Effects of the Addition of Citric Acid on Tannin-Sucrose Adhesive and Physical Properties of the Particleboard

Zhongyuan Zhao,* Kenji Umemura, and Kozo Kanayama

The effects of citric acid on the curing properties of tannin-sucrose adhesives and on the physical properties of particleboard utilizing the adhesives were investigated. The citric acid content of tannin-sucrose adhesive was adjusted to 0.4, 1.8, 4.6, 13.8, 20.0, and 33.3%, which corresponded to pH values of the adhesive 40 wt% solution at 3.8, 3.0, 2.5, 2.0, 1.8, and 1.5, respectively. Thermal analysis showed that with increasing of citric acid content, the temperature of significant weight loss and the endothermic reaction of the tannin-sucrose-citric acid adhesive was reduced. When the adhesives were heated at 200 °C, boiled for 4 h, and adjusted to 20.0 and 33.3% citric acid content, the insoluble matter was increased significantly, and an absorption band derived from ester linkages and another peak, possibly from dimethylene ether bridges, were observed by FT-IR. The particleboards bonded with 20.0 and 33.3% citric acid adhesives at 200 °C satisfied the physical requirements of the type 18 Japanese Industrial Standards A 5908 (2003). Consequently, the addition of citric acid promoted the reaction between tannin and sucrose at a lower temperature, and the optimal hot pressing temperature decreased from 220 to 200 °C. The mechanical properties and water resistance of the particleboards were also enhanced.

Keywords: Wood adhesive; Tannin; Sucrose; Citric acid; Particleboard

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INTRODUCTION

Wood-based materials are commonly bonded with synthetic resins such as formaldehyde-based, isocyanate-based, and vinyl acetate resins (Yang *et al.* 2006; Mansouri *et al.* 2007). Because wood-based materials are generally used in housing construction and furniture manufacturing, they are frequently present in living environments (Sellers 2001; Zheng *et al.* 2007). However, many synthetic resin adhesives contain harmful chemical substances such as formaldehyde that cause environmental problems and health disorders. In addition, most synthetic resins are derived from fossil resources, which are limited resources. To address these problems, natural adhesives derived from biomass resources such as protein, tannin, lignin, and starch are being derived (Trosa and Pizzi 1997; Lei *et al.* 2008; Li *et al*. 2015). Generally, some chemicals, such as phenol and hexamine, which are derived from fossil resources, must be added to these conventional natural adhesives to obtain good bonding performance (Pizzi 2006; Jang *et al.* 2011; Wang *et al.* 2011). Therefore, development of natural adhesives that do not include chemicals derived from fossil resources is extremely desirable.

Wattle tannin and sucrose can be combined to make an adhesive for particleboard manufacturing (Zhao and Umemura 2014, 2015). The mechanical properties of the particleboard produced with a tannin-sucrose ratio of 25/75, resin content of 30 to 40 wt%,

hot pressing temperature of 220 °C, and hot pressing time of 10 min satisfied the requirements of the Japanese Industrial Standards A 5908 (2003) type 18. However, the hot pressing temperature, time, and resin content were much higher than that of particleboard produced using commercial synthetic resins. During the reaction between tannin and sucrose, 5-hydroxymethyl-2-furfural (5-HMF) forms from the decomposition of sucrose, and then 5-HMF reacts with tannin (Zhao and Umemura 2015). Methods or substances that accelerate sucrose decomposition to 5-HMF seems to improve the manufacturing conditions of particleboard bonded with tannin and sucrose.

Acid compounds catalyze the decomposition of sucrose, thus increasing the yield of 5-HMF (Locas and Yaylayan 2008; Sievers *et al*. 2009). In addition, acid compounds reduce the temperature of 5-HMF formation (Haworth and Jones 1944; Asghari and Yoshida 2006). Citric acid is effective in forming 5-HMF from saccharides (Asghari and Yoshida 2006). Citric acid (2-hydroxy-1,2,3-propanetricarboxylic acid) has a certain acidity (Grudpan *et al.* 1998). It is an organic polycarboxylic acid containing three carboxyl groups, and it is mainly produced by the fermentation of sucrose (Shu and Johnson 1948). As a safe natural substance, citric acid is widely used in food, beverages, and pharmaceuticals (Černá *et al*. 2003; Summers and Enever 2006). In addition, citric acid can be used as a natural adhesive to manufacture wood-based materials (Umemura *et al.* 2012), and it reacts with sucrose (Umemura *et al*. 2013, 2015). Therefore, the addition of citric acid is expected to promote the curing reaction of the adhesive and reduce the required hot-pressing temperature. In this study, citric acid was incorporated into a tannin and sucrose adhesive, and the effect of this addition on the hot pressing temperature of particleboard was investigated.

EXPERIMENTAL

Materials

Adhesive components

Wattle tannin (commercial name: tannic acid ME) was purchased from the Fuji Chemical Industry Co. (Wakayama, Japan). Sucrose (guaranteed reagent) and citric acid (extra pure reagent) were purchased from Nacalai Tesque, Inc. (Kyoto, Japan). These materials were used without further purification and were dried in a vacuum oven at 60 °C for 15 h before use.

