# Evaluation of the Mechanical and Physical Properties of Plywood Adhesively Bonded with Cellulose Nanofiber

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## Abstract

The use of plywood made from domestic softwoods is being promoted in Japan. Synthetic adhesives derived from fossil resources are typically used to manufacture plywood. However, because of problems such as high environmental impact and difficulty in recycling, research on the boards produced using adhesives derived from natural products has been active. In this study, we focused on cellulose nanofiber (CNF), a natural material that has been attracting attention in recent years, to determine if plywood can be made using CNF as the binder and examined the effects of different hot-pressing temperatures and veneer surface pretreatments (wetting and scratching treatment). The results showed that CNF-supplemented plywood could be produced regardless of hot-pressing temperature or pretreatment. CNF-supplemented plywood with pretreatment at 220°C showed the highest shear strength, >0.4 MPa. Pretreatment improved the bending properties, whereas pressing temperature did not affect the bending properties of CNF-supplemented plywood. CNF-supplemented plywood with pretreatment fulfilled the required modulus of rupture of E50-F160 structural plywood of Japanese Agricultural Standards, whereas modulus of elasticity and shear strength on them did not. Those results indicate that CNF is a useful binder for plywood and pretreatment on veneer surfaces enhances the adhesion performance of plywood.

Plywood is a wood material consisting of three or more rotary-cut wood veneers that are laminated such that the alternating layers are perpendicular to each other. Plywood was the most produced wood material in Japan as of 2020 (Forestry Agency 2021). Most logs used for plywood in Japan originate from Southeast Asian wood. However, because of restrictions on log exports in exporting countries and other factors, the use of domestic softwood lumber, mainly Japanese cedar, cypress, and larch, is rapidly increasing. The share of domestic lumber in domestic plywood production reached 91 percent in Japan in 2020 (Forestry Agency 2021). Japanese cedar (*Cryptomeria japonica*) is the most abundant domestic softwood species produced in Japan (Forestry Agency 2021). Therefore, its effective utilization is important. Plywood is usually manufactured from softwoods using adhesives derived from fossil resources, causing environmental impacts during production. The emission of formaldehyde and volatile organic compounds also causes health hazards. To solve these problems, recently high-density polyethylene films have been used as formaldehyde-free adhesives for plywood fabrication (Ashori et al. 2023a, 2023b). The use of binderless boards is also being explored (Sato 2012). Plywood is also being produced through self-bonding between veneers by scratching the veneer surfaces before hot pressing (Kurokochi et al. 2019). The adhesion of binderless boards is affected by the thermal denaturation of the chemical components in the wood and plant biomass, which results in higher

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temperatures and compression rates than during typical plywood production.

Cellulose nanofiber (CNF) is a fibrous material with a diameter of 3 to 100 nm that is obtained from various plants by physical and chemical methods. CNF is a high-performance material that is lightweight yet has very high mechanical properties and a very low coefficient of linear thermal expansion (Yano 2007). Recently many researchers investigated phenol–formaldehyde adhesive incorporated with cellulose nanomaterials to improve the adhesion property of wood-based materials (Naghizadeh et al. 2024). The recent use of CNF as a natural binder to produce boards has also been investigated. Previous studies have reported that CNF addition improves the bending strength and water resistance of powderboard made from wood flour, fiberboard made from Monterey pine or corn, and particleboard made from recycled wood (Kojima et al. 2013, 2018, 2021; Theng et al. 2015). Recently, fiber-based insulation material bonded with CNF met the standard for cellulosic fiber insulating board on ASTM C-208 (Hossain and Tajvidi 2024).

However, most studies of the boards produced using CNF have focused on particleboard or fiberboard, and their application in plywood production has not been studied. Adding CNF likely improves board performance through hydrogen bonding and physical entanglement with the elements, and these effects may also be manifested during plywood production. CNFs are often produced as highly viscous, homogeneous waterborne dispersion fluids. Adhesive for plywood needs adequate wettability and viscosity for uniform application on the surface of the veneer; therefore CNF would be suitable for use as a binder for plywood. Previous studies have reported that self-bonding strength between veneers improves as the moisture content of the veneer surface increases (Kurokochi et al. 2019).

