

Cellulose Nanofiber as Complete Natural Binder for Particleboard

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Abstract

The demand for wood-based materials as an alternative to plywood is increasing, and a synthetic adhesive is typically used for the fabrication of such materials. The identification of replacements for standard adhesives poses a major challenge. In this study, we investigated the mechanical properties of particleboard bonded by cellulose nanofiber (CNF) and compared the mechanical properties of particleboard bonded by CNF with those of particleboard bonded by adhesive. CNF was added together with wood particles during the fabrication process. The target CNF addition amounts were 0, 3, 5, 10, and 20 weight percent. The bending and internal bond strengths of the particleboard with CNF increased, and its water absorption decreased as CNF content increased. The comparison between the boards prepared with adhesives and those with CNF showed that the properties of boards prepared with 20 weight percent CNF were comparable to those of boards prepared with 1 weight percent synthetic adhesive.

Wood-based materials are increasingly in demand, and their production is also increasing. For example, plywood is in high demand as a building material, while particleboard and medium-density fiberboard (MDF) are often used as furniture material. However, particleboard and MDF are expected to be useful as alternatives to plywood. Recently, the contents of particleboard and MDF for construction were added to the Japanese Industrial Standards (JIS) under JIS A 5903 and 5908 (JIS 2014, 2015). A synthetic adhesive is typically used for the production of wood-based materials. The most common synthetic adhesives used in the wood-based materials industry are urea-formaldehyde (UF) and phenol-formaldehyde (PF) resins. UF resins have many advantages, including low cost, ease of use under a wide variety of curing conditions, low cure temperatures, water solubility, and so on. UF is usually used in furniture products. The PF resins are more expensive than UF resins; however, they are more popular due to their high water resistance, which makes them suitable for outdoor applications. The major drawbacks to these adhesives are that some of them contain volatile organic compounds that are harmful to human health, and the final products are difficult to recycle. Thus, considering their use of synthetic adhesives, wood-based materials are not necessarily environmentally friendly.

For this reason, some studies have focused on the development of natural, material-based wood adhesives using bioresources. For example, some natural adhesives are composed of citric acid (Umemura et al. 2012a, 2012b, 2012c, 2013) or lactic acid (Ikeda et al. 2008, Takatani et al. 2008), but these have not yet been put to practical use. In this study, we explored options involving nanotechnology. Nanotechnology has been rapidly developing in many fields.

A “nanofiber” is an object with two similar external dimensions in the nanoscale, with a length range of approximately 1 to 100 nm and a third dimension that is significantly larger (ISO 2011). There are many types of nanofibers. However, cellulose nanofiber (CNF) has received attention in numerous fields worldwide.

CNF is a nanofiber composed predominantly of cellulose (ISO 2017). Cellulose is the most abundant organic compound derived from biomass. It is estimated that about 10 billion tons of cellulose are produced each year (Azizi et al. 2005). CNF has been reported to have better physical and mechanical properties compared with other fibers (Nishino et al. 2004, Iwamoto et al. 2009, Heath and Thielemans 2010, Saito et al. 2011). The use of CNF has been explored in a variety of materials, such as CNF films and CNF nanocomposites (Iwamoto et al. 2005, Shams et al. 2011). Of the various uses for CNF, the combination of CNF and paper is of particular interest. CNF improves the mechanical and gas barrier properties of paper, and this combination is

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expected to be applied in fields such as food packaging and printing (Bardet and Bras 2014). The properties of paper can be improved by the addition of CNF without any special treatment because both pulp and CNF are hydrophilic, which is highly advantageous for the commercial application of this combination. Because the material in wood-based panels is also hydrophilic, the performance improvement achieved using CNF in wood-based panels is expected to be similar to that achieved with paper.

In a previous study, we investigated the addition of CNF to wood-based panels as an alternative to adhesives. We previously reported that the mechanical properties of wood flour board and fiberboard were enhanced by the addition of CNF in certain amounts (Kojima et al. 2014, 2015, 2016). For example, the bending strength of the wood flour board without CNF at target density of 1.0 g/cm³ was 4.5 MPa, and it increased to 7.7, 13.7, and 8.2 MPa with addition of 5, 10, 15 percent CNF, respectively (Kojima et al. 2013). Previous experiments by our group showed the improvement of wood-based panels by CNF in panels composed of relatively small elements, such as wood flour and fiber. However, it is not clear whether CNF is able to improve the properties of particleboard, which is composed of larger elements. It has previously been reported that boards made with 2.5 cm-long flakes were the strongest in all respects compared with boards that were made from 0.6 cm-long flakes, planer shavings, fines, and sawdust (Heebink and Hann 1959). It is necessary to determine the properties of particleboard using CNF as binder.

Therefore, the purpose of this study is to investigate the reinforcement effects achieved by using CNF in particleboard, which is composed of larger elements than the materials used in previous studies. In addition, few studies have focused on the comparison between wood-based panels produced using commercial adhesive and those produced with CNF. It is important to compare adhesive and CNF to examine whether CNF is useful as a binder in the production of wood-based panels. Thus, this article presents the results obtained by fabricating particleboard using CNF or adhesive and comparing the properties of these boards.

