Investigation of a New Natural Particleboard Adhesive Composed of Tannin and Sucrose. 2. Effect of Pressing Temperature and Time on Board Properties, and Characterization of Adhesive

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In a previous study by the authors, particleboard was manufactured using a new natural adhesive composed of tannin and sucrose. The optimal ratio between tannin and sucrose was 25/75, and the suitable resin content was 30 to 40 wt%. In this study, the effects of hot pressing temperature and hot pressing time on board properties were investigated. The optimal values for the hot pressing temperature and hot pressing time were found to be 220 °C and 10 min, respectively. When the particleboard was made under these optimum conditions, the physical properties of the particleboard bonded with tannin and sucrose met the requirement of the JIS A 5908 type 18 standard (2003). Thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), insoluble matter, and Fourier transform infrared spectroscopy (FT-IR) tests were carried out. The results of TGA and DSC measurements showed that the weight loss and endothermic reaction of the adhesive composed of tannin and sucrose at a ratio of 25/75 occurred at 204 and 215 °C, respectively. When the adhesive was heated at 220 °C for longer than 10 min, the level of insoluble matter was higher than 70 wt%. FT-IR analysis showed the existence of a furan ring, a carbonyl group, and dimethylene ether bridges in the cured adhesives before and after the boiling treatment. When the heating time was longer than 10 min, no further change of chemical structure was observed.

Keywords: Wood adhesive; Tannin; Sucrose; Particleboard

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INTRODUCTION

Wood-based materials are widely used. These materials are considered to be environmentally friendly due to the fact that they incorporate lignocellulosic biomass, which is a renewable resource, as a raw material. Usually, synthetic resin adhesives derived from fossil resources are used in wood-based materials to obtain satisfactory bond performance (Yang *et al.* 2006; Mansouri *et al.* 2007; Krug 2010; Hoong *et al.* 2011). However, as the shortage of fossil resources and the deterioration of global environment have been serious problems, it is necessary to develop renewable resources instead of fossil resources. Some researchers have investigated wood adhesives made at least partly from bio-based materials such as protein, tannin, and lignin (Trosa and Pizzi 1997; Pizzi 2006; Lei *et al.* 2008; Jang *et al.* 2011). Recently, citric acid and sucrose were used as an adhesive for the particleboard, and sucrose was found to be effective for enhancement of bond performance (Umemura *et al.* 2013). In addition, it was recently found that a mixture of wattle tannin and sucrose can act as an adhesive for wood (Umemura 2013). Wattle tannin is composed of resorcinol A-rings, providing the material with strong nucleophilicity. The phenolic nuclei could react with formaldehyde with high reactivity, yielding a rate of reaction 10 to 50 times higher than the reaction of phenol with formaldehyde (Kim and Kim 2003).

Sucrose is a common disaccharide, which in principle can be prepared by dehydration synthesis from glucose and fructose molecules. During the heating treatment of sucrose, the caramelization reaction takes place, and 5-hydroxymhthylfurfural (5-HMF) is formed as a main product (Lee *et al.* 2011; Jeong *et al.* 2013). Considering the chemical characteristics of tannin and sucrose, it is possible that the polymerization could happen between tannin and sucrose during the heating treatment.

In a previous study by the authors (Zhao and Umemura 2014), particleboard bonded with tannin and sucrose was manufactured. The mechanical properties of particleboard produced with a tannin-to-sucrose ratio of 25/75 and a resin content of 30 to 40 wt% satisfied the requirement of the JIS A 5908 type 18 standard (2003). In the present study, the effects of the hot pressing temperature and hot pressing time on the physical properties of the particleboard were investigated. The relationship between curing process and temperature was also studied by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The insoluble matter of the cured adhesives was measured, and the change of the chemical structure of adhesives cured for different heating times was studied by Fourier transform infrared spectroscopy (FT-IR).

MATERIALS AND METHODS

Materials

Particles

Recycled wood particles obtained from a particleboard company were used as raw materials, and the particles were screened using a sieving device to collect particles in the range between 5.9 and 0.9 mm. The moisture content of the original particles was 3 to 4 wt%. Before the manufacturing of the particleboards, the particles were dried in an oven at 80 $^{\circ}$ C for 12 h until the moisture content at 2 wt%.

Adhesive

Wattle tannin (tannic acid ME) was purchased from Fuji Chemical Industry Co. (Wakayama, Japan), and sucrose was purchased from Nacalai Tesque, Inc. (Kyoto, Japan). Those materials were used without further purification and dried in a vacuum oven at 60 °C for 15 h before using. Based on the previous paper (Zhao and Umemura 2014), tannin and sucrose were mixed with a ratio of 25/75, and then the mixture was dissolved in distilled water with a concentration of the solution at 40 wt%. The solution was used as an adhesive, and the viscosity and pH of the solution at 20 °C were 51.3 mPa·s and 4.76, respectively.