The objective of this study was to determine whether domestic softwood plywood can be produced using CNF. The effects of different hot-pressing temperatures on the physical and mechanical properties of plywood during fabrication and the effects of wetting and scratching treatment on the veneer surfaces were investigated. To determine the effect of CNF addition on the physical and mechanical properties of plywood, the properties of the plywood with and without CNF were evaluated under the same fabrication conditions.

## Materials and Methods

### Materials

A total of 144 rotary-cut Japanese cedar veneers (200 by 200 by 3.5 mm) from a sapwood region was used to prepare the plywood. Veneers that have a smaller number of lathe check were manually selected by visual inspection. A commercial CNF slurry (BinFi-s, WFo-10010; Sugino Machine Ltd., Uozu, Japan) consisting of 10 weight percent CNF and 90 weight percent water was used in this study. A nominal fiber diameter, specific surface area, average degree of polymerization, and viscosity were 10 to 50 nm, 120 $m^2$/g, 200, and 110 Pa·s, respectively. Before the experiment, the CNF was diluted to 2.5 weight percent with distilled water using a homogenizer (Polytron, PT2500; Kinematica, Malters, Switzerland).

### Plywood manufacturing

Table 1 shows the plywood manufacturing conditions. A total of eight manufacturing conditions combining pretreatment, CNF application, and pressing temperature was tested. Six replications were manufactured per condition. In this study, inner (pith side) and outer surfaces were not considered for pretreatment and layer configuration since the number of lathe check was low. The pretreatment application consisted of wetting the surfaces of the veneers with running water for 1 minute, and then a steel wire brush (SUN UP Channel brush no. 29457, Taiyo Shokai Co. Ltd., Osaka, Japan) was used to make 50 scratches in a circular motion (Kurokochi et al. 2019) before CNF application. A 2.5 weight percent CNF slurry was manually applied on both sides of the plywood veneer using a roller at a spread rate of 250 g/$m^2$ (6.25 g/$m^2$ for dry CNF basis). This means that the addition of water by spreading CNF slurry was 9.75 g per one veneer surface. The three-layered plywood was hot pressed (180°C or 220°C) for 20 minutes at 2.0 MPa after cold pressing for 9 minutes at 2.0 MPa using a tabletop hot press (SA-302; Tester Sangyo Co., Ltd., Tokyo, Japan). Two distance bars (thickness 9 mm) were used for thickness control. All prepared plywood was stored at a constant temperature of 20°C and 65 percent relative humidity for $>$3 days.

### Mechanical tests

Eight shear strength test specimens (25 by 81 mm) were cut from five of the six panels, for a total of 40 specimens. According to the Japanese Agricultural Standard for plywood (Japanese Agricultural Standard Association 2020), dry and wet shear strength tests were conducted using 20 specimens for each test. The shear strength tests were performed under a load speed of 5 mm/min using a universal testing machine (RTC-1250A; Orientec Co., Ltd., Tokyo, Japan). After testing, shear strength was calculated as follows:

$$\text{Shear strength} = \frac{P_s}{S} \quad (1)$$

where $P_s$ is the maximum test load and $S$ is the bonding area.

Two bending test specimens (40 by 188 mm) were cut from five of the six panels, for a total of 10 specimens. A

*Table 1.—Manufacturing conditions of plywood samples.*

| Condition name | Pretreatment[a] | Cellulose nanofiber (CNF) | Pressing temperature (°C) |
|---|---|---|---|
| 180 (control) | –[b] | – | 180 |
| 220 (control) | –[c] | – | 220 |
| 180P | + | – | 180 |
| 220P | + | – | 220 |
| 180C | – | + | 180 |
| 220C | – | + | 220 |
| 180PC | + | + | 180 |
| 220PC | + | + | 220 |

