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ORIGINAL ARTICLE

Binding effect of cellulose nanofibers in wood flour board

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Abstract Wood-based materials are extensively used for residual construction worldwide, especially in Japan. Most wood-based materials are fabricated using adhesives, some of which are not environmentally friendly. As an alternative to chemical adhesives, we explored this issue using nanofiber technology, especially the use of cellulose nanofibers (CNF), as reinforcement in wood flour (WF) board to replace chemical adhesives. We found that CNF could be easily made by pulverization in a ball mill. The physical and mechanical properties of WF board were improved by the three-dimensional binding effects of the CNF.

Keywords Wood-based materials · Cellulose · Nanofiber · Binder · Pulverize

Introduction

Wood-based materials are extensively used for residual construction worldwide, especially in Japan. These materials can be made from virgin or recycled wood, and most are fabricated using adhesives. Various kinds of adhesives are used, some of which are not environmentally friendly, and finding replacements for such chemical adhesives poses major challenges.

Natural materials (e.g., citric acid) have been used as binders [1]. In this research, we explored what nanofiber technology could offer. Nanotechnology is developing rapidly in many disciplines. In general, the term nanofiber refers to a nano-sized fiber and is defined as a fibrous material with a diameter of about 1–100 nm and a length more than 100 times the diameter. A fiber that has a surface and inner structure controlled at the nanoscale is called a nanostructured fiber [2]. This is true even for fibers having diameters exceeding 100 nm.

Extensive research has been conducted concerning the development and applications of cellulose nanofiber (CNF). Over a trillion tons of it exist worldwide. CNF is well known to have better physical and mechanical properties compared to other fibers [3]. Development of new materials using CNF has been conducted in many fields. For example, in relation to polymer composites, Okubo et al. [4] fabricated composites using polylactic acid and bamboo fibers extracted by steam explosion method with fibrillated cellulose from wood pulp as an enhancer. However, to the author’s knowledge, the application of CNF technology to wood-based materials has not been reported. If CNF could be used as a binder, wood-based materials could be manufactured without using chemical adhesives, and thereby become more environmentally friendly.

In this study, we focused on wood flour (WF) board, which is a simple wood-based material. The utility of CNF as a binder was explored by mixing it with the WF. A method was developed to make CNF using a ball mill, and its morphology was studied. The binding effect of CNF with WF was investigated as a function of CNF content and its particle size.

Materials and methods

Materials

The cellulose used in this study was commercial cellulose powder (KC Flock W100GK; Nippon Paper Chemicals

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Co., Ltd., Japan). WF board was made from WF (ARBOCEL®C100; Rettenmaier Japan Co., Ltd., Japan) having an average particle size of about 100 μm.

Pulverization of cellulose

Cellulose powder (13.5 g) was mixed with distilled water (200 g) and fibrillated using a ball mill (Pulverisette 6; Fritsch Japan Co., Ltd., Japan). The degree of pulverization was controlled by the time and rotational rate of the ball mill. The pulverizing time was set at five levels: 1, 2, 4, 8, and 16 h. The rotational rate of the ball mill was set at three levels: 150, 200, and 250 rpm. Fifteen different CNF slurries were studied. A slurry made from untreated (non-pulverized) cellulose powder was the control. The particle size of the CNF after ball milling was measured using a laser diffraction particle-size distribution analyzer (Partica LA-950; Horiba, Ltd., Japan). To prevent flocculation, the CNF slurry was replaced by alcohol, and CNF powder was manufactured. The surface morphology of the CNF powder was observed with a scanning electron microscope (SEM) (TM1000; Hitachi, Japan).

Fabrication of WF board

WF boards were made from mixtures of WF and the 16 CNF slurries noted above. This was done to study the effect of CNF form on the physical and mechanical properties of the WF boards. A single composition was examined, i.e., 80 wt% WF + 20 wt% CNF. WF (54 g, dry weight) was mixed with a CNF slurry (CNF 13.5 g + distilled water 200 g) in a polyethylene bag, and a hand-formed mat (15 cm × 15 cm) was made using a metal frame. Wire screens were placed on the upper and lower surfaces of the formed mat to accelerate water transfer during subsequent pressing. The hand-formed mats were pressed for 15 min at 120 °C at 2.4 MPa using a hot press (Tabletop Test Press SA-302; Tester Sangyo Co., Ltd., Japan). WF boards 15 × 15 × 0.3 cm having a density of 1.00 g/cm^3 were manufactured.