Methods

Manufacture of particleboard

The adhesive solution was sprayed onto particles in a blender. The resin contents were 30 and 40 wt% based on the weight of the oven-dried particles. The sprayed particles were dried at 80 °C for 12 h, and the moisture content after drying was 3 to 6 wt%. Then the particles were mat-formed using a forming box of 300×300 mm. The particleboards

were manufactured in two Groups. In Group 1, hot pressing temperatures of 160, 180, 200, and 220 °C were used, and the hot pressing time was 10 min. In Group 2, the hot pressing temperature was 220 °C, and hot pressing times of 5, 7, 10, and 15 min were used. Both in Groups 1 and 2, one board was manufactured for every condition. The thickness was controlled using a distance bar of 9 mm thickness. The size of the manufactured board was $300 \times 300 \times 9$ mm, and the target density was 0.8 g cm⁻³. The obtained boards were conditioned for 1 week at 20 °C and 60% relative humidity (RH). The details of the manufacturing conditions are shown in Table 1.

Groups	Mixture ratio of tannin and sucrose (wt%)	Resin content (wt%)	Pressing temperature (°C)	Pressing time (min)	Target density (g cm-3)
Group1	25:75	30 40	160	10	0.8
		30 40	180		
		30 40	200		
		30 40	220		
Group2	25:75	30 40	220	5	- 0.8
		30 40		7	
		30 40		10	
		30 40		15	

Table 1. Manufacturing Conditions of Particleboard

Evaluation of Board Properties

The boards were evaluated according to the Japanese Industrial Standard for particleboard (JIS A 5908. 2003). The static three-point bending test was carried out under dry condition on a $200 \times 30 \times 9$ mm sample from each board, and the effective span and loading speed were 150 mm and 10 mm/min, respectively. The modulus of rupture (MOR) and the modulus of elasticity (MOE) were calculated. The internal bond strength (IB) test was performed on a $50 \times 50 \times 9$ mm sample with a loading speed of 2 mm/min, and thickness swelling (TS) after water immersion for 24 h at 20 °C was measured in samples of the same size. Following the TS test, thickness and weight changes under a cyclic accelerated aging treatment (drying at 105 °C for 10 h, warm water immersion at 70 °C for 24 h, drying at 105 °C for 10 h, boiling water immersion for 4 h, and drying at 105 °C for 10 h) were measured. Each experiment was performed in quintuplicate, and the average value and standard deviation were calculated. Statistical significance was considered for p values < 0.5. The MOR, MOE, and IB of the boards shown in the figures are values corrected for target density based on each regression line between actual values and sample densities of the mechanical properties.

Thermal Analysis

Tannin and sucrose were dissolved in distilled water with the ratio of tannin to sucrose at 100/0, 25/75, and 0/100. The concentration of each solution was adjusted to 40

wt%. From every kind of the solution, 20 g solution was taken out and divided in 10 aluminum cups and dried in an oven at 80 °C for 12 h. After that, the samples were pulverized into less than 250- μ m mesh size to obtain uncured adhesive powder. TGA was carried out using a TGA 2050 (TA Instruments Japan). The samples were scanned from room temperature to 400 °C at a rate of 10 °C /min under nitrogen purging. The DSC measurement was conducted using a DSC 2910 (TA Instruments, Japan). The samples were encapsulated in an aluminum pan and scanned from room temperature to 400 °C at a rate of 10 °C /min under nitrogen purging.

Insoluble Matter and FT-IR

Tannin and sucrose were dissolved in distilled water with a ratio of tannin to sucrose of 25/75, and uncured adhesive powder was obtained, in the same manner as was done for the thermal analysis. The uncured adhesive powder was divided into 7 parts. The powder was heated in an oven at 220 °C for 1, 3, 5, 7, 10, 15, and 20 min, respectively. Then 2 g samples were sampled from every part of the cured adhesives, and the samples were then boiled in distilled water for 4 h to obtain the insoluble matter. The boiling treatment for every sample was carried out in triplicate. All of the samples that were obtained from the heat treatments and boiling treatments were vacuum-dried at 60 °C for 15 h.

Infrared spectra were obtained with a Fourier transform infrared spectrophotometer (FT/IR-4200, JAS-CO Corporation) using the KBr disk method and were recorded with an average of 32 scans at a resolution of 4 cm⁻¹.

RESULTS AND DISCUSSION

Effects of the Hot Pressing Temperature

The particleboard was manufactured at different hot pressing temperatures with resin contents of 30 and 40 wt%. Figures 1 and 2 show the influence of hot pressing temperature on MOR and MOE.