[a] Before hot-pressing process, the veneer's surface was wet with running water for a minute and then scratched with a steel wire brush.
[b] –, without pretreatment or addition of CNF.
[c] +, with pretreatment or addition of CNF.

three-point bending test was performed at a span of 150 mm and a load speed of 10 mm/min using a universal testing machine (TCM-1000; Shinkom Nagano, Japan). After testing, two bending properties, modulus of rupture (MOR) and modulus of elasticity (MOE), were calculated as follows:

$$\text{MOR} = \frac{3P_bL}{2ah^2} \quad (2)$$

$$\text{MOE} = \frac{\Delta P}{\Delta\sigma} \times \frac{L^2}{4ah^3} \quad (3)$$

where $P_b$ is the maximum test load and $L$, $a$, and $h$ are the length, width, and thickness of the specimen, respectively. $\Delta P/\Delta\sigma$ is the slope of the initial straight-line portion of the load deflection.

## Density profile

Two density profile specimens (50 by 50 mm) were cut from one of the six panels. The density distribution was measured in the thickness direction using a commercial density profiler (DAX-6000; GreCon, Alfeld, Germany).

## Scanning electron microscopy

The interfacial surface of the plywood was observed with a scanning electron microscope (JSM-6000Plus; JEOL, Tokyo, Japan) after the shear strength tests. The observed surface was coated with gold at a target film thickness of 10 nm using a coating device (DII-29010SCTR; JEOL).

# Results and Discussion

## Plywood manufacturing

We were unable to produce plywood in conditions of 180 and 220. Under the pretreatment alone conditions, only 220P (220°C, pretreatment) plywood was successfully produced. The 180P (180°C, pretreatment) veneers were delaminated after hot pressing or during specimen cutting. A previous study reported that high moisture content on the bonding surface under hot-pressing temperatures of >180°C caused the denaturation of hemicellulose and lignin, resulting in self bonding (Ando and Sato 2010). Thus, it appears that pretreatment increases the contact area between the veneers and the moisture content enhances self-bonding strength, allowing the production of plywood. However, the development of self bonding was insufficient at 180°C, and we assumed that the veneers delaminated easily after fabrication. The 220°C temperature treatment provided higher self bonding than 180°C, as reported previously (Ando and Sato 2010). However, plywood was produced under all conditions with added CNF, regardless of pretreatment. Bonding strength develops when adding CNF because of hydrogen bonds and physical entanglement between the wood and CNF (Kojima et al. 2013, 2018, 2021). We speculate that the increased moisture content of the bonding surface resulted in the development of self bonding between the veneers, resulting in the production of plywood. The mean and standard deviation of the density of the plywood in this study was 0.37 ± 0.02 g/cm$^3$, regardless of the manufacturing conditions. As the plywood thickness was approximately 9 mm, compression ratio was equivalent to that of normal softwood plywood. Moreover, the plywood could be produced under all conditions with CNF addition, indicating that CNF can be used as a plywood binder. In the following sections, we evaluate various properties of the plywood prepared in this study.

## Mechanical properties

Figure 1 shows the relationship between the plywood preparation conditions and dry shear strength and wet shear strength. First, looking at the conditions under which one of the two treatments was applied, shear strength developed only at condition 180C (180°C, CNF added). At 220°C, the shear strength of the plywood of 220C (220°C, CNF added) was significantly higher than that of 220P. Therefore, we inferred that adding CNF contributed more than the pretreatment to improving the bonding strength. Shear strength was significantly higher under conditions of 180PC (180°C, pretreatment and CNF added) and 220PC (220°C, pretreatment and CNF added) than under conditions with only one of the treatments (180P, 180C, 220P, and 220C) at 180°C and 220°C. It is likely a result of the increased contact area between the CNF and veneer because of pretreatment, which led to increased hydrogen bonding and physical entanglement. Under both treatments, the shear strength in the 220PC condition was