Materials and Methods

Materials

Recycled particles (Okura Industrial Co., Ltd.) were used to fabricate particleboards. Figure 1 shows a representation of the particles used in this study. Sieve screen weight data analysis was used to characterize the shape of particles, as previously performed by Sackey and Smith (2009). Particles were screened with a sieve shaker (VSS-50, Tsutsui Scientific Instrument Co., Ltd.) into five different size classes, 4.7, 8.6, 16, 30, and 36 mesh. First, 50 g of the particles were placed on top of the sieve and shaken for 30 minutes. Then, the weight of the particles retained on each sieve after shaking was measured. We repeated this measurement three times and calculated the mean weight on each sieve after shaking. The mean weight percentage of particle sizes is shown in Table 1. The largest mass fraction was collected in the 16 mesh sieve. When the mass fraction of 16 mesh and 8.6 mesh was combined, the width or thickness of 86 percent of particles was 1 to 4 mm.

CNF (BiNF-i-s WMa-10010, Sugino Machine, Ltd.) was used as a 10 weight percent water suspension. The nominally average fiber diameter was 10 to 50 nm, and

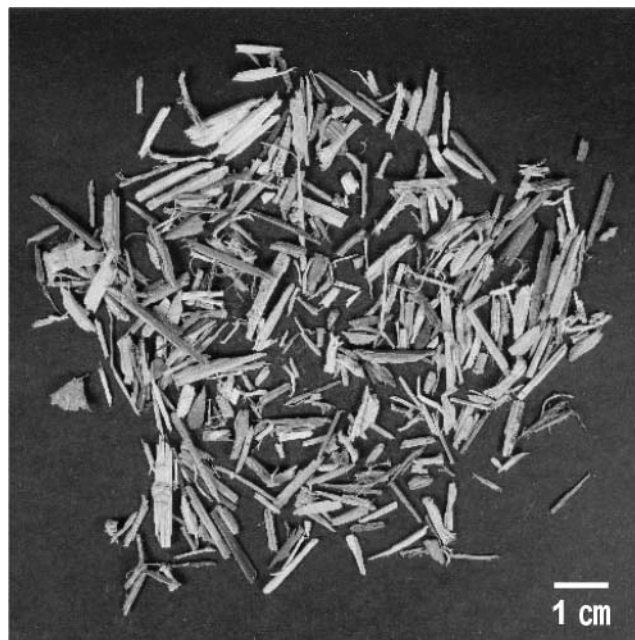


Figure 1.—Representative particle shape used in this study.

the nominally average viscosity was 110 Pa s. The CNF suspension was subjected to tert-butyl alcohol (t-BuOH) displacement and freeze-dried to prevent aggregation. Then, the surface morphology of the freeze-dried CNF was observed with a scanning electron microscope (SEM; JSM-6510LV; JEOL, Ltd.). Urea-formaldehyde resin (UF), with solids content of 65 percent, viscosity of 0.23 Pa s, and Ph of 7.2 (TB-86, Oshika Co., Ltd.); and phenol-formaldehyde resin (PF), with solids content of 40 percent, viscosity of 1.1 Pa s, and Ph of 11.8 (PR-9800, Sumitomo Bakelite Co., Ltd.) were used as the commercial adhesives to compare with CNF.

Fabrication of particleboard with CNF

The CNF was diluted to 3 weight percent. The CNF suspension was added to wood particles at 0 (control), 3, 5, 10, and 20 weight percent and mixed by hand. The preparation containing 20 weight percent CNF contained 491 g water, and the mat moisture content became 323 percent. Therefore, to adjust moisture content, supplementary water was added to the preparations containing 0, 3, 5, and 10 weight percent CNF. The mixture was then poured into a wooden forming box with internal dimensions of 150 by 150 mm, and a hand-formed mat was made. The mats were pressed for 15 minutes at 180°C and 2.4 MPa using a

Table 1.—Weight percentage of each particle size class (%).

Mesh size ^a	Weight percentage (%)
4.7(+)	4.02
8.6(+)	31.47
16(+)	54.12
30(+)	10.12
36(+)	0.12
36(–)	0.06

^a Mesh size opening: (+) particles retained on the sieve; (–) particles pass through the sieve. Each mean is the result of $n = 3$ sample bags with each bag containing 50 g particle.

hot press (Tabletop Test Press SA-302; Tester Sangyo Co., Ltd., Japan). Two metal bar stops with a thickness of 9 mm were used for the thickness control. Particleboards with dimensions of 150 by 150 by 9 mm and a density of 0.75 g/cm³ were fabricated. For all experiments, four fiberboards were produced for each condition. All boards were conditioned at 20°C and 65 percent relative humidity for at least 3 days before testing. Adhesives were not used.