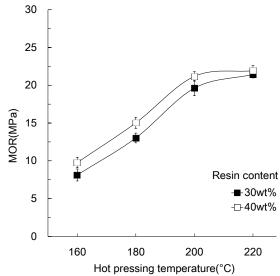


Fig. 1. Effect of hot pressing temperature on MOR in bending test. Error bars indicate standard deviations.

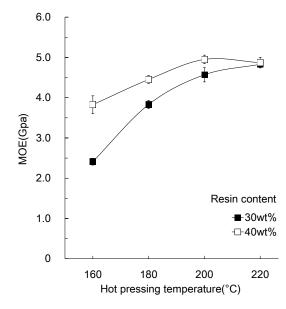


Fig. 2. Effect of hot pressing temperature on MOE in bending test. Error bars indicate standard deviations.

As the hot pressing temperature was increased, the values of MOR and MOE were enhanced irrespective of the resin content, and the values obtained at a 40 wt% resin content were a little higher than those of the 30 wt% resin content at the same hot pressing temperature. The maximum average values of MOR and MOE were 21.9 MPa (40 wt%-220 °C) and 5.0 GPa (40 wt%-200 °C), respectively. Analysis of variance (ANOVA) showed no significant difference in MOR and MOE among the three particleboards bonded with resin content at hot pressing temperature levels of 40-200, 30-220, and 40-220. In addition, those bending properties were comparable to the requirement of the type 18 JIS A 5908 standard (MOR>18MPa) (JIS A 5908. 2003). The results indicated that when the hot pressing temperature was higher than 200 °C, the influence of hot pressing temperature on the bending properties was small, and a temperature that is 200 °C or higher is required to obtain satisfactory bending properties of the particleboard bonded with tannin and sucrose.

Figure 3 shows the IB strength of the particleboard. Under both of the 30 and 40 wt% resin content conditions, the IB strength was obviously enhanced as the hot pressing temperature increased, and the maximum average value was 1.6 MPa, obtained from the board with a 40 wt% resin content at 220 °C. When the hot pressing temperature was higher than 200 °C, the IB strength of all the samples satisfied the requirement of the type 18 JIS A 5908 (IB>0.3MPa) (JIS A 5908. 2003).

It is worth noting that, if the hot pressing temperature was less than or equal to 200 °C, the IB strength of the board with 30 wt% was higher than that of the 40 wt% board. This phenomenon was perhaps due to the decrease in wood particles when the resin content was increased under the same board density. This means that the decrease in wood particles led to more void areas in the particleboard, and the increase in void areas had a negative effect on IB strength (Arabi *et al.* 2011). However, the opposite results were obtained when the hot pressing temperature was 220 °C, as the IB strength at a 40 wt% resin content was

a little higher than that at 30 wt%. This would be due to the promotion of adhesiveness at $220 \,^{\circ}$ C.

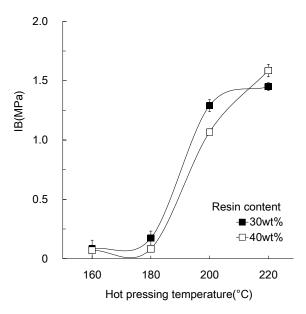


Fig. 3. Effect of hot pressing temperature on IB strength. Error bars indicate standard deviations.

Figure 4 shows the thickness changes of the particleboard. In the first immersion treatment (immersion at 20 °C for 24 h), the samples manufactured at 160 and 180 °C were decomposed, which indicated that the water resistance of those boards was very weak. Comparing with the results obtained from the board hot pressed at 200 and 220 °C, the TS values decreased greatly when the board was hot pressed at 220 °C. The TS values of 30 and 40 wt% resin content at 220 °C were 10 and 7%, respectively, which satisfied the requirement of the type 18 JIS A 5908 (TS<12%) (JIS A 5908. 2003).

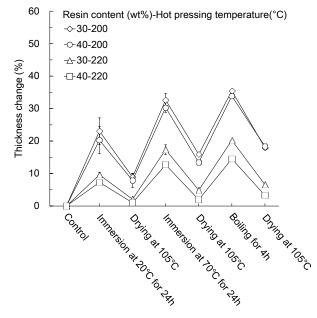


Fig. 4. Effect of hot pressing temperature on thickness change in a cyclic accelerated aging treatment. Error bars indicate standard deviations.

The subsequent cyclic accelerated aging treatment brought a stepwise increase in the thickness of the samples, and the boards pressed at 220 $^{\circ}$ C showed lower thickness swelling values than the boards pressed at 200 $^{\circ}$ C in each immersion treatment. This indicated that increasing the hot pressing temperature to 220 $^{\circ}$ C greatly promoted the water resistance of the particleboard bonded with tannin and sucrose.