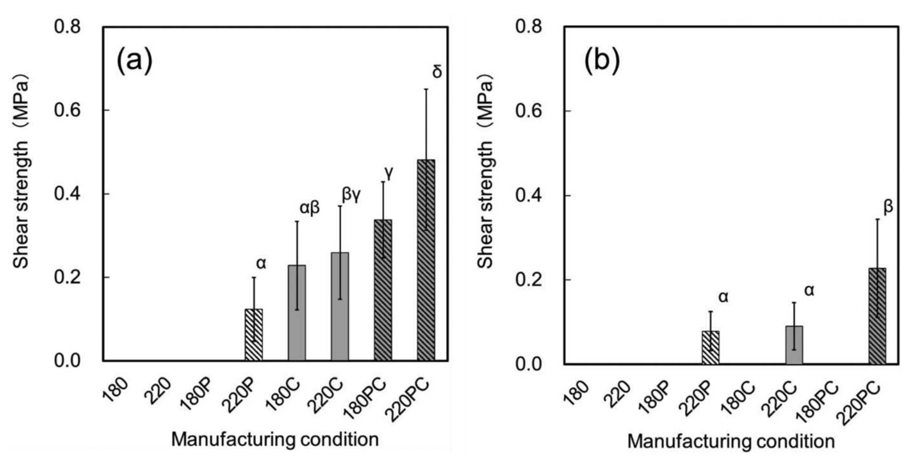


*Figure 1.—(a) Dry and (b) wet shear strength of plywood samples under different manufacturing conditions. Details of the manufacturing conditions are listed in Table 1. Vertical bars indicate standard deviation. Different letters indicate significant differences at a 5 percent significance level (Tukey's test).*

significantly higher than that in the 180PC condition. These results suggest that the combined effects of self-bonding strength due to the high hot-pressing temperature and CNF addition resulted in higher bonding strength.

Wet shear strength of all manufacturing conditions did not meet the type II softwood plywood of Japanese Agricultural Standards (0.4 MPa, Japanese Agricultural Standard Association 2020). Our wet shear strength testing results showed that the specimens delaminated during hot water soaking under all conditions at a hot-pressing temperature of 180°C (Fig. 1b). As 180C and 180PC, which developed strength in the dry shear strength test, also delaminated, we inferred that the hydrogen bonding and physical entanglement caused by adding CNF decreased after water immersion. In contrast, wet shear strength developed in 220PC, 220C, and 220P at 220°C as observed under the dry conditions. Therefore, the high self-bonding strength developed by hot pressing at 220°C persisted during the water soaking test. This result is consistent with a previous study (Kurokochi et al. 2019). The 220C treatment specimens were stronger than the 220P treatment specimens in the dry shear test (Fig. 1a), and the 220P and 220C treatment specimens were comparable with those in the soaking test (Fig. 1b). As the contact area between veneers was small, we assumed that the self-bonding strength of the 220C specimens was lower than that of the 220P specimens; however, the strength of the 220C and 220P specimens was similar to those in the soaking test. These results may have three possible explanations. First, CNF is stronger and denser than veneer, such that the presence of CNF on the bonding surface may have contributed to the increased strength. Second, cellulose does not denature at 250°C, but its crystallinity is affected by lower temperatures (Hayashi et al. 1975). Also, Bhuyan et al. (2000) reported that the crystallinity of cellulose powder and spruce increased for <1 hour of heat treatment and the increment was greater at higher temperatures (180 to 220°C). Thus, the presence of CNF and increased crystallinity may have resulted in the 220C treatment specimens having bonding strength comparable with that of the 220P treatment specimens. Third, self bonding occurred by the pyrolysis of hemicellulose and lignin in veneer. According to Chen et al. (2019) the first stage of thermal decomposition of hemicellulose starts at <220°C primarily because of dehydration. Then in the second stage in the temperature range of 210 to 340°C, the dissociation of *O*-acetyl side-chain fragmentation reactions happen to form acetic acid. Umemura et al. (2013) reported that the self-bonding mechanism of particleboards with citric acid is mainly due to the ester linkages between the carboxyl groups of citric acid and hydroxyl groups of wood components. There is a possibility that acetic acid derived from hemicellulose forms ester linkages between hydroxyl groups of the veneer or CNF. The 220PC treatment specimens exhibited the greatest strength under the wet condition. The combined effect of pretreatment and CNF addition persisted after the immersion test under dry conditions. It is likely due to the density and crystallinity effects mentioned above, either alone or combined, becoming more pronounced in both treatments, as well as the high self-bonding strength due to hot pressing at 220°C.