The relative amounts of CNF and WF were varied to study the effect of composition on the physical and mechanical properties of the WF board. The mixing condition was fixed at 250 rpm/2 h. Three WF:CNF compositions were evaluated, namely, 80:20, 90:10, and 95:5.

WF boards were also made with untreated cellulose powder and without CNF or cellulose powder using the above method. Two boards were produced for each condition. All boards were conditioned at 20 °C and 65 % relative humidity for at least 2 weeks before testing.

Because the purpose of this study was to clarify the binder effect of CNF, no adhesives or other additives were used.

Physical testing

After conditioning, six pieces measuring 12 × 2 cm were taken from each board for a three-point bending test. The following conditions were used: span, 10 cm; loading speed 1 cm/min. The modulus of rupture (MOR) and modulus of elasticity (MOE) were calculated.

Water absorption was determined by measuring the change in weight and thickness of the pieces before and after soaking in water at 20 °C for 24 h. An unstressed part of the bending test specimen was used for this test.

Results and discussion

Influence of CNF form

Table 1 shows the median particle size after ball milling. Smaller particle sizes were obtained with longer pulverizing times at the same rotational rate. For a fixed pulverizing time, a higher rotational rate gave a smaller particle size. The smallest particle size was obtained under the 250 rpm/16 h condition, and this particle size was about one-fifth that of the untreated cellulose powder. Fibrillation increased when the pulverization was done at high moisture content because of fiber swelling. Fibrous cellulose was produced because the water entered between cellulose fibers, and hydrogen bonding between the cellulose fibers was disrupted [5]. Initially, the fibers simply separated and cellulose with many fibrils was produced, but fiber breakage eventually occurred. Consequently, the particle sizes for 150 rpm/1 h and /2 h shown in Table 1 were larger values than control size. Moreover, the fiber size decreased with increasing pulverizing time. This trend was more evident at higher rotational rates of the ball mill. The morphology of the CNF was studied with the SEM. Figure 1 shows SEM photographs of the cellulose powder before and after ball milling (250 rpm/1 h). Figure 1c shows an area under higher magnification. The surface of the untreated fiber was very smooth, while a rougher surface and pulverization was observed for the ball-milled fiber. As shown in Fig. 1c, the cellulose powder was fiberized and many nano-sized fibrils were formed on the

Table 1 Median particle size after ball milling

Rotational rate (rpm)	Pulverizing time (h)				
	1 (μm)	2 (μm)	4 (μm)	8 (μm)	16 (μm)
150	48.4	43.5	35.8	35.3	27.8
200	39.8	30.1	21.3	13.0	9.5
250	30.2	22.9	14.4	11.8	8.6

