Reinforcement of wood flour board containing ligno-cellulose nanofiber made from recycled wood

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Key words

wood-based materials, wood flour, cellulose nanofiber, ligno-cellulose nanofiber, wet pulverize

Abstract

Wood-based materials are widely used in residential construction. These materials can be made from virgin or recycled wood, and most of the materials are fabricated with chemical adhesives. Finding replacements for such chemical adhesives poses major challenges. This study explored nanofiber technology as an alternative to these adhesives. Previous studies have shown that the three-dimensional binding effects of cellulose nanofiber (CNF) and ligno-cellulose nanofiber (LCNF), when mixed with wood flour, can significantly improve the physical and mechanical properties of wood flour board. We use the word "LCNF" as the surface nanofibrillated wood flour. Previous studies have also highlighted problems that occur during compounding and board manufacturing. In this study, a reliable method was established to mix wood flour and LCNF. The method involved a compounding machine, which facilitated board manufacturing safely. Physical and mechanical properties of the resulting wood flour boards were significantly improved with the addition of LCNF, due to close binding between LCNF and wood flour particles.

Text

Introduction

Wood-based materials are used extensively in residential construction, particularly in Japan. These materials can be made from virgin wood, recycled wood, unused wood species, or thinning wood. Many of the materials are fabricated with various adhesives. Most of the currently available wood adhesives, such as formaldehyde-based resins, vinyl acetate resins, and isocyanate-based resins, are composed of various materials derived from fossil fuels.

Identifying replacements for these chemical adhesives poses major challenges. The global focus on sustainability demands development of novel, natural adhesives that do not depend on fossil fuels or synthetic chemicals. Some studies have focused on developing natural, material-based wood adhesives, using bio-resources [1]. For example, some natural adhesives are composed of citric acid [2-5] or lactic acid [6, 7].

In this study, we explored options involving nanofiber technology. Nanotechnology has been 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 [8]. This is true even for fibers that have diameters exceeding 100 nm.

There are many types of nanofiber. In particularly, cellulose nanofiber (CNF) has received attention in numerous fields. Over a trillion tons of CNF exist worldwide. CNF is known to have better physical and mechanical properties than most other fibers [9]. Developing new materials that incorporate CNF is a high priority [10-14]. However, use of CNF technology in wood-based materials has not been reported. Results from studies focusing on the relationship between fiber shape and mechanical properties for medium-density fiberboard (MDF) have been reported, but there has been no mention of nano-order fiber [15-17].

In a previous study, we investigated the effects of adding CNF to wood flour [18]. The resulting properties of the CNF/wood flour boards were evaluated, with a focus on the binding effects of CNF. We observed that wet ball-milling of commercial cellulose powder led to the formation of nanostructured fibers with nano-sized surface fibrils. Moreover, the physical and mechanical properties of the wood flour boards were significantly enhanced by the addition of CNF, due to three-dimensional binding between CNF and wood flour.

In another study, ligno-cellulose nanofiber (LCNF) was made from wood flour, using a disk mill [19]. In this context, CNF refers to nanofibers made from cellulose alone. In terms of reinforcement of wood flour, cellulose nanofiber is better than LCNF. However, in terms of productivity, LCNF is better than CNF because CNF requires a lot of process including delignification. Thus we employed LCNF. The fabrication of LCNF by disk milling is simple and effective, and its incorporation into wood flour board significantly enhances the physical and mechanical properties of the board. When CNF or LCNF were initially mixed with wood flour, both materials were mixed manually. It is difficult to uniformly mix LCNF into wood flour. CNF and LCNF were made by wet milling, resulting in a moisture content of the mixture (CNF or LCNF and wood flour) over 300%. Hot pressing of raw materials with high moisture content often reduces the quality of fabricated board.

In this study, we focused on establishing a reliable board manufacturing procedure to solve these problems. The binding effect of LCNF with wood flour was investigated as a function of LCNF content and its particle size.

Materials and Methods

Materials

Recycled wood flour, mainly consisting of particleboard, was obtained from Toclas Corporation (Shizuoka, Japan). The average size of wood flour fibers was about 220 μm. This wood flour was used both as a base material for wood flour board and as a material for LCNF.

Pulverization of wood flour to make LCNF

Wood flour (13.5 g) was mixed with distilled water (200 g) and pulverized using a ball mill (Pulverizette 6; Fritsch Japan Co., Ltd., Japan). The degree of pulverization was controlled by the time and rotational rate of the ball mill. Pulverizing time was set at six levels: 1, 2, 4, 8, 16, and 32 hours. The rotational rate of the ball mill was set at three levels: 100, 150, and 200 rpm. Eighteen different LCNF slurries were prepared and tested. A slurry made from untreated (non-pulverized) wood flour was the control. LCNF particle size after pulverization was measured with a laser diffraction particle size distribution analyzer (Partica LA-950; Horiba, Ltd., Kyoto, Japan). To prevent flocculation, the LCNF slurry was replaced with alcohol. After that samples were freeze dried, and LCNF powder was produced. Surface morphology of the LCNF powder was observed with a scanning electron microscope (SEM) (JSM-6510LV2; JEOL Ltd., Japan).