Figure 2 shows the MOR and MOE bending properties for each of the plywood fabrication conditions. Although MOR on 180PC and 220PC fulfilled the required conditions of E50-F160 structural plywood of Japanese Agricultural Standards, even the highest MOE obtained from 220PC was <10 percent of that requirement. MOR and MOE were similar for all conditions, with no significant difference in bending properties between 180°C and 220°C under the same treatment and CNF conditions. Thus, unlike shear strength, self bonding due to denaturation of the chemical components did not affect the bending properties. Bending properties of 180PC and 220PC were higher than those with one treatment condition alone (180C, 180P, 220C, and 220P). The reason for this increase in bending properties in association with simultaneous pretreatment and CNF is thought to be the same as that for the increase in wet shear strength. To discuss these properties in greater depth, we analyzed the layer density distribution and the fractured surfaces of the shear specimens.

## Distribution of layer density

Figure 3 shows the layer density distribution for the plywood. As two samples were measured and similar trends were observed, only one sample from each condition is shown in the figure. A small peak of 0.35 to 0.36 g/cm$^3$ was observed at the adhesive layer formed with pretreatment and without CNF

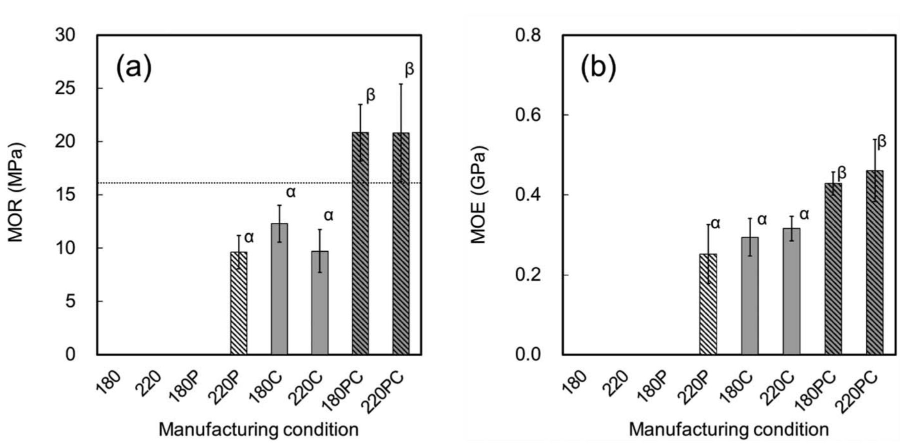


*Figure 2.—Bending properties of plywood samples under different manufacturing conditions. Details of the manufacturing conditions are shown in Table 1. (a) Modulus of rupture (MOR). (b) Modulus of elasticity (MOE). Vertical bars indicate standard deviation. Different letters indicate significant differences at a 5 percent significance level (Tukey's test). Dotted line indicates the required MOR of E50-F160 structural plywood of Japanese Agricultural Standards.*