The changes in mat temperature during hot pressing were measured at the center of the mat using a thermocouple sensor (Type T/copper-constantan) and a data logger (midi Logger GL200A, Graphtec). This measurement aimed to clarify the influence of CNF addition on temperature behavior inside the mat during hot pressing. Measurement started when the top plate of the hot press contacted the mat surface, and it finished when the temperature at the core reached the temperature of the press (180°C in this study). Temperature measurements were carried out for the boards prepared using 0 and 10 weight percent CNF. As discussed above, the mat moisture content in both conditions was adjusted to 323 percent.

Fabrication of particleboard with commercial adhesive

The particles were sprayed with UF and PF resin in a rotating drum blender at a resin content of 1, 3, and 5 weight percent based on the air-dried weight of the particles. No supplementary water was added to adjust the moisture content to the board. The procedures of forming and hot pressing were the same as described above.

Physical testing

After conditioning, 10 pieces (120 by 25 mm in size) were cut from each board for use in a three-point bending test with a universal testing machine. The following conditions were imposed: a span of 100 mm and a loading speed of 3 mm/min. The modulus of rupture (MOR) and modulus of elasticity (MOE) were calculated. After the bending test, two pieces (25 by 25 mm) were cut from edge parts of the bending test specimen for internal bond strength (IB) and water adsorption tests. The IB test was performed under a loading speed of 3 mm/min. Water adsorption was determined by measuring the weight and thickness of the pieces before and after soaking in water at 20°C for 24 hours. Ten samples were used for each test. In the control boards, the water adsorption test conditions would have caused these specimens to disintegrate because the boards were fragile since they were fabricated without any bonding agent. Therefore, the water absorption test was not carried out in these boards. A commercial density profiler (DAX-6000, GreCon) based on an X-ray system was used. Specimens had dimensions of 50 by 50 mm.

Results and Discussion

Binding effects of CNF in particleboard

Figure 2 shows the measurements of the change in mat temperature during hot pressing in the 0 and 10 weight percent CNF boards. In both boards, after the platen reached the mat surface, the core temperature increased rapidly to approximately 100°C and then remained constant for a period of time. After that, the core temperature increased slowly toward the 180°C platen temperature. This behavior is similar to the previously reported typical core temperature change observed during hot pressing (Rofii et al. 2014). However, there were some differences in the temperature

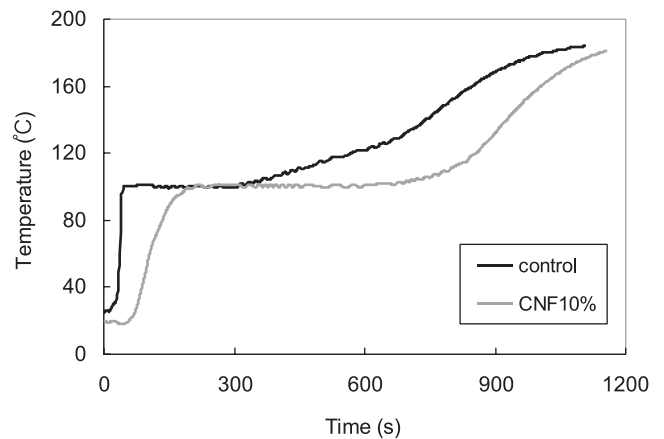


Figure 2.—The core temperature change curve as influenced by cellulose nanofiber (CNF).

changes in the 0 and 10 weight percent CNF boards. First, the time point when the core temperature reached 100°C was approximately 100 seconds in the case of the 0 weight percent CNF board and approximately 200 seconds for the 10 weight percent CNF board. Therefore, the presence of CNF caused a slower increase in core temperature. Second, the duration of time when the core temperature remained constant in the case of the 0 weight percent CNF board was 250 seconds, whereas this duration in the 10 weight percent CNF board was approximately 500 seconds. It is likely that the CNF absorbs a large quantity of water and adheres to wood particles, so little water flows out of the preparation at the time of mat formation, resulting in vaporization of this water during hot pressing. In the case of the board without CNF, some water flowed out after formation of the mat.

Pictures of the fabricated particleboards are shown in Figure 3. The particleboard without CNF was fragile, and it was difficult to cut the edges finely because no binding agent was added. By contrast, it was possible to make fine edge cuts in the particleboard prepared with 10 weight percent CNF. Moreover, the wood particles in the particleboard prepared with 10 weight percent CNF became whitish in color because the aggregates of CNF adhered to the particles. The thicknesses of the board for 0, 3, 5, 10, and 20 weight percent were 9.0, 8.9, 8.9, 8.8, and 8.8 mm, respectively. The thicknesses of the boards were same as target thickness (9 mm). The densities of the board for 0, 3, 5, 10, and 20 weight percent were 0.57, 0.67, 0.67, 0.68, and 0.69 g/cm³, respectively. The density without CNF was lower than the others because the weight of board without CNF was lower due to some particles falling off the board during cutting.