Figure 5 shows the weight changes during the cyclic accelerated aging treatment. In the first immersion treatment, the weight increase of the boards pressed at 220 °C was obviously lower than those of the boards pressed at 200 °C. This indicated that the water absorption was inhibited by increasing the hot pressing temperature. Weight decreases ranging from 1.9 to 4.5% were observed after the first drying treatment, indicating that some elution from the samples occurred. The weight increases of 70 °C water immersion and boiling treatment were higher than the first treatment. This phenomenon was possible due to the decreasing of water resistance of adhesive and the penetration of water into the wood. (Umemura *et al.* 2013). In the last drying treatment, the weight decreases of the boards pressed at 200 and 220 °C ranged from 11.8 to 14.7% and 4.7 to 5.4%, respectively. There was a smaller weight decrease when the hot pressing temperature was 220 °C, which means that less elution occurred during the cyclic accelerated aging treatment. This indicated that the curing of tannin and sucrose adhesive at 220 °C led to a more complete insolubilization than curing at 200 °C.

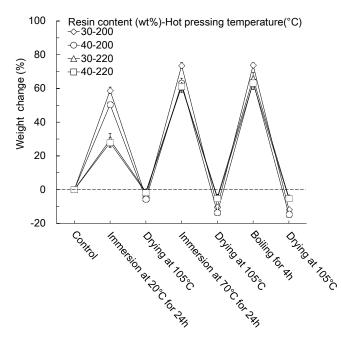


Fig. 5. Effect of hot pressing temperature on weight change in a cyclic accelerated aging treatment. Error bars indicate standard deviations.

Effects of Hot Pressing Time

Based on the results obtained, the effects of hot pressing time on the physical properties of particleboard bonded with 30 and 40 wt% resin content were investigated at 220 °C. Figures 6 and 7 show the influence of hot pressing time on MOR and MOE. The results show that the bending properties of all the samples satisfied the requirement of the type 18 JIS A 5908 standard (JIS A 5908. 2003). The maximum values of MOR and MOE were 23.5 MPa and 5.3 GPa, respectively, with a hot pressing time of 7 min and 40 wt% resin content. Based on the ANOVA analysis, there were no significant differences of

MOR among all samples. In terms of MOE, the values were increased in the hot pressing time range of 5 to 7 min, and decreased from 7 to 15 min. This means that prolonging hot pressing time would not promote the bending properties of the particleboard.

Figure 8 shows the effect of hot pressing time on IB strength. Both the 30 and 40 wt% resin contents showed the same trend in IB values with increasing hot pressing time. A significant increase was observed as the hot pressing time was prolonged from 5 to 10 min. This phenomenon would be due to a more efficient curing in the core layer. A slight decrease was observed in 15 min. The maximum value was 1.6 MPa in the case of a 40 wt% resin content and 10 min, and the IB values of all the samples except for the boards pressed for 5 min with a 30 wt% resin content were comparable to the requirement of the type 18 JIS A 5908 standard (JIS A 5908. 2003).

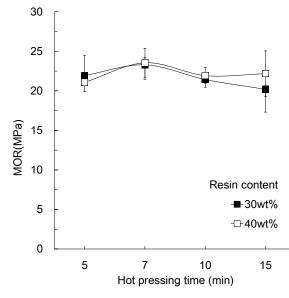


Fig. 6. Effect of hot pressing time on MOR in bending test. Error bars indicate standard deviations.

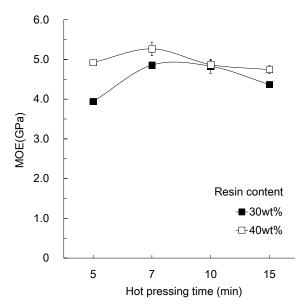


Fig. 7. Effect of hot pressing time on MOE in bending test. Error bars indicate standard deviations.

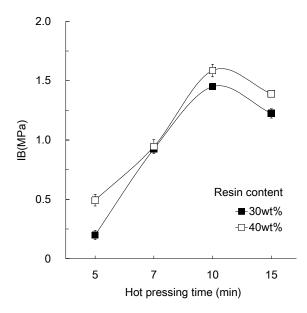


Fig. 8. Effect of hot pressing time on IB strength. Error bars indicate standard deviations.

Judging from the results of the bending and IB tests, when the hot pressing time was in the range of 5 to 7 min, the bending properties of the boards were good; however, the IB strength was weak. This phenomenon was explained as follows: the short hot pressing time led to sufficient curing at the surface but insufficient curing in the core of the particleboard, and the bending properties and IB properties of the particleboard were more strongly affected by the surface and core layers, respectively.

Figure 9 shows the thickness changes of the particleboard samples bonded with different hot pressing times and resin contents in the cyclic accelerated aging treatment.