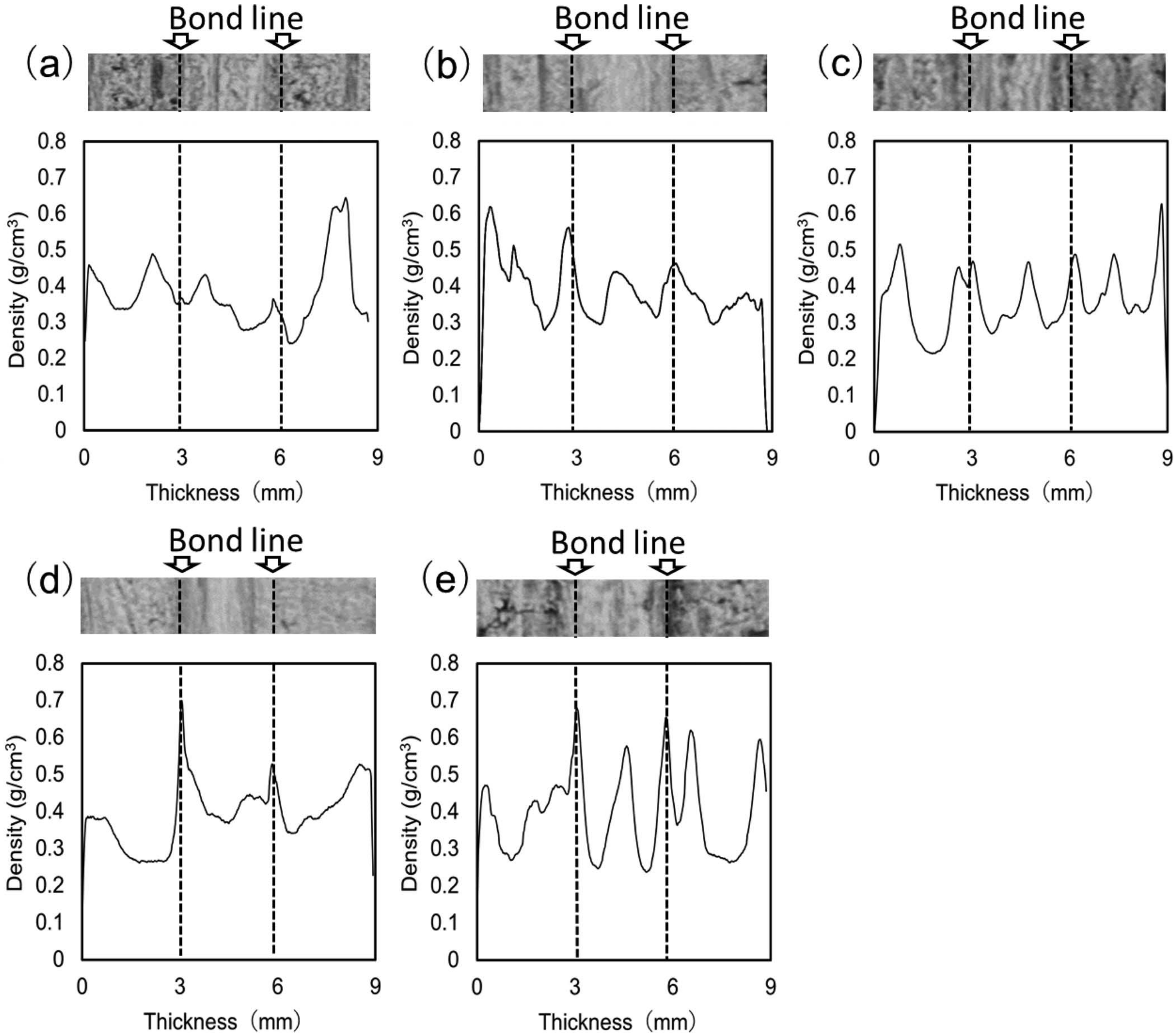


*Figure 3.—Distribution of layer density under conditions (a) 220P, (b) 180C, (c) 220C, (d) 180PC, and (e) 220PC (Table 1). Upper images are photographs of the specimens corresponding to each graph.*