Figure 4 shows the results of the vertical density profiles of the boards prepared with CNF. In the board without CNF, it was difficult to cut out a sample; thus, no vertical density profile test was carried out. The profiles of the CNF-added boards show two peak densities in the outer layers of the board, with the lowest density in the core layer. This tendency was similar to that of conventional particleboards with synthetic adhesive. In these fabricated particleboards, the mat moisture content before hot pressing was high due to the presence of CNF. The surface layer was compressed sufficiently, but the heat transfer to the core was delayed, decreasing the compression, which may have resulted in the observed density distribution. In addition, the densities in the outer layers increased as the CNF content increased. The

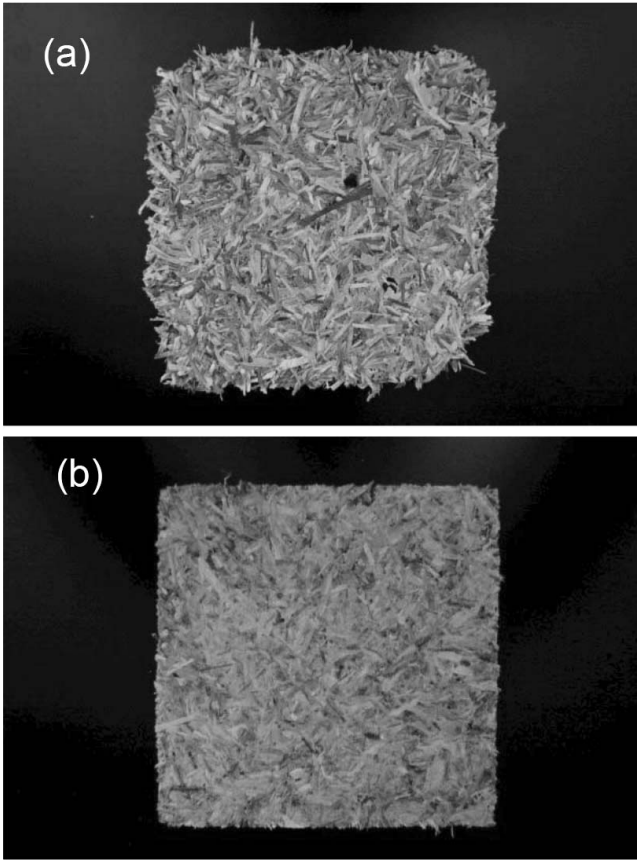


Figure 3.—Manufactured board (a) control (no binder), (b) cellulose nanofiber (10 wt%).

maximum density in the 3 weight percent CNF board was 0.78 g/cm^3 , whereas it reached 0.90 g/cm^3 in the 20 weight percent CNF board. This was attributable to aggregates of CNF that adhered to the wood particles, as shown in Figure 3. In the board prepared using 20 weight percent CNF, aggregates of CNF appear to not only cover the board surface but also fill the space between wood particles on the board surface. Therefore, no gaps were present on the board surface, resulting in the high density of the surface layer.

Figure 5 shows the relationship between the bending properties and CNF content of the fabricated particleboards. These results indicate that the MOR and MOE of the boards with CNF added were improved. It appears that the interparticle bond strength of the particleboard was enhanced by the addition of CNF, since CNF adhered to the wood particles, became entangled in the surface of the particles, and filled in between particles as aggregates. Furthermore, the MOR and MOE gradually increased with increasing CNF content from 3 to 20 weight percent. The specific MOR (MOR/mean board density) was $0.34 \text{ MPa}/(\text{g/cm}^3)$; without CNF, however, it was increased to 2.94 , 3.44 , 4.40 , and $9.01 \text{ MPa}/(\text{g/cm}^3)$ by adding 3, 5, 10, and 20 weight percent CNF, respectively. It is obvious that the increasing MOR is not due to the density.

Figure 6 shows the relationship between the IB and CNF contents of the fabricated boards. Similar to the findings shown for bending properties, the IB of the boards with CNF added was improved. This result also indicates that the interparticle bond strength of the boards was enhanced by the

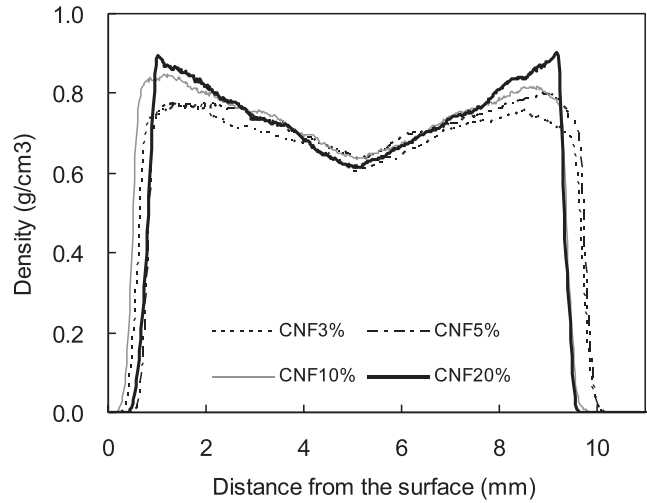


Figure 4.—Vertical density profiles as influenced by cellulose nanofiber (CNF) content.