Particles

Recycled wood particles were obtained from a particleboard company, and they were screened using a sieving device to collect particles between 0.9 mm and 5.9 mm. The moisture content of the original particles was 3 to 4 wt%. Before particleboard manufacturing, particles were dried in an oven at 80 °C for 12 h to a final moisture content of 2 wt%.

Methods

pH adjustment of adhesive solution by adding citric acid

The citric acid content in tannin-sucrose adhesive was determined by the adjustment of pH values. The optimal ratio of tannin to sucrose is 25:75 (Zhao and Umemura 2014). In this study, 25 g of tannin and 75 g of sucrose were mixed in a beaker, and 150 g of distilled water was added to the mixture to blend a tannin-sucrose adhesive

solution at a concentration of 40 wt%. Citric acid was also dissolved in distilled water at a 40 wt% concentration. Subsequently, 0.5 g of the citric acid solution was added stepwise to the tannin-sucrose solution. The viscosity and pH of the solution at 20 °C was measured by a rotational viscometer (Viscolead One, Fungilab S.A., Barcelona, Spain) with cone-plate type R2 and a pH meter (D-51, Horiba Scientific, Kyoto, Japan), respectively. This process was repeated several times until the tannin-sucrose solution reached pH 1.5. Basic information regarding the tannin-sucrose-citric acid adhesives is presented in Table 1.

Table 1. Formulation of Tannin-Sucrose-Citric Acid Adhesives

The ratio of tannin:sucrose:citric acid	Addition of citric acid solution (g)	Citric acid content in adhesive (wt%)	Concentration (wt%)	Viscosity at 20 °C (mPa·s)	pH
25:75:0.0	0.0	0.0	40	51.3	4.8
25:75:0.4	1.0	0.4		50.5	3.8
25:75:1.8	4.5	1.8		48.7	3.0
25:75:4.8	12.0	4.6		43.5	2.5
25:75:16	40.0	13.8		42.7	2.0
25:75:25	62.5	20.0		40.4	1.8
25:75:50	125.0	33.3		36.5	1.5

Thermal analysis

After pH adjustments, 100 g of adhesive was prepared for each variation in citric acid content. In each version of the adhesive solution, 20 g of the solution was divided into 10 aluminum cups and dried in an oven at 80 °C for 12 h. The samples were then pulverized to smaller than 250-μm mesh size to obtain the uncured adhesive powder. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were carried out using a TGA 2050 (TA Instruments, Tokyo, Japan) and DSC 2910 (TA Instruments, Tokyo, Japan), respectively, and the samples were scanned from room temperature to 400 °C at a rate of 10 °C/min under nitrogen purging with the flow rate at 100 mL/min and 40 mL/min, respectively.

Insoluble matter and FT-IR

As shown in Table 1, 100 g adhesive solutions with of 4.6%, 13.8%, 20.0%, and 33.3% citric acid were prepared. Uncured adhesive powder was obtained in the same manner as in the thermal analysis. Each of the uncured adhesive powders was divided into four parts, and these were heated in an oven at 160, 180, 200, or 220 °C for 10 min. Next, 2-g samples of each cured adhesive were boiled in distilled water for 4 h to obtain the insoluble matter. The boiling treatment was carried out in triplicate. All samples obtained from heating 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, JASCO Corporation, MD, USA) using the KBr disk method; they were scanned from 700 to 4000 cm^{-1}, and recorded with an average of 32 scans at a resolution of 4 cm^{-1}.

Manufacture of particleboard

Tannin, sucrose, and citric acid were dissolved in distilled water with 4.6%, 13.8%, 20.0%, and 33.3% citric acid contents and a solution concentration of 40 wt% (Table 1). The adhesive solution was sprayed onto particles in a blender at 30 wt% resin content based

on the weight of the oven-dried particles. After spraying, the particles were dried at 80 °C for 12 h until the moisture content was 3 to 6 wt%. The particles were mat-formed using a 300 × 300 mm forming box. For every variation in citric acid content, particleboards were manufactured with hot pressing temperatures of 160, 180, 200, and 220 °C; the hot pressing time was 10 min. The thickness was controlled using a 9-mm distance bar during pressing. The size of the manufactured board was 300 × 300 × 9 mm, and the target density was 0.8 g/cm^3.

Evaluation of board properties

The boards were conditioned for 1 week at 20 °C and 60% relative humidity (RH) and then evaluated according to the Japanese Industrial Standard for particleboard (JIS A 5908 2003). The average density of the particleboards after conditioning was calculated by measuring the dimension and weight of all the samples obtained from each particleboard. The mechanical properties of the particleboards were investigated with a universal testing machine (Model 4411, Instron Corp., PA, USA). The static three-point bending test was carried out on a 200 × 30 × 9 mm specimen 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 obtained. The internal bond strength (IB) test was performed on a 50 × 50 mm specimen with a loading speed of 2 mm/min, and thickness swelling (TS) after water immersion for 24 h at 20 °C was measured in specimens 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 to investigate the water resistance in severe conditions. Each experiment was performed in quintuplicate, and the average value and standard deviation were calculated. Statistical significance of ANOVA analysis was considered for p values < 0.05. The MOR, MOE, and IB values were corrected for target density based on each regression line between actual values and sample densities of the mechanical properties.