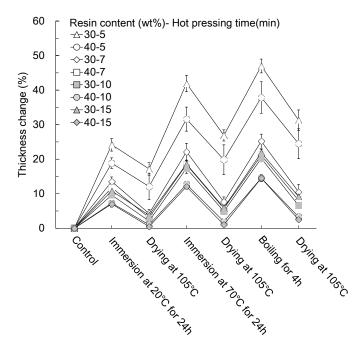


Fig. 9. Effect of hot pressing time on thickness change in a cyclic accelerated aging treatment. Error bars indicate standard deviations.

In the first immersion treatment, the TS values under the same resin content decreased in the time range of 5 to 10 min, and then they remained the same at 10 min and 15 min. In addition, the TS values of the boards bonded with a 40 wt% resin content were lower than those with a 30 wt% resin content at every hot pressing time. The minimum TS value was 7%, obtained from the boards bonded with a 40 wt% for 10 and 15 min. When the particleboard were manufactured with a 40 wt% resin content for 7 min, and 30 or 40 wt% resin content for 10 or 15 min, the TS values were comparable to the requirement of the type 18 JIS A 5908 standard (JIS A 5908. 2003). The subsequent cyclic accelerated aging treatment brought a stepwise increase in the thickness of the samples, and the least thickness changes were obtained in samples bonded with a 40 wt% resin content with 10 or 15 min. This indicated that the prolonging of hot pressing time contributed to the inhibition of the thickness change of the boards.

Figure 10 shows the weight changes of the particleboard bonded with different hot pressing times in the cyclic accelerated aging treatment. In the first immersion treatment, the weight increase between the boards hot-pressed for 10 and 15 min was lower than the boards hot-pressed for 5 and 7 min, meaning that water absorption was inhibited by prolonging the hot pressing time. The weight change range after the first drying treatment was -1.2 to -7.8%, indicating that some elution including adhesive occurred. In the 70 °C immersion treatment, the weight increase ranged from about 58 to 79% and dropped as the hot pressing time increased. The overall weight change between 70 °C immersion treatment and boiling treatment were almost the same, and the values were higher than the first immersion treatment. The final weight decrease with drying at 105 °C ranged from 4.7 to 14.8%, and a long hot pressing time corresponded to a slight weight decrease. Based on the results shown in Figs. 9 and 10, prolonging the hot pressing time brought good water resistance of the board.

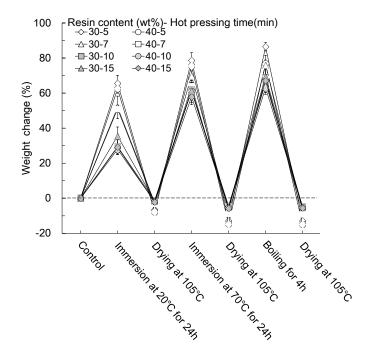


Fig. 10. Effect of hot pressing time on weight change in a cyclic accelerated aging treatment. Error bars indicate standard deviations.

Judging from the results obtained from Figs. 9 and 10, the optimal hot pressing time of the particleboard bonded with tannin and sucrose was 10 min. In terms of resin content, the best physical properties were obtained from the boards bonded with 40 wt% resin content, although the physical properties of the boards bonded with 30 wt% resin content also satisfied the requirement of the type 18 JIS A 5908 standard (JIS A 5908. 2003).

Thermal Analysis

Figure 11 shows the thermogravimetric (TG) and derivative TG (DTG) curves of the tannin, adhesive, and sucrose. Tannin (100/0) showed two-step degradation from the DTG curve, with the first stage at around 193 °C and the second stage at around 240 °C. In the first stage, 10% weight loss was observed from the TG curve. Judging from a previous study (Gaugler and Grigsby 2009), the first and the second stages can be attributed to the decomposition of polysaccharide from tannin and the decomposition of tannin, respectively. Sucrose (0/100) showed two-step degradation from the DTG curve. The first step at around 220 °C relates to the caramelization phase of sucrose (Gintner *et al.* 1989; Eggleston *et al.* 1996). The second step at around 270 °C was due to the production of a black aerated char-like solid (Eggleston *et al.* 1996). The adhesive (25/75) had a significant weight decrease at around 204 °C according to the DTG. Considering the results of tannin and sucrose, some reaction must occur between tannin and sucrose.