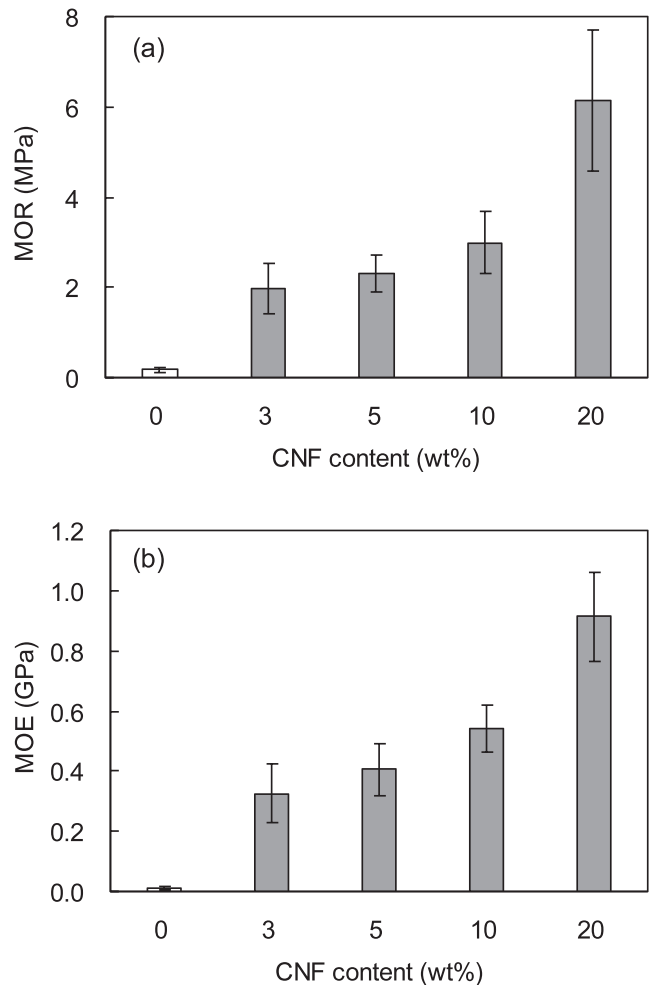


Figure 5.—Bending properties of particleboards containing cellulose nanofiber (CNF). (a) Modulus of rupture (MOR), (b) modulus of elasticity (MOE). Vertical bars indicate standard deviations.

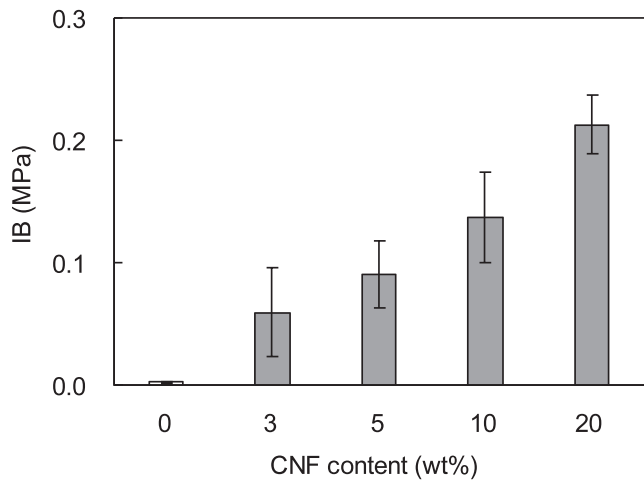


Figure 6.—Internal bond (IB) strength of particleboards containing cellulose nanofiber (CNF). Vertical bars indicate standard deviations.

addition of CNF. The IB gradually increased with increasing CNF content from 3 to 20 weight percent.

Figure 7 shows thickness swelling (TS) and weight change (WC) measured in the water absorption test. The