RESULTS AND DISCUSSION

Adhesive Analysis

Thermal analysis

In the thermogravimetric (TG) analysis of adhesives adjusted to different citric acid contents, weight loss in the tannin-sucrose-citric acid adhesive started at a lower temperature than in the tannin-sucrose adhesive (Fig. 1a). The tannin-sucrose adhesive (0 wt% citric acid content) exhibited a sharp weight loss from 186 to 217 °C. When citric acid was added, a gradual weight loss was observed, and the starting weight loss temperature decreased with increasing citric acid content.

In the derivative TG (DTG) curves (Fig. 1b), tannin-sucrose adhesive (0 wt% citric acid content) had a noticeably weight loss at 204 °C; this result was attributed to the reaction between tannin and sucrose (Zhao and Umemura 2015). In the tannin-sucrose-citric acid adhesives, the temperatures of significant weight loss in adhesives with 0.4, 1.8, 4.6, 13.8, 20.0, and 33.3 wt% citric acid content were 202, 200, 176, 162, 159, and 151 °C, respectively. As the citric acid content increased, the temperature of significant weight loss decreased. Because acid compounds catalyze sucrose decomposition at lower temperatures (Haworth and Jones 1944; Asghari and Yoshida 2006), the observed results reflected the

decomposition of sucrose or another reaction between tannin, sucrose, and citric acid. A shoulder at 200 °C in the 20.0 and 33.3 wt% citric acid adhesive curves reflected an apparent degradation of citric acid (Umemura *et al*. 2012). Another shoulder located at 300 °C was observed in the curves produced from all tannin-sucrose-citric acid adhesives, and this decline was possibly caused by decomposition of the cured adhesives.

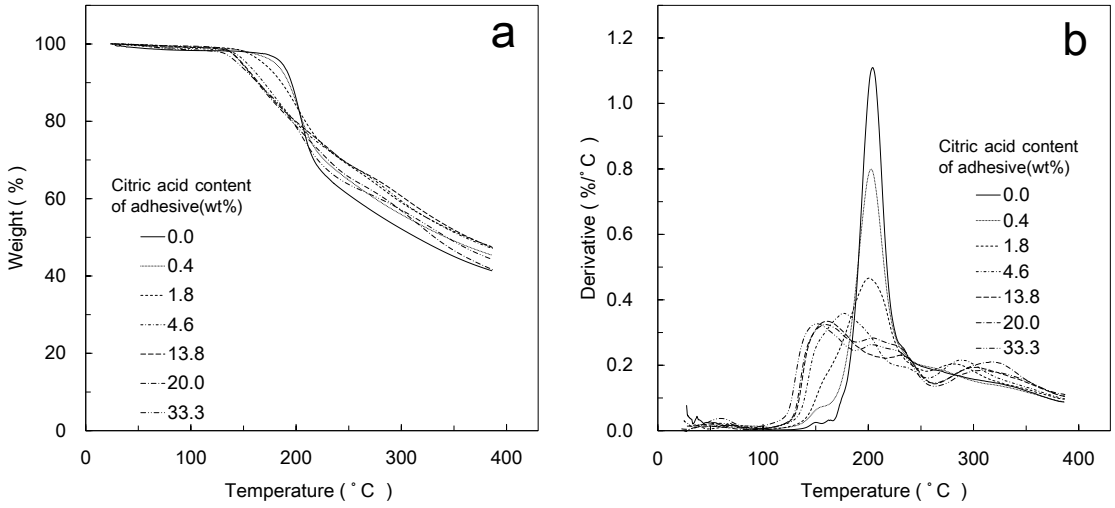

Fig. 1. TG (a) and DTG (b) curves of the adhesives with different citric acid contents

DSC curves of the adhesives showed that tannin-sucrose adhesive (0 wt% citric acid content) had two endothermic peaks (Fig. 2). The peak located at 180 °C was due to the melting of sucrose (Eggleston *et al.* 1996), and the peak located at 215 °C was attributed to the caramelization of sucrose or polymerization (Eggleston *et al.* 1996; Zhao and Umemura 2015).

In tannin-sucrose-citric acid adhesives with 0.4 and 1.8 wt% citric acid, a shoulder was detected at 157 °C, and endothermic peaks were observed at 210 and 194 °C. TG analysis of these two adhesives indicated that the weight loss at 157 °C was not noticeable. Therefore, the shoulder must have been due to the melting of some substance, which could have been citric acid (Barbooti and Al-Sammerrai 1986). The endothermic peak located at 210 °C was due to caramelization or polymerization reactions.

When the citric acid content was equal to or greater than 4.6 wt%, a broad endothermic peak was observed at 155 °C in all adhesives. In accordance with the TG analysis, this peak was probably due to the loss of sucrose or some reaction between tannin, sucrose, and citric acid. In adhesives with 20.0 and 33.3 wt% citric acid, a small endothermic shoulder was observed at 212 °C, which was due to the loss of citric acid (Barbooti and Al-Sammerrai 1986).