Control (untreated cellulose powder): 41.4

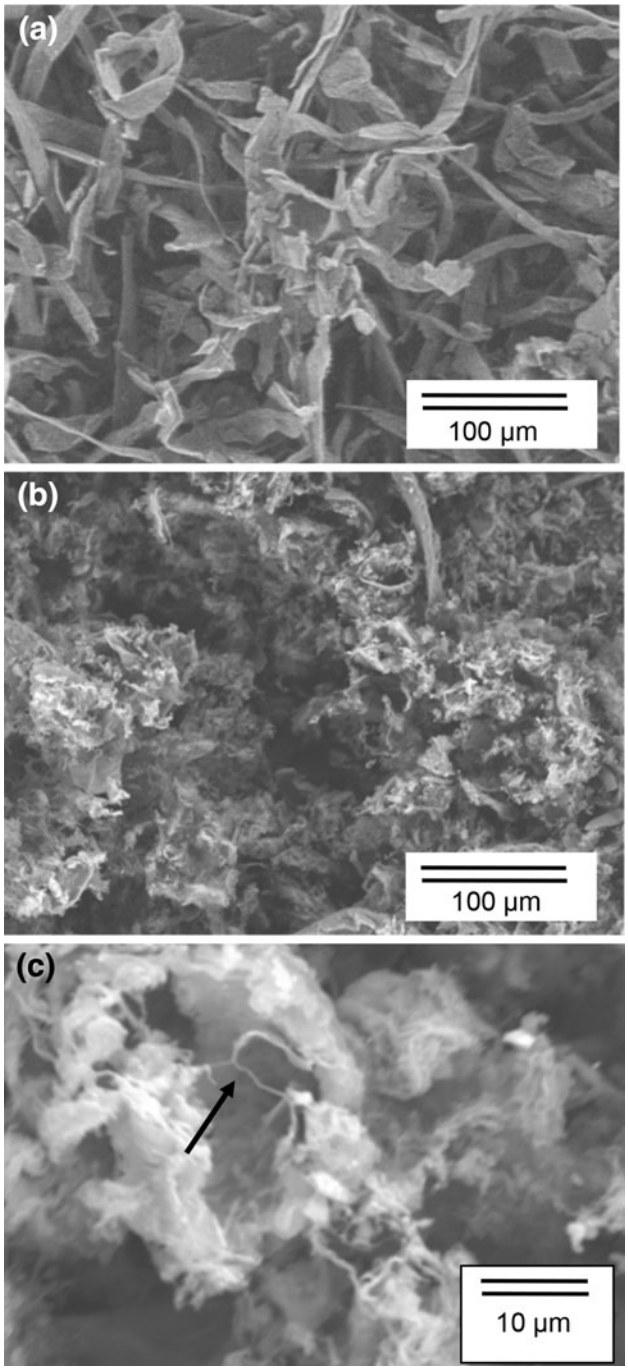

Fig. 1 SEM *photographs* of the cellulose powder before and after ball milling. **a** Untreated cellulose powder; **b** 250 rpm/1 h treatment; **c** Enlargement of **b** allow: CNF

surface: a nanostructured fiber with nanoscale surface fibrils on the surface was formed after ball milling.

The binding effect of CNF in the WF board

Boards were made for a single composition (i.e., WF:CNF = 80:20) to evaluate the binding effect of the CNF (Table 2). The measured board densities were 0.97–1.16 g/cm^3, very close to the target density of 1.00 g/cm^3 (Table 2). The density of boards made under the conditions of 200 rpm/16 h and 250 rpm/16 h exceeded 1.00 g/cm^3. The density did not change significantly for boards made at 150 rpm, while the densities of boards made at 200 and 250 rpm increased with increasing pulverizing time. With pulverizing time, the particle size of the CNF was diminished. Smaller CNF particles mixed more intimately with the WF, resulting in less void space in the pressed sheets. Figure 2a, b show the bending properties of boards containing CNF. For all rotational rates, the MOR and MOE of the boards containing CNF were higher than those for the board made from WF only, which indicated the binding effect of CNF between the WF particles. The MOR and MOE increased steadily with increasing pulverizing time, for all rotational rates, but the effect was enhanced at higher pulverizing rates. This is because under such conditions, many nano-sized fibrils were formed on the fiber surfaces, and the nano-sized fibrils effectively trapped WF particles [4]. However, the moduli of the boards made with untreated cellulose powder were lower than those for boards made from WF only. This suggested that the cellulose powder particles that lacked nano-sized surface fibrils could not act to bind the WF particles. Figure 3 shows the thickness swelling (TS) and weight change (WC) data for the water absorption test. The TS and WC values for boards with CNF were lower than those of the boards fabricated with WF only, or with untreated cellulose powder. Incorporation of CNF thus improved the

Table 2 Board densities: effect of changing pulverizing conditions

Samples	Density (g/cm^3)	Standard deviation
WF	0.97	0.04
Control (untreated)	0.98	0.03
150 rpm/1 h	0.97	0.03
150 rpm/2 h	0.98	0.03
150 rpm/4 h	0.98	0.02
150 rpm/8 h	0.98	0.03
150 rpm/16 h	0.98	0.02
200 rpm/1 h	0.98	0.04
200 rpm/2 h	0.98	0.02
200 rpm/4 h	1.01	0.03
200 rpm/8 h	1.03	0.02
200 rpm/16 h	1.11	0.02
250 rpm/1 h	0.98	0.03
250 rpm/2 h	1.01	0.03
250 rpm/4 h	1.03	0.02
250 rpm/8 h	1.09	0.04
250 rpm/16 h	1.16	0.05