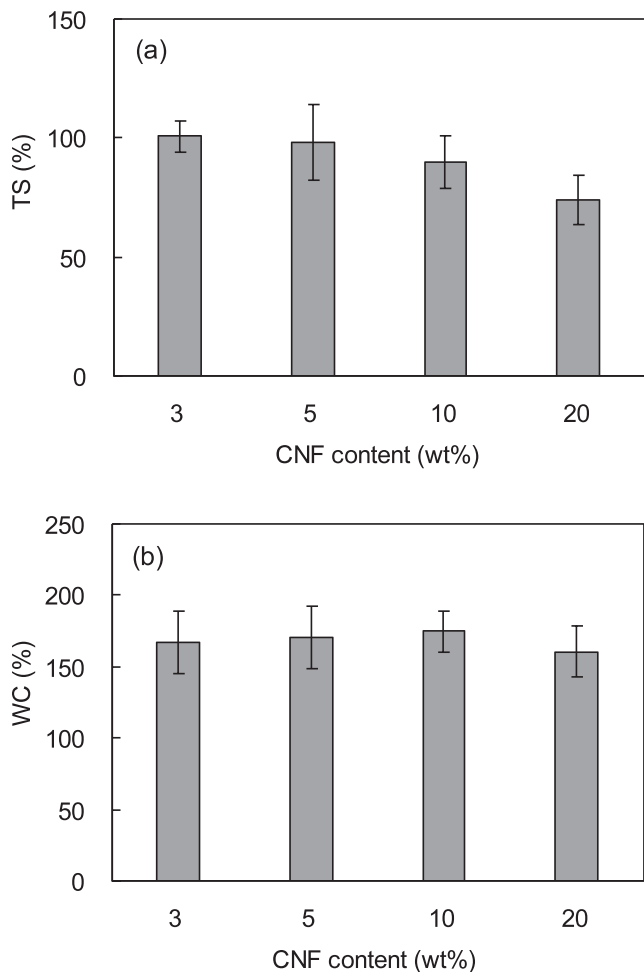


Figure 7.—Thickness swelling and weight change measured with the water adsorption test. (a) thickness swelling (TS), (b) weight change (WC). Vertical bars indicate standard deviations.

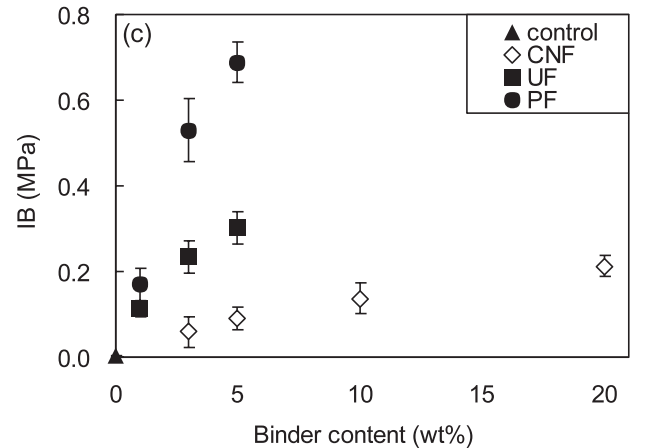
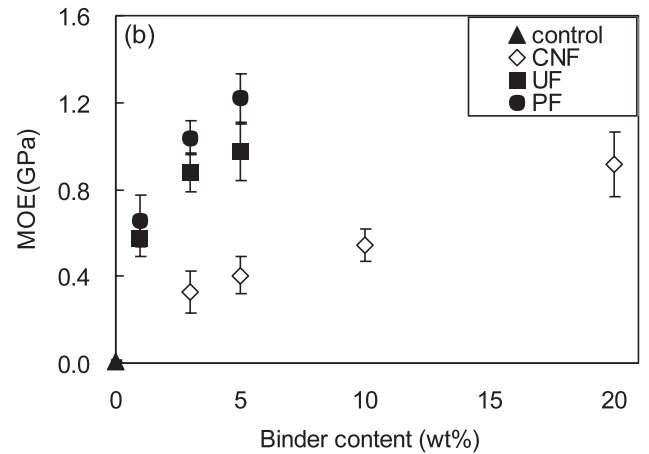
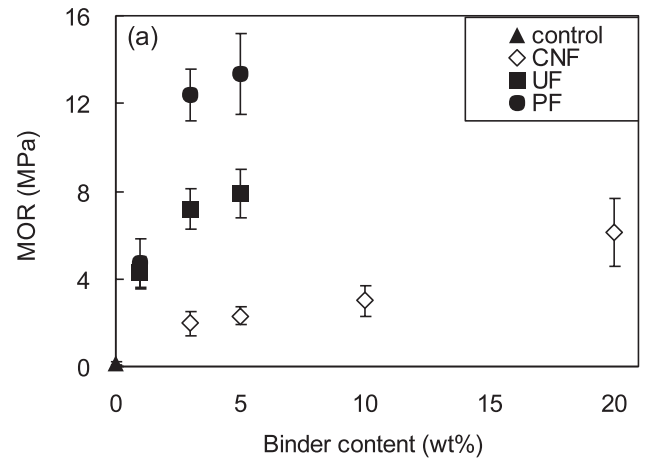


Figure 8.—Bending and internal bond (IB) strengths of particleboards containing cellulose nanofiber (CNF) or adhesives. (a) Modulus of rupture (MOR), (b) modulus of elasticity (MOE), and (c) IB. Vertical bars indicate standard deviations. UF = urea-formaldehyde resin; PF = phenol-formaldehyde resin.

water absorption test was not performed for the 0 weight percent CNF board because the specimen was disintegrated by the water soaking. By contrast, the samples prepared with CNF maintained their shapes even after water absorption, which suggests that the CNF had a reinforcing

effect on the boards. The TS decreased with increasing CNF content from 3 to 20 weight percent. It appears that CNF enhances entanglement at the surface of the particles. The WC results showed no significant differences based on the amount of CNF added to the boards. Therefore, TS decreased as the amount of added CNF increased, whereas WC did not decrease. Because the CNF and the particles are both hydrophilic materials, no difference was found in WC because the amount of water absorbed was about the same; however, due to particle entanglement and hydrogen bonding caused by CNF, the TS decreased with the increase in added CNF.