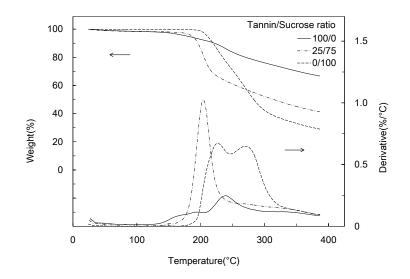


Fig. 11. TG and DTG curves of wattle tannin, adhesive and sucrose

Figure 12 shows the DSC curves of tannin, adhesive, and sucrose. The results for tannin (100/0) show an exothermic peak at around 255 °C. In research on the thermal degradation of pine tannin, an exothermic peak at around 255 °C was observed, and this was attributed to the charring of tannin (Gaugler and Grigsby 2009). Therefore, the peak observed in the present study was likewise attributed to the charring of tannin. In the case of sucrose (0/100), two sharp endotherms were observed at 180 and 226 °C. Considering the results of TG analysis of sucrose above, there was no peak at 180 °C; therefore, the endotherm peak at 180 °C was attributed to the melting of sucrose. The previous research also reported that the peaks at 180 and 226 °C were attributable to the melting and caramelization, respectively (Eggleston *et al.* 1996). When testing a mixed adhesive (25/75), two endothermic peaks were observed. The first endotherm at around 180 °C,

given what was shown in the sucrose-only condition, was due to the melting of sucrose. The second endotherm appeared at around 215 °C, which was lower than the second peak of the sucrose; therefore, this endotherm may be due to the caramelization of sucrose and the reaction between the products from caramelization and tannin. Based on the results obtained from Figs. 11 and 12, the reaction of the mixture composed of tannin and sucrose at a ratio of 25/75 occurred at a temperature higher than 200 °C. These results were consistent with the good physical properties of particleboard manufactured at 220 °C.

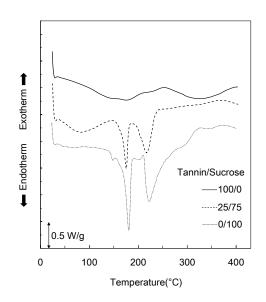


Fig. 12. DSC curves of wattle tannin, adhesive, and sucrose

Insoluble Matter and FT-IR analysis

The adhesive (25/75) was heated at 220 °C for different heating times, and insoluble matter after boiling treatment were observed. The results are shown in Fig. 13.

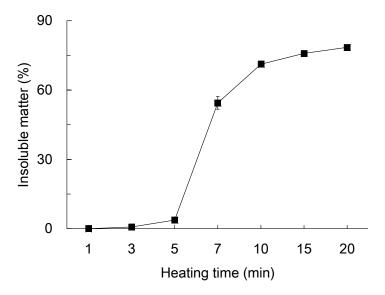
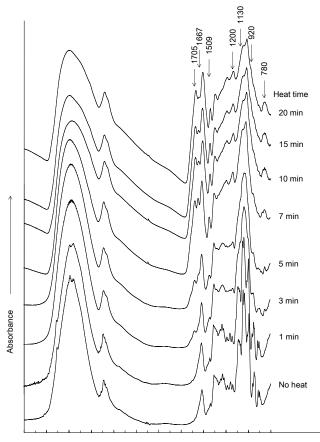


Fig. 13. Effect of heating time on insoluble matter. Error bars indicate standard deviations.

As the heating time was increased from 1 to 5 min, the value of insoluble matter was small, but when the heating time was longer than 5 min, the value of insoluble matter increased obviously. This phenomenon indicated that the curing of adhesive progressed significantly. When the heating time was longer than 10 min, the insoluble matter was higher than 70 wt% and changed slightly.

The effect of heating time on the chemical structure of the adhesive heated at 220 °C is shown in Fig. 14. Compared with the peaks obtained from the adhesive that had not undergone heat treatment, the absorption bands at around 1705, 1667, 1509, 1200, and 780 cm⁻¹ appeared clearly. The peak at around 1705 cm⁻¹ can be attributed to C=O stretching derived from the carbonyl group (Vaz and Ribeiro-Claro 2003). The peaks near 1509 and 780 cm⁻¹ are characteristic of the C=C stretching vibration and the unsubstituted CH=CH of the furan ring, respectively (Chuntanapum and Matsumura. 2009; Zhang *et al.* 2012; Alakhras and Holze 2007). The peak located around 1200 cm⁻¹ was associated with the the dimethylene ether bridges and/or -CO stretching of the benzene nucleus (-CH2OCH2-) (Kim and Kim 2003; Choi *et al.* 2002). A new peak located at 1667 cm⁻¹ was ascribed to the C=O stretching of 5-HMF (Chuntanapum and Matsumura 2009). Disappearance of two absorption peaks at around 1130 cm⁻¹ and 920 cm⁻¹ was observed. Those peaks were attributed to the glycosidic linkage (C-O-C) contribution (Kacurakova *et al.* 2000) and pyranose ring of sucrose (Seino *et al.* 1984). This indicated the decomposition of sucrose.