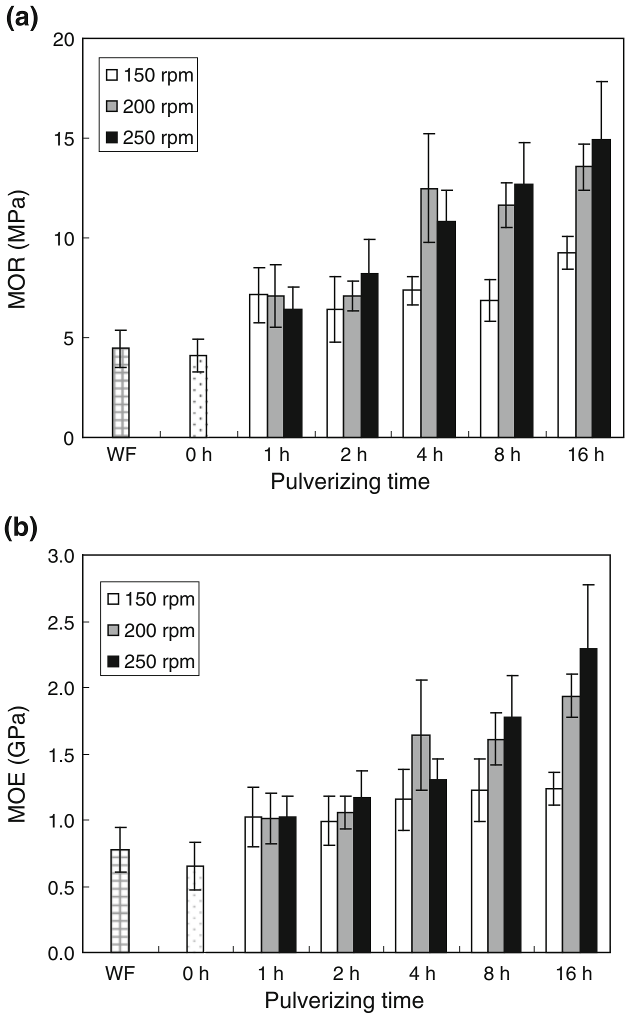

Fig. 2 Bending properties of WF boards containing CNF. **a** MOR; **b** MOE. *Vertical bars* indicate standard deviations