The MOR, IB, and TS of the board with 20 weight percent CNF, which had the best mechanical properties in this study, were 6.1 MPa, 0.21 MPa, and 74 percent, respectively. According to JIS A 5908, the standard values of MOR, IB, and TS to use as manufactured material were 13.0 MPa or more, 0.20 MPa or more, and 12 percent or less (JIS 2015). The IB of the board with CNF was able to exceed the standard value, but the MOR and TS was not. The MOR and TS must be improved for practical use.

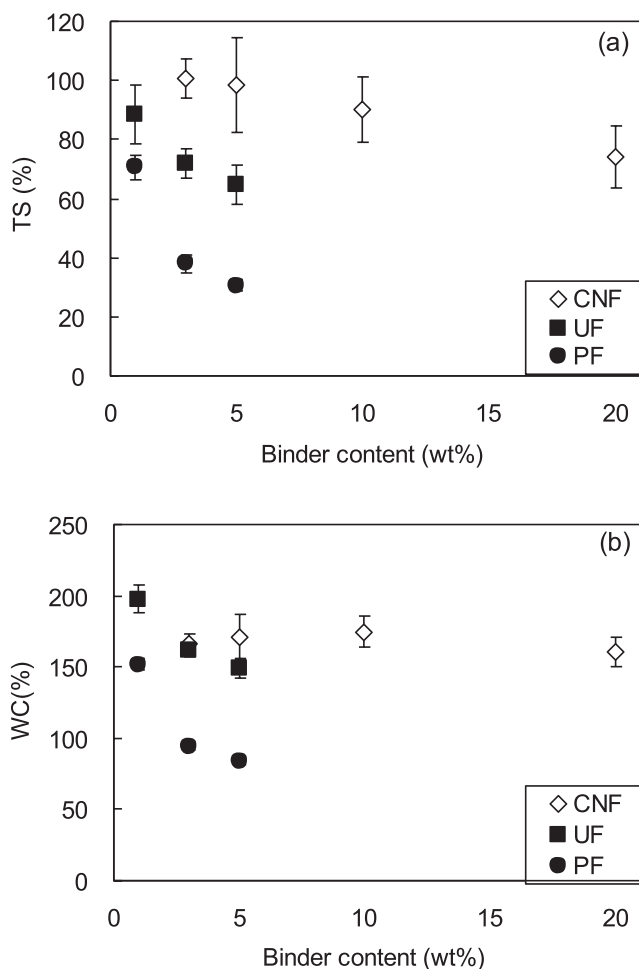


Figure 9.—Thickness swelling and weight change of particleboards containing cellulose nanofiber (CNF) or adhesives. (a) thickness swelling (TS), (b) weight change (WC). Vertical bars indicate standard deviations. UF = urea-formaldehyde resin; PF = phenol-formaldehyde resin.

Comparison of particleboards fabricated using CNF and adhesive

Figure 8 shows the evolution of the MOR (Fig. 8a), MOE (Fig. 8b), and IB (Fig. 8c) of the boards as a function of the percentage of CNF or the percentage of two kinds of resin (UF and PF). The mechanical strength of the board increased with the increasing amounts of CNF or resins. In addition, the mechanical properties of the board with 20 weight percent CNF was higher than those with 1 weight percent UF or 1 weight percent PF.

Figure 9 shows the evolution of the TS (Fig. 9a) and WC (Fig. 9b) of the boards as a function of the percentages of CNF or two kinds of resin (UF and PF). Like the mechanical properties, the water-resistance properties of the board with