In sum, increasing citric acid content decreased the temperature of weight loss and endothermic reactions from 220 to 155 °C, demonstrating that citric acid effectively reduced the reaction temperature of the adhesive. When the citric acid content was equal to or greater than 4.6 wt%, the reaction temperature was noticeably reduced. Therefore, in the next set of experiments, tannin-sucrose-citric acid adhesives with 4.6, 13.8, 20.0, and 33.3 wt% citric acid contents were tested, and tannin-sucrose adhesive with 0 wt% citric acid content was used as a reference.

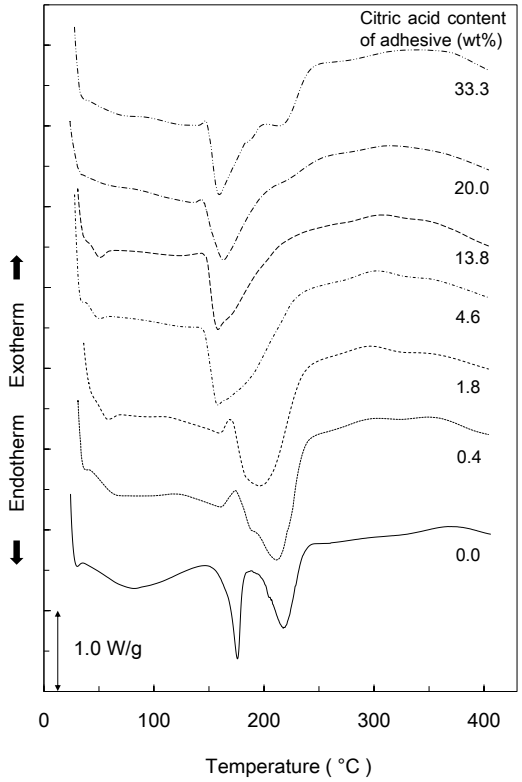

Fig. 2. DSC curves of the adhesives with different citric acid contents

Insoluble matter

The adhesives were heated at 160, 180, 200, and 220 °C for 10 min, and the insoluble matter after boiling was measured (Fig. 3).

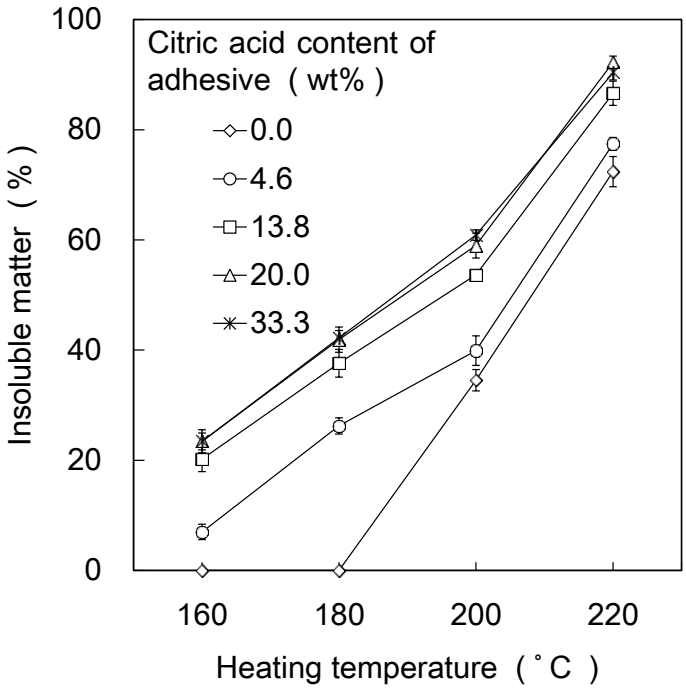

Fig. 3. Insoluble matter of the adhesives with different citric acid contents

As the heating temperature was increased, the insoluble matter also increased considerably. In each heating temperature condition, the insoluble matter increased with increasing citric acid content, indicating that the addition of citric acid improved adhesive curing. Analysis of variance (ANOVA) showed no significant (p>0.05) difference between the adhesives with 20.0 and 33.3 wt% citric acid in all heating temperature conditions. Notably, when the heating temperature was 160 or 180 °C, there was no insoluble matter in 0 wt% citric acid tannin-sucrose adhesive, but some insoluble matter was present in adhesives containing citric acid. Judging from the thermal analysis, this result reflects the reaction of adhesives with citric acid at a lower temperature than in the tannin-sucrose adhesive. When the heating temperature was 200 °C, adhesives with 20.0 and 33.3 wt% citric acid contained 60% insoluble matter, nearly two times higher than the 0 wt% citric acid tannin-sucrose adhesive.