4000 3700 3400 3100 2800 2500 2200 1900 1600 1300 1000 700

Fig. 14. FT-IR curves of the curing adhesives

To observe the chemical structure change of cured adhesives, FT-IR spectra of the insoluble residue after boiling treatment were obtained, and the results are shown in Fig. 15. No results of the heating of the adhesives for 1 and 3 min were obtained because very little residue remained, as shown in Fig. 13. The absorption band at around 1667 cm⁻¹ was not observed in any samples due to the solubility of 5-HMF. The absorption bands at around 1705, 1509, 1200, and 780 cm⁻¹ were observed, indicating that the functional groups mentioned above existed in the cured adhesive. The FT-IR spectra of the adhesives heated for 10, 15, and 20 min were almost the same, which means that the chemical structures of the adhesive heated for more than 10 min were almost the same. Judging from these results, the cured adhesive had a furan ring structure and dimethylene ether bridges, and the water resistance was high. When the heating time was 10 min, higher values of insoluble substances were obtained, and the chemical structure of the adhesion system seemed to be stable. Therefore, a heating time of 10 min is needed to cure the adhesive sufficiently.

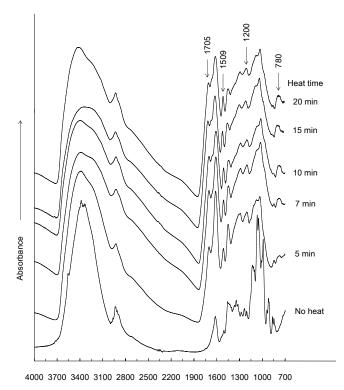


Fig. 15. FT-IR curves of the residue after the boiling treatment of the curing adhesives

CONCLUSIONS

1. The mechanical properties and water resistance of the particleboard increased as the hot pressing temperature and time increased. The optimum hot pressing temperature was 220 °C, and the optimum hot pressing time was 10 min, respectively. The MOR, MOE, IB, and TS of the boards manufactured under the optimum conditions were higher than the requirement of the type 18 JIS A 5908 standard (MOR>18 MPa, MOE>3 GPa, IB>0.3 GPa, TS>12%).

- 2. Thermal analysis showed that the reaction between tannin and sucrose (only at the ratio of 25/75) occurred at temperatures greater than 200 °C, which explained the promotion of the particleboard properties when the hot pressing temperature was increased to 220 °C.
- 3. When the heating time was longer than 10 min, the insoluble matter content was higher than 70 wt%. The FT-IR results showed the existence of a furan ring, a carbonyl group, and dimethylene ether bridges. In a comparison of the FT-IR spectra of the adhesives heated for different heating times, no obvious change of chemical structure was found in the hot pressing time range of 10 to 20 min. The results of boards were supported by thermal analysis and FT-IR.

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REFERENCES CITED

- Alakhras, F., and Holze, R. (2007). "In situ UV-vis-and FT-IR-spectroscopy of electrochemically synthesized furan-thiophene copolymers," *J. Synthetic Metals* 157, 109-119. DOI:10.1016/j.synthmet.2006.12.011.
- Arabi, M., Faezipour, M., Layeghi, M., and Enayati, A. A. (2011). "Interaction analysis between slenderness ratio and resin content on mechanical properties of particleboard," *J. For. Res.* 23, 461-464. DOI: 10.007/s11676-011-0188-2.
- Choi, M. H., Byun, H. Y., and Chung, I. J. (2002). "The effect of chain length of flexible diacid on morphology and mechanical property of modified phenolic resin," *J. Polymer* 43, 4437-4444. DOI:10.1016/S0032-3861(02)00226-4.
- Chuntanapum, A., and Matsumura, Y. (2009). "Formation of tarry material from 5-HMF in subcritical and supercritical water," *Ind. Eng. Chem. Res.* 48, 9837-9846. DOI: 10.1021/ie900423g.
- Eggleston, G., Trask-Morrell, B. J., and Vercellotti, J. R. (1996). "Use of differential scanning calorimetry and thermogravimetric analysis to characterize the thermal degradation of crystalline sucrose and dried sucrose-salt residues," *J. Agric. Food. Chem.* 44, 3319-3325. DOI: 10.1021/jf950836z.
- Gaugler, M., and Grigsby, W. J. (2009). "Thermal degradation of condensed tannins from radiata pine bark," J. Wood. Chem. Technol. 29, 305-321. DOI:10.1080/02773810903165671.
- Gintner, Z., Vegh, A., and Ritlop, B. (1989). "Determination of sucrose content of cariogenic diets by thermal analysis," *J. Thermal Analysis* 35, 1399-1404. DOI: 10.1007/BF01912916.
- Hoong, Y. B., Paridah, M. T., Loh, Y. F., Jalaluddin, H., and Chuah, L. A. (2011). "A new source of natural adhesive: *Acacia mangium* bark extractsco-polymerized with phenol-formaldehyde (PF) for bonding *Mempisang* (Annonaceae spp.) veneers," *Int. J. Adhes. Adhes.* 31, 164-167. DOI:10.1016/j.ijadhadh.2010.12.002

Jang, Y., Huang, J., and Li, K. (2011). "A new formaldehyde-free wood adhesive from renewable materials," *Int. J. Adhes. Adhes.* 31, 754-759. DOI:10.1016/j.ijadhadh.2011.07.003.