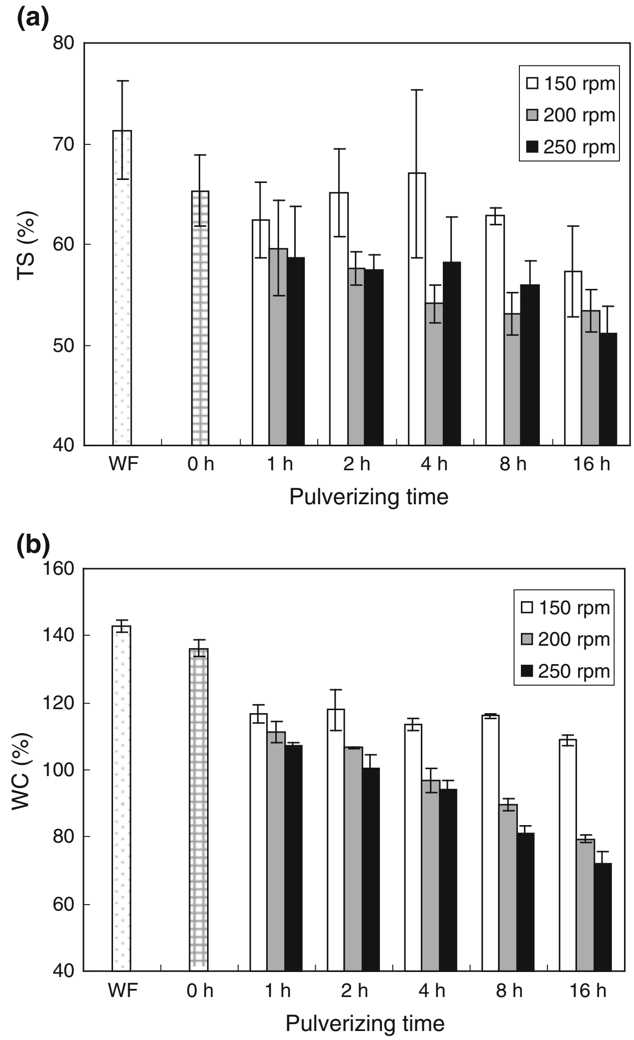

Fig. 3 Thickness swelling and weight change with the water absorption test. **a** Thickness swelling (TS); **b** weight change (WC). *Vertical bars* indicate standard deviations

water resistance. Moreover, high rotational rates or long pulverizing times tended to improve this property. The TS and WC values for board with untreated cellulose powder were lower than those of the board fabricated with WF only. This is the reason that the cellulose was poorer absorber than the WF.

The effect of composition on physical and mechanical properties of WF boards was also studied. Table 3 shows the measured density of the manufactured boards. The control was the WF board made with untreated cellulose powder. Figure 4 shows the bending properties (MOR, MOE) of the boards as a function of cellulose content. The highest moduli were found for the board having a WF:CNF of 90:10, which was considered the optimal composition. Increasing the CNF content above 20 % caused the moduli to decrease, which was attributable to flocculation of the

Table 3 Board densities: effect of changing composition

Samples	Density (g/cm^3)	Standard deviation
WF	0.97	0.04
WF:CNF = 95:5		
Control (untreated)	0.95	0.03
250 rpm/2 h	0.92	0.03
WF:CNF = 90:10		
Control (untreated)	0.94	0.03
250 rpm/2 h	0.92	0.05
WF:CNF = 80:20		
Control (untreated)	0.98	0.03
250 rpm/2 h	1.01	0.03

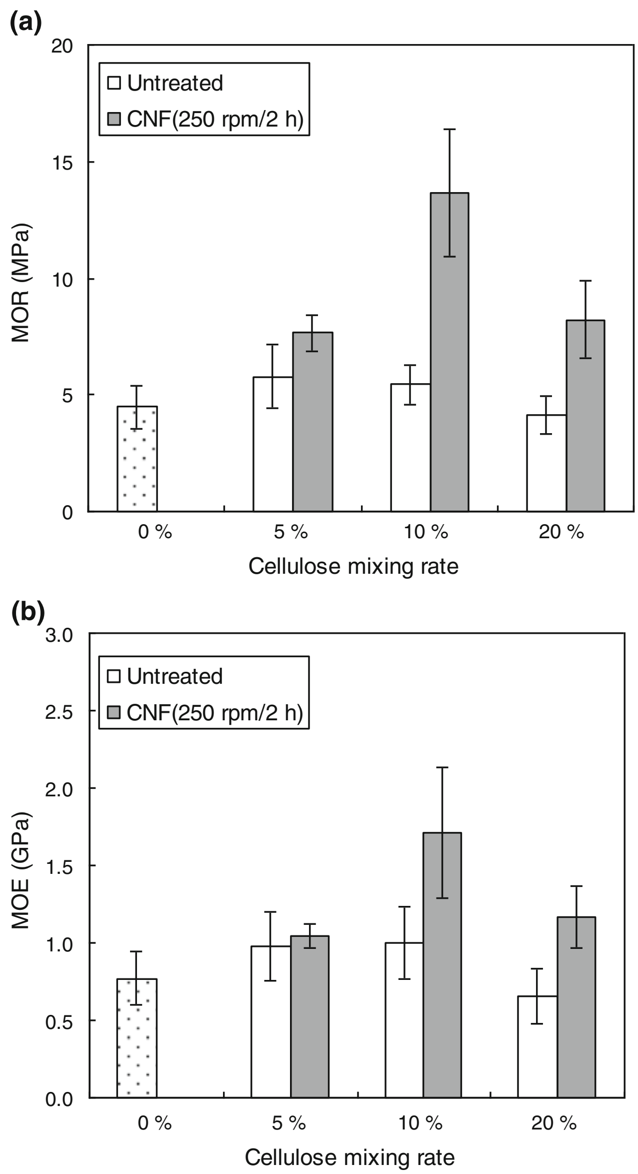

Fig. 4 Bending properties of WF boards containing cellulose powder and CNF. **a** MOR; **b** MOE. *Vertical bars* indicate standard deviations