FT-IR analysis

The effect of citric acid on the chemical structure of the adhesives cured at 200 °C for 10 min before and after boiling treatment was investigated by FT-IR (Fig. 4). Compared with the peaks obtained from tannin-sucrose adhesive (0 wt% citric acid content), the absorption bands of the tannin-sucrose-citric acid adhesives appeared at 2630 and 1731 cm^{-1}, and these two peaks located at 1190 to 1200 cm^{-1} and 780 cm^{-1} increased as citric acid content increased (Fig. 4a). The broad absorption band at 2630 cm^{-1} belongs to the vibration of H-bonded carboxylic OH groups (Zagar and Grdadolnik 2003). The peak at 1731 cm^{-1} can be attributed to C=O stretching derived from the carboxyl group and/or ester group (Yang and Wang 1996; Zagar and Grdadolnik 2003). The peak at 1190 cm^{-1} is due to C-O stretching from the carboxyl group and/or dimethylene ether bridges (Mawhinney and Yates 2001). Meanwhile, the peak located at 780 cm^{-1} is derived from the unsubstituted CH=CH of the furan ring (Cimino *et al.* 1972). In the tannin-sucrose adhesive (0 wt% citric acid content), the peak located at 1705 cm^{-1} is derived from the C=O carbonyl group (Sellitti *et al.* 1990). The peak located at 1200 cm^{-1} can be attributed to C-O-C dimethylene ether bridges in the phenolic resin and wattle tannin adhesives (Choi *et al.* 2002; Kim and Kim 2003); therefore, the peak located at 1200 cm^{-1} derived from tannin-sucrose adhesive (0 wt% citric acid content) was possibly due to the dimethylene ether bridges.

The peaks derived from citric acid appeared strongly when the citric acid content of the adhesives was high, and some water-soluble substances were contained in the samples. The chemical structure of the insoluble matter in cured adhesives was observed by FT-IR measurements after the cured adhesives were boiled (Fig. 4b). The absorption band at 2630 cm^{-1} was barely recognizable in all spectra, indicating that carboxyl groups did not exist in the insoluble matter. The peaks located at 1731 and 1190 cm^{-1} were only observed in the insoluble matter of the adhesives with 20.0 and 33.3 wt% citric acid. The peaks increased with increasing citric acid content, implying that the ester linkage and dimethylene ether bridges were more abundant as the citric acid increased. The peaks at 1705 and 1200 cm^{-1} were observed in the insoluble matter of the adhesives with 0, 4.6, and 13.8 wt% citric acid, indicating that the chemical structure of the insoluble matter of these adhesives was very similar. The peak at 780 cm^{-1} increased slightly as citric acid increased, suggesting that the furan ring content in the insoluble matter increased. In sum, when the citric acid content of adhesive was adjusted to 20.0 and 33.3%, ester linkages and dimethylene ether bridges were formed. With increased citric acid content, the furan ring content increased, and more chemical bonds derived from tannin participated in the reaction.

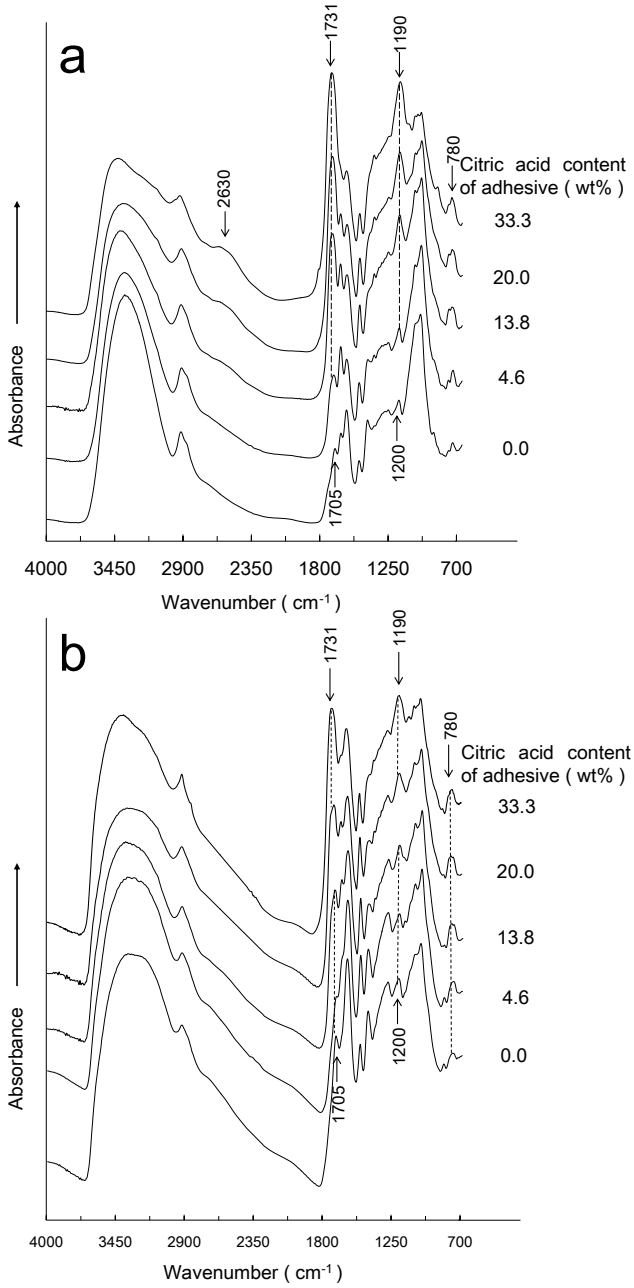

Fig. 4. Infrared spectra of the adhesives heated at 200 °C for 10 min (a) before and (b) after boiling