(condition 220P, Fig. 3a), whereas for conditions 180C (Fig. 3b) and 220PC (Fig. 3e), a peak of 0.44 to 0.70 g/cm$^3$ was observed at the adhesive layer when CNF was added. The density of CNF is 1.5 g/cm$^3$, which is much higher than that of the veneer used in this test. Therefore, the high density obtained by adding CNF is considered a reasonable result. The peak densities of the adhesive layers in Figures 3b and 3c conditions 180C and 220C were 0.44 to 0.54 g/cm$^3$, and those with conditions 180PC and 220PC were 0.52 to 0.70g/cm$^3$ (Figs. 3d, 3e). These peak values increased with pretreatment because of the entanglement of CNF with veneer fluff caused by the pretreatment. Also, there is a possibility that the addition of water accompanied by spreading CNF accelerates the plasticization on the interface during pressing. Densification in the interface layer may have a positive effect on the mechanical properties. These results support a relationship between the mechanical properties associated with pretreatment and CNF and those associated with a lack of pretreatment and CNF. In addition, as bending properties are generally enhanced by including high-density, high-strength materials, the higher density of the adhesive layer may have contributed to the increased bending properties. In contrast, the densities of the 220PC and 180PC adhesive layers were almost the same, with significantly different dry shear strength. The densities of the adhesive layers prepared with and without pretreatment and with and without CNF were different, but there were no significant differences in wet shear strength or bending properties. These results suggest that self-bonding strength due to denaturation of the components cannot be determined from the layer density distribution.

Density peaks observed outside of the adhesive layer were due to the consolidation of the surface layer and the latewood portion of the veneer.

## Interfacial surface

Figure 4 shows scanning electron microscopy images corresponding to the specimen surfaces. Only the 220°C results are