Japanese Industrial Standards (JIS) (2003). "JIS A 5908.Particleboard."

- Jeong, J. A., Antonyraj, C., Shin, S., Kim, S., Kim, B., Lee, K. Y., and Cho, J. K. (2013). "Commercially attractive process for production of 5-hydroxymethyl-2-furfural from high fructose corn syrup," *J. Ind. Eng. Chem.* 19, 1106-1111. doi:10.1016/j.jiec.2012.12.004.
- Kacurakova, M., Capek, P., Sasinkova, V., Wellner, N., and Ebringerova, A. (2000).
 "FT-IR study of plant cell wall model compounds: Pectic polysaccharides and hemicelluloses," *Carbohyd Polym.* 43, 196-203. DIO:10.1016/S0144-8617(00)00151-X.
- Kim, S., and Kim, H. (2003). "Curing behavior and viscoelastic properties of pine and wattle tannin-based adhesives studied by dynamic mechanical thermal analysis and FT-IR-ATR spectroscopy," J. Adhesion Sci. Technol. 17, 1369-1383. DOI:10.1163/156856103769172797.
- Krug, D. (2010). "Use of proteins as binders for wood-based panels," *Eur. J. Wood Prod.* 68, 289-301. DOI: 10.1007/s00107-010-0464-4.
- Lee, J. W., Thomas, L. C., Jerrell, J., Feng, H., Cadwallader, K. J., and Schmidt, S. J. (2011). "Investigation of thermal decomposition as the kinetic process that causes the loss of crystalline structure in sucrose using a chemical analysis approach," *J. Agric. Food.* Chem. 59, 702-712. DOI: 10.1021/jf104235d.
- Lei, H., Pizzi, A., and Du, G. B. (2008). "Environmentally friendly mixed tannin/lignin wood resins," *J. Appl. Polym. Sci.* 107(1), 203-209. DOI: 10.1002/app.27011.
- Mansouri, N.-E., Pizzi, A., and Salvado, J. (2007). "Lignin-based polycondensation resins for wood adhesives," J. Appl. Polym. Sci.103, 1690-1699. DOI: 10.1002/app.25098.
- Pizzi, A. (2006). "Recent development in eco-efficient bio-based adhesives for wood bonding: Opportunities and issues," J. Adhes. Sci. Technol. 20, 829-846. DOI:10.1163/156856106777638635.
- Seino, H., Uchibori, T., Nishitani, T., and Inamasu, S. (1984). "Enzymatic synthesis of carbohydrate esters of fatty acid (I) esterification of sucrose, glucose, fructose and sorbitol," JAOCS 61(11), 1761-1765. DOI: 10.1007/BF02582144
- Trosa, A., and Pizzi, A. (1997). "Stability and performance of tannin-accelerated PF resins for plywood," *J. Holz. als. Roh- und. Werkstoff.* 55, 306.
- Umemura, K. (2013). "Condensed tannin-containing composition which is cured by application of heat and pressure," WO2013018707 A1.
- Umemura, K., Sugihara, O., and Kawai, S. (2013). "Investigation of a new natural adhesive composed of citric acid and sucrose for particleboard," *J. Wood. Sci.* 59, 203-208. DOI:10.1007/S10086-013-1326-6.
- Vaz, P. D., and Ribeiro-Claro, P. J. A. (2003). "C-H...O hydrogen bonds in liquid cyclohexanone revealed by the v C=O splitting and the v C-H blue shift," *J. Raman Spectrosc.* 34, 863-867. DOI: 10.1002/jrs.1066.
- Yang, I., Kuo, M., Myers, D. J., and Pu, A. (2006). "Comparison of protein-based adhesive resins for wood composites," J. Wood. Sci. 52, 503-508. DOI: 10.1007/s10086-006-0804-5.

- Zhang, M., Yang, H., Liu, Y., Sun, X., Zhang, D., Xue, D. (2012). "Hydrophobic precipitation of carbonaceous spheres from fructose by a hydrothermal process," J. *Carbon* 50, 2155-2161. DOI:10.1016/j.carbon.2012.01.024.
- Zhao, Z., and Umemura, K. (2014). "Investigation of a new natural particleboard adhesive composed of tannin and sucrose," *J. Wood. Sci.* 60, 269-277. DOI: 10.1007/s10086-014-1405-3.

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