Evaluation of Board Properties

Bending properties

Particleboards bonded using adhesives with various citric acid contents were manufactured at different hot pressing temperatures, and MOR and MOE were evaluated (Fig. 5). As the hot pressing temperature increased, both MOR and MOE were enhanced in all particleboards. When the pressing temperature was equal to or higher than 200 °C, the bending properties of the particleboards satisfied the requirements of the 18 type JIS A 5908 standard (MOR > 18 MPa, MOE > 3 GPa) (JIS A 5908 2003). With hot pressing

temperatures of 160, 180, and 200 °C, the MOR and MOE values of the boards bonded with adhesives containing citric acid were higher than those bonded with tannin-sucrose adhesive, as the curing temperature of the tannin-sucrose-citric acid adhesives was lower. The maximum average MOR and MOE values were 21.5 MPa and 4.9 GPa, respectively, which were obtained from the board bonded with 33.3% citric acid adhesive at 200 °C. Analysis of variance (ANOVA) showed no significant difference (p>0.05) in MOR or MOE in boards bonded with 20.0 and 33.3% citric acid adhesives at 200 °C, and there was no significant (p>0.05) difference in any boards hot-pressed at 220 °C.

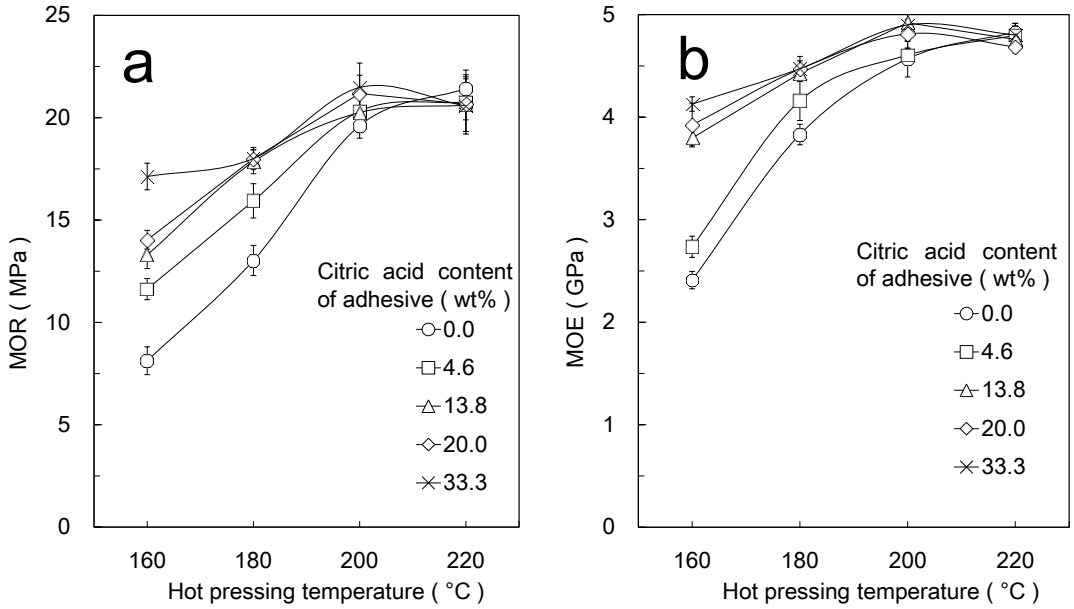

Fig. 5. Effects of the citric content of adhesive and hot pressing temperature on (a) MOR and (b) MOE in bending properties. Error bars indicate standard deviations.

IB properties

The IB of the particleboards was also evaluated (Fig. 6). The strength increased with increasing hot pressing temperature, irrespective of the citric acid content. The IB of the boards bonded with tannin-sucrose-citric acid adhesives at all hot pressing temperatures satisfied the requirements of the 18 type JIS A 5908 standard (IB > 0.3 MPa) (JIS A 5908 2003). However, the maximum value obtained from the board bonded with tannin-sucrose (0 wt% citric acid content) adhesive at 220 °C was 1.45 MPa. Analysis of variance (ANOVA) showed no significant difference (p>0.05) between the boards bonded with 20.0 and 33.3% citric acid adhesives at all hot pressing temperatures. When the hot pressing temperature was equal to or lower than 180 °C, the IB of the board bonded with tannin-sucrose adhesive was very low; however, the particleboard bonded with the adhesives with citric acid exhibited superior IB because of the curing of citric acid-containing adhesives at 160 and 180 °C. As the hot pressing temperature increased to 200 and 220 °C, the opposite result was obtained, with IB increasing with the decreasing citric acid content. This phenomenon requires further clarification.

Overall, when the hot pressing temperature was 160 or 180 °C, the mechanical properties of the particleboard increased with increasing citric acid (Fig. 5, 6). However, citric acid did not enhance particleboard mechanical properties when the hot pressing temperature was 200 or 220 °C.