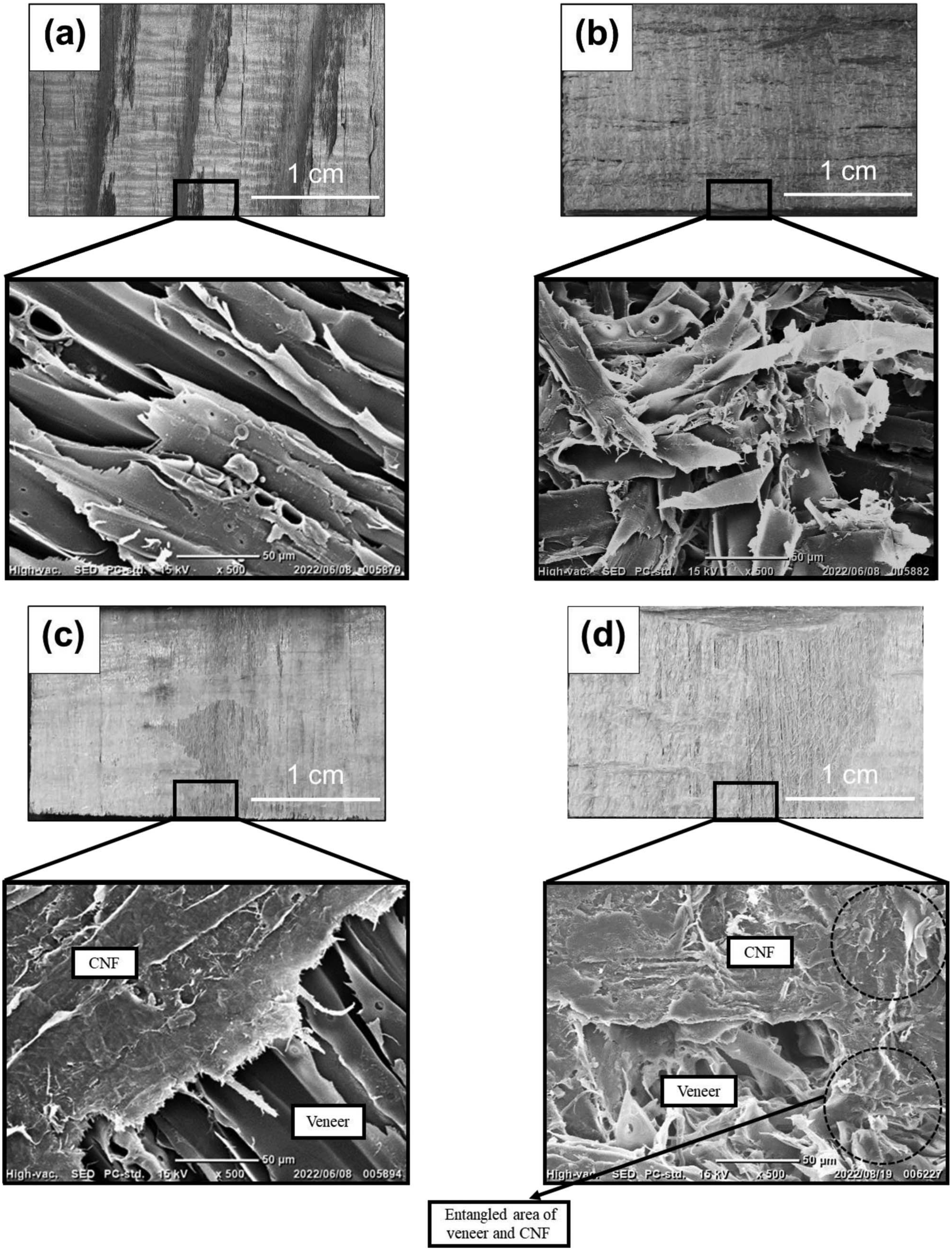


*Figure 4.—Representative interfacial plywood surfaces and corresponding scanning electron microscopy images (×500) for conditions (a) 220, (b) 220P, (c) 220C, and (d) 220PC (Table 1).*

shown, as no difference was observed between the 180°C and 220°C surfaces. Tracheids were uniformly aligned under the condition with no pretreatment and no CNF addition (condition 220, Fig. 4a), but the surface became rougher and the direction of the tracheids became more random under the condition with pretreatment and no CNF addition (condition 220P, Fig. 4b). This increase in surface area is thought to have contributed to the development of self bonding by increasing the number of bonding surfaces between the veneers. This finding is similar to that of a previous study (Kurokochi et al. 2019). Filmlike CNF covered the veneer under the condition without pretreatment and with CNF addition (condition 220C, Fig. 4c), whereas veneers with increased surface area and filmlike CNF formed under the pretreatment and CNF condition (220PC, Fig. 4d). The CNF and veneers were observed to be entangled. Therefore, pretreatment increased the contact area between the veneer and CNF, and their complex entanglement resulted in the formation of a high-density adhesive layer. These observations are consistent with the results and discussion of mechanical properties, as well as the layer density distribution data.

## Conclusions

In this study, whether domestic softwood plywood could be produced with CNF was determined. Also, the effects of different hot-pressing temperatures during fabrication and the presence or absence of wetting and scratching treatment on the veneer surfaces were examined.

Plywood containing CNF had higher dry shear strength than plywood produced with pretreatment alone. Shear strength was significantly higher under conditions of 180PC and 220PC than under conditions with only one of the treatments (180P, 180C, 220P, and 220C) at 180°C and 220°C. Pretreatment improved the bending properties, whereas pressing temperature did not affect the bending properties. MOR on CNF-supplemented plywood with pretreatment fulfilled the required conditions of E50-F60 structural plywood of Japanese Agricultural Standards. It is likely a result of the increased contact area between the CNF and veneer due to pretreatment.

The results of this study suggest that CNF is useful as a plywood binder. However, the veneer delaminated at the adhesive layer on some samples instead of producing a bending fracture. Moreover, except for MOR on 180PC and 220PC, mechanical properties did not meet Japanese Agricultural Standards. Therefore, the adhesive strength of CNF was insufficient, and future studies are needed to increase the adhesive strength of CNF by varying the type and amount of CNFs added. Also, in this study, 9 minutes of cold pressing and 20 minutes of hot pressing were applied to remove water inside the plywood since CNF slurry contains considerable water. Moubarik et al. (2009) has reported that five layers of plywood of 10-mm thickness bound by cornstarch and tannin in phenol–formaldehyde resins were manufactured in 6 minutes. Pressing is a key factor to determine the production speed. Shortening the pressing time is one of the issues to improve for practical application.


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