontinuity of the adhesive layer and the reduction in the bonding strength. Furthermore, highest bonding strength and best plywood was produced when the hot-pressing time and the adhesive consumption was about 182 s and 239 g/m², respectively. The significant interaction between hot pressing time and adhesive consumption can be observed from the ellipse of the contour in Fig. 6b.

Based on the above analysis of the interaction among the three factors, and without exceeding the range of conditions selected by each factor, the optimum technological parameters of each factor were obtained by response surface analysis software. The bonding strength reached its maximum value at a hot-pressing temperature of 145.2 °C, a hot-pressing time of 182.7 s, and a sizing amount of 239.3 g/m². The prediction value of bonding strength of excellent plywood is 2.19 MPa.



Fig. 4. (a) 3D response surface plots and (b) contour lines showing the interaction effects of the hot-pressing temperature and pressing time on the bonding strength



Fig. 5. (a) 3D response surface plots and (b) contour lines showing the interaction effects of hotpressing temperature and adhesive consumption on bonding strength



Fig. 6. (a) 3D response surface plots and (b) contour lines showing the interaction effects of hotpressing time and adhesive consumption on bonding strength

Validation Method of the Model

Three layers of plywood were prepared according to the optimum hot-pressing process parameters obtained by the response surface tests. It was replicated five times to determine the stability of hot-pressing process by the bonding strength of the plywood. As shown in Table 5, the average dry strength of plywood pressed with starch-based chitosan adhesive was 2.23 MPa, which is not significantly different from the theoretical predicted value of 2.19 MPa (the relative error is only 1.83%). This means that the quadratic regression model established by response surface fits well with the actual hot-pressing process.

Number	Moisture Content (%)	Bond Strength of Dry Form (MPa)
1	9.3	2.17
2	9.4	2.28
3	8.8	2.13
4	8.7	2.41
5	8	2.16
Average	8.84	2.23

Table 5. Results of The Model Verification Method

Advantages of Starch-based Chitosan Film

Three main advantages of the chitosan-modified starch films were as follows, based on the present work:

- a) Ease of application: The use of starch-based chitosan film enable uniform veneer surface coating which enhance adherend bonding.
- b) Good shelf-life: Due to its low moisture content (1%), it can be used when stored at room temperature and stay dry for half a year. Starch adhesives without chitosan would be inhabited by mildew after storing for one week at room temperature.
- c) Ease of transportation: starch-based chitosan film is a thermosetting solid, solventfree, easy to load and unload.

CONCLUSIONS

- 1. Response surface analysis showed that the optimum hot-pressing conditions for chitosan-modified starch plywood were 145.2 °C, 182.7 s, and 239.3 g/m² adhesive consumption. The predicted value of the optimal plywood bonding strength was 2.19 MPa.
- 2. The model verification demonstrated that the actual bonding strength of plywood pressed by the optimal hot-pressing process parameters were slightly higher than the predicted value, and the relative error was only 1.83%. This shows that the fitting equation matched well with the actual situation.
- 3. Statistical analysis showed that the effect of adhesive consumption on bonding strength was very significant. The effect of hot-pressing temperature on bonding strength was also significant. In addition, there was significant interaction between hot-pressing temperature, hot-pressing time, and the amount of adhesive consumed. The established quadratic model has practical significance, and the optimal process parameters obtained by the response surface method have practical reference value.

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