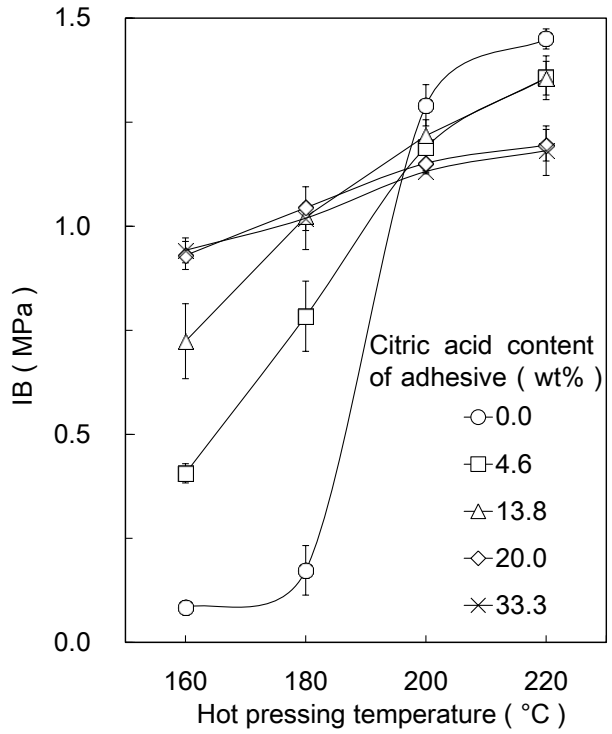

Fig. 6. Effects of adhesive citric acid content and hot pressing temperature on IB. Error bars indicate standard deviations.

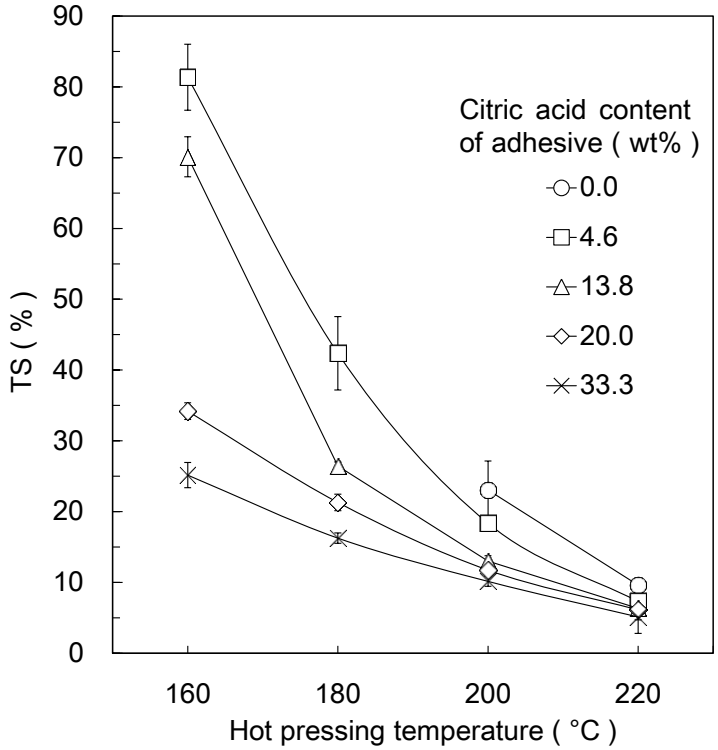

Fig. 7. Effects of citric content and hot pressing temperature on TS. Error bars indicate standard deviations.

Water resistance properties

Figure 7 shows the TS analysis of the particleboards. When the particleboards were bonded with tannin-sucrose (0 wt% citric acid content) adhesive at 160 or 180 °C, the boards decomposed during the immersion treatment, showing very low water resistance. As the hot pressing temperature increased, the TS values of the particleboards decreased irrespective of the citric acid content of the adhesive. At each hot pressing temperature, the TS values decreased with increasing citric acid, indicating that the addition of citric acid improved particleboard TS properties. The best value was 5%, obtained from the board bonded with 33.3% citric acid adhesive at 220 °C. For the particleboards bonded with 20.0 and 33.3% citric acid adhesives at 200 °C and for all boards manufactured at 220 °C, the TS values satisfied the requirements of the 18 type JIS A 5908 standard (TS < 12%) (JIS A 5908 2003).

To further clarify the effect of citric acid on water resistance, a cyclic accelerated aging treatment was performed (Fig. 8), and the thickness changes of the particleboards bonded at 200 °C were measured (Fig. 8a). The results for immersion at 20 °C for 24 h were similar to those shown in Fig. 7. Generally, a stepwise increase in the thickness of the boards was observed. For boards bonded with tannin-sucrose adhesive at 200 °C, the final thickness change was 17.9%, whereas the board manufactured with 33.3% citric acid adhesive showed a 6.1% change.

When the particleboards were bonded at 220°C, the thickness changes were similar to the results shown in Fig. 8a; however, the thickness changes after every treatment were lower than the changes in the boards manufactured at 200 °C. The least final thickness change was 2.1%, which was obtained from the board manufactured with 33.3% citric acid adhesive at 220 °C.