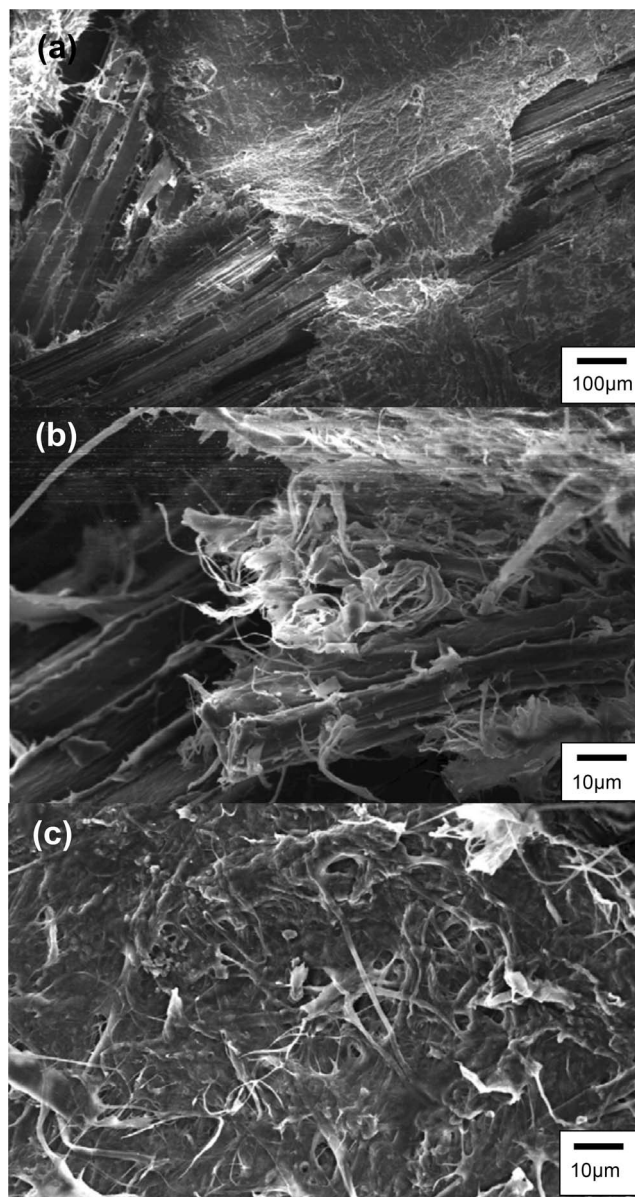


Figure 10.—Scanning electron microscopy photographs of the particleboard with cellulose nanofiber (CNF). (a), (c) Aggregated CNF formed a nonporous membrane-like structure, (b) CNF entangled the surface of wood particles.

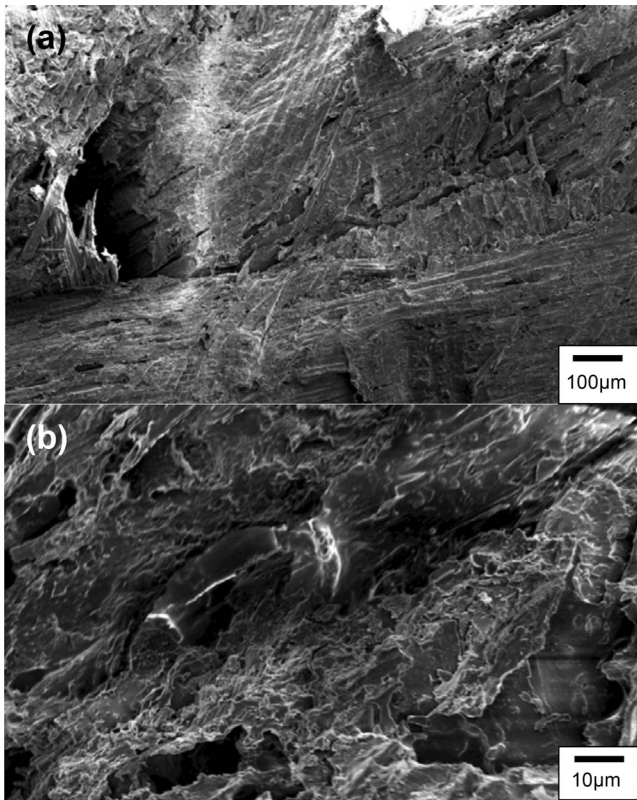


Figure 11.—Scanning electron microscopy photographs of the particleboard bonded by urea-formaldehyde resin. (a) Wood particle covered with cured adhesives, (b) a different structure from the cellulose nanofiber aggregates formed by cured adhesive.

20 weight percent CNF were higher than those of boards with 1 weight percent UF or PF. Therefore, it is suggested that the properties of the board with 20 weight percent CNF corresponded to those of the boards with 1 weight percent UF or PF.

Figure 10 shows SEM images of the board with CNF. As can be seen in Figure 10a, aggregates of CNF covered the wood particles. The CNF in the board did not exist as individual fibers with nanoscale diameters, but as large aggregates. Figure 10b shows that CNF entangled the surface of wood particles. Figure 10c shows that aggregated CNF formed a “nonporous membrane-like” structure. The aggregates are attributed to the physical entanglement and hydrogen bonding of CNF.

Figure 11 shows SEM images of the board using UF as binder. Figure 11a shows that cured adhesive covers the wood particles, and Figure 11b shows that cured adhesive forms a different structure from the CNF aggregates. The production of particleboard with 20 weight percent CNF is too costly to be practical, even if this particleboard achieved higher performance than particleboard with 1 weight percent UF or PF. Consequently, the application of CNF alone as a reinforcing material for particleboards is not realistic. Further investigation should be conducted to determine practical uses for CNF as an alternative to synthetic adhesive in particleboard.

Conclusions

In this study, the mechanical properties of particleboard bonded by CNF were investigated and compared with those of particleboard bonded by adhesive. CNF improved the particleboard properties in a manner similar to previous studies conducted using wood flour boards and fiber boards. The improvement was more prominent as the amount of CNF in the boards increased. It appears that the interparticle bonds of the board were enhanced by CNF addition because CNF adhered to wood particles, became entangled with the surface of the particles, and filled in between particles as aggregates. The comparison between the adhesives and CNF showed that the properties of the board with 20 weight percent CNF corresponded to those of the boards with 1 weight percent UF or PF. However, the production cost of particleboard with 20 weight percent CNF is too high to be practical. Further investigation should be conducted to determine practical uses for CNF as an alternative to synthetic adhesive in particleboard fabrication.

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