CNF [6], which likely became more important as pulverization progressed. The mixing condition chosen for this part of the study (i.e., 250 rpm/2 h) was just sufficient to generate a binding effect by the CNF. Figure 5 shows the thickness swelling and weight change results for the water absorption test. For the boards made with untreated cellulose powder, the WC was largely unaffected by the composition. However, for the boards with CNF (250 rpm/2 h), TS and WC for 5 % CNF content tended to keep same values for WF board, and to decrease with increasing CNF content. This was because the CNF was a poorer absorber than the WF.

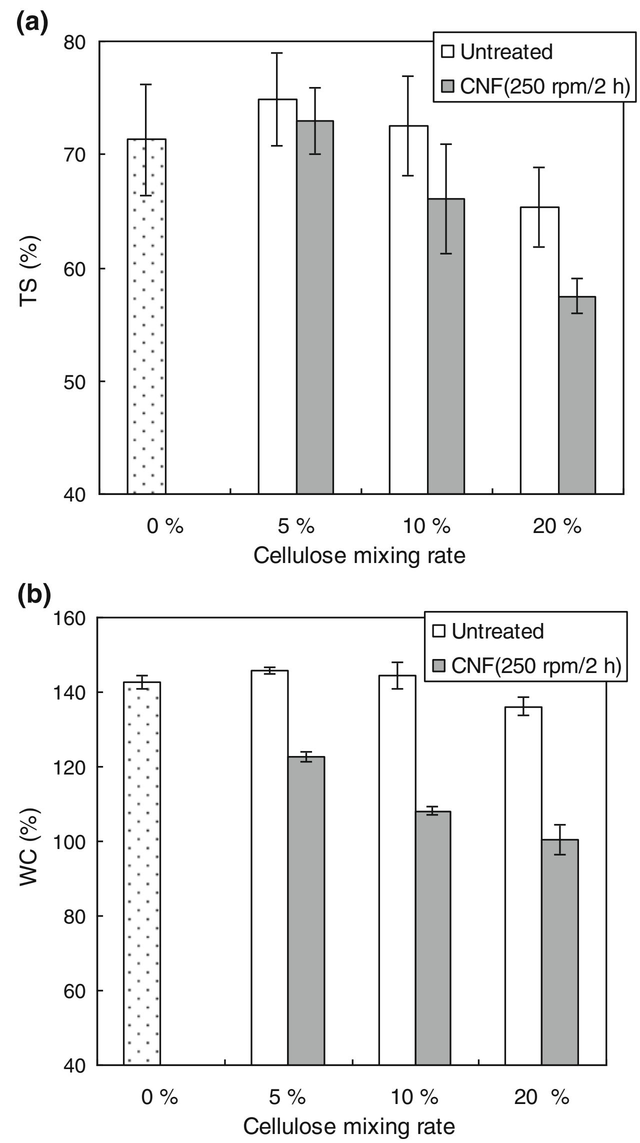

Fig. 5 Thickness swelling and weight change with the water absorption test. **a** Thickness swelling (TS); **b** weight change (WC). *Vertical bars* indicate standard deviations

Conclusions

This research studied the effects of adding CNF to WF on the properties of WF boards, with a focus on the binding effect of CNF in this system. Wet ball-milling of commercial cellulose powder led to the formation of nanostructured fibers with nano-sized surface fibrils. Increasing the pulverizing time or rotational rate reduced the particle size of the CNF. However, flocculation of the CNF particles could have occurred at long pulverizing times. The physical and mechanical properties of WF boards could be improved with the addition of CNF because of a binding effect between the CNF and WF particles.

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