The weight changes of the particleboards bonded at 200 °C in a cyclic accelerated aging treatment were measured (Fig. 8b). In the first water immersion treatment, water absorption was similar to the TS data. This result indicated that the addition of citric acid to the adhesives inhibited water absorption by the particleboards. The weight decrease in the subsequent drying treatment of all boards ranged from -4.5% to -1.6%, suggesting that a slight elution of the adhesive components occurred. The weight increase of all the samples in the 70 °C immersion treatment was higher than in the first treatment. This effect could be due to the lowering of the water resistance of the adhesive and the penetration of water into the wood (Umemura *et al*. 2013).

The weight change of each board after boiling treatment was almost the same as in the warm water treatment, indicating that the boards exhibited excellent stability during boiling. The least final weight change of the board manufactured at 200 °C was -8.0%, which was obtained from particleboards bonded with 20.0 and 33.3% citric acid adhesives. The boards bonded with adhesives containing citric acid showed smaller final weight changes than those manufactured with the tannin-sucrose adhesive. The particleboards bonded at 220 °C showed the same trend of weight changes in the cyclic accelerated aging treatment.

The final weight change of the board manufactured using tannin-sucrose adhesive at 220 °C was -4.7%, whereas the board manufactured with 33.3% citric acid adhesive showed a relatively small value at -3.8%. Thus, the addition of citric acid enhanced water resistance in the particleboards.

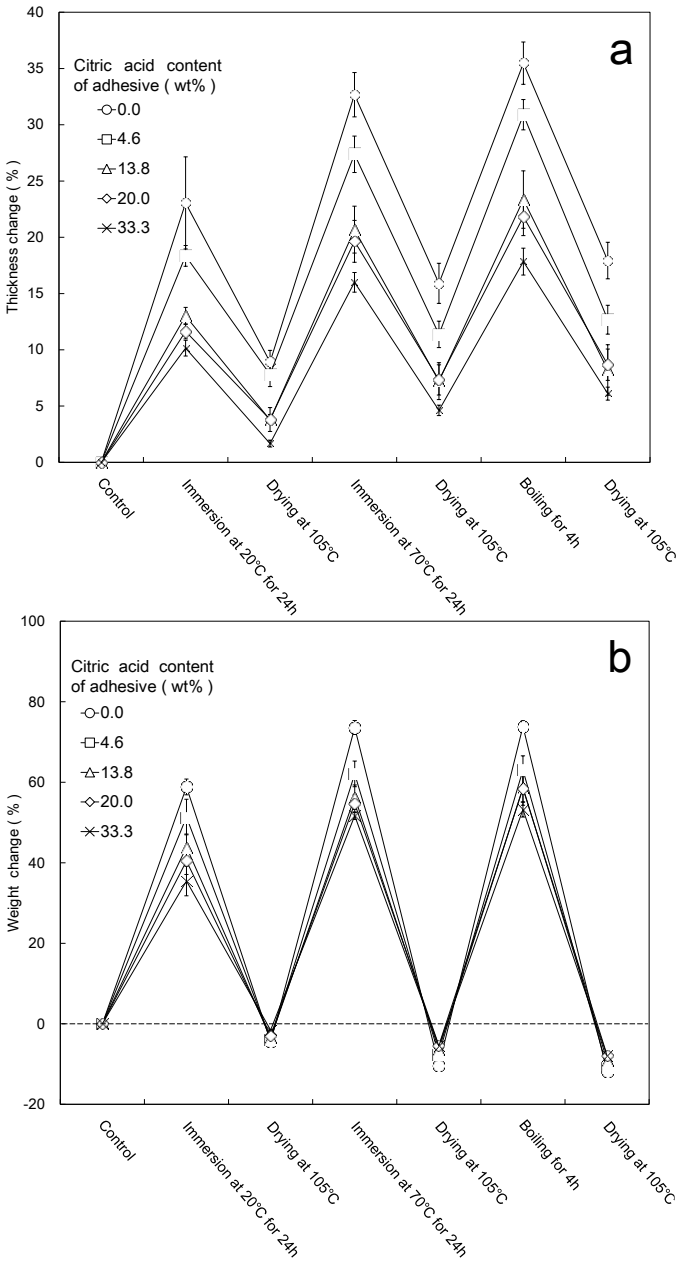

Fig. 8. Effects of the citric acid content of adhesive on (a) thickness changes and (b) weight changes in particleboards bonded at 200 °C in a cyclic accelerated aging treatment. Error bars indicate standard deviations.

CONCLUSIONS

1. The addition of citric acid to adhesive reduced the required reaction temperature, and the insoluble matter remaining from 20.0 and 33.3% citric acid adhesives at 200 °C was two times higher than in the tannin-sucrose adhesive. In addition, ester linkages and dimethylene ether bridges were recognized by FT-IR.

2. The physical properties of the particleboards bonded with tannin-sucrose-citric acid adhesive with 20.0 and 33.3% citric acid at 200 °C above satisfied the requirements of the type 18 JIS A 5908 standard (2003).
3. The addition of citric acid promoted the reaction between tannin and sucrose at a lower temperature and decreased the hot pressing temperature to 200 °C while enhancing the bending properties and water resistance of particleboards.

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