Fabrication of wood flour board with LCNF

Wood flour boards were made from mixtures of wood flour and the 18 LCNF slurries. We observed the effects of LCNF on the physical and mechanical properties of wood flour boards. A single composition (80 wt% wood flour + 20 wt% LCNF) was examined. Wood flour (54 g, dry weight) was mixed with LCNF slurry (CNF 13.5 g + distilled water 200 g) in a polyethylene bag. The moisture content of the mixture was over 300%. If hot pressing occurred without desiccation, it was difficult to safely make the board. When the mat moisture content is over 300%, it is impossible to produce the uniform board because excessive steam pressure will occurred, and moisture inside the mat will not completely evaporate during hot pressing and it will burst. Therefore, the mixture was compounded and dried with a compounding machine (Trimix, Inoue MFG. Inc, Japan) to less than 30% moisture content. Compounding and drying were performed at 40 rpm, 80°C, and for 15 minutes.

As a next step, 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 mat to accelerate water transfer during pressing. The mats were pressed for 10 min at 120°C and 0.85 MPa, using a hot press (Tabletop Test Press SA-302; Tester Sangyo Co., Ltd., Japan). Wood flour boards (15 cm × 15 cm × 0.3 cm) were manufactured with a targeted density of 1.00 g/cm^3. It is possible to make boards with lower density, but it is difficult to use the lower density boards. As a result, we selected the density of 1.0 g/cm^3.

As another experiment, the physical properties of the wood flour board were evaluated as a function of the relative amounts of LCNF and wood flour. The LCNF used in this experiment was fixed at 200 rpm/4 h. Three wood flour:LCNF combinations (80:20, 90:10, and 95:5) were evaluated.

For all experiments, two boards were produced for each treatment. All boards were conditioned at 20°C and 65% relative humidity for at least two weeks before testing. No adhesives or other additives were used.

Physical testing

After conditioning, four 12 cm x 2.5 cm pieces were cut from each board, for use in a three-point bending test with a universal testing machine. The following conditions were imposed: span 10 cm, loading speed 3 mm/min. The moduli of rupture (MOR) and modulus of elasticity (MOE) were calculated.

After the bending test, two pieces (2.5 cm × 2.5 cm) were cut from unstressed parts of the bending test specimen for the 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 change in weight and thickness of the pieces before and after soaking in water at 20°C for 24 h.

Results and Discussion

Evaluation of LCNF structure

Table 1 shows the median particle size of LCNF after ball milling. The size of the control (untreated wood flour) was about 220 μm. Smaller particle sizes were obtained with longer pulverizing times and at a fixed rotational rate. For a fixed pulverizing time, a higher rotational rate produced a smaller particle size. The smallest particle size was produced at 200 rpm/32 h, less than 1/30 of the particle size of untreated wood flour.

Figure 1 shows the particle size distributions of the control, 200 rpm/1 h, 200 rpm/8 h, and 200 rpm/32 h treatments. Smaller particle sizes were obtained with longer pulverizing times. The variation in particle size of the control was relatively large. Less variation in particle size resulted from increased pulverizing time. Therefore, it is possible to control the size uniformity of wood flour particles by adjusting the settings of the ball mill. The peaks for 32-hour treatments are shown in two parts. Smaller peaks (0.1-1μm) were obtained from the small fibril formed by wet ball milling peeled from the surface of wood flour.

The morphology of the LCNF was studied with the SEM. Figure 2 shows SEM photographs of the wood flour before and after ball milling (100 rpm/4 h and 200 rpm/4 h). The surface of the untreated fiber was very smooth, while a rougher surface and pulverization were observed for the ball milled wood flour. Nanostructured fibers with nanoscale surface fibrils were formed on the surface of wood flour after ball milling. We have confirmed that the size of LCNF nanofibrils made by ball milling is the same CNF nanofibrils made by ball milling [18].

Binding effect of LCNF in the wood flour board

A compounding machine was used in the mixing process, and the LCNF and wood flour were mixed simultaneously during the drying process. Figure 3 shows the manufactured board in this study. Compounding machine was applied to mix the LCNF and wood flour homogeneously and extract water from compound. As shown in Fig.3, the use of compounding machine made it possible to produce boards with uniform quality.

Boards were produced at a single composition (i.e., wood flour: LCNF = 80:20) to evaluate the binding effect of the LCNF (Table 2). The board densities were 0.91-1.19 g/cm^3, which was very close to the targeted density of 1.00 g/cm^3. The densities for untreated wood flour, lower rotational rate, and shorter pulverizing time were lower than the targeted density. On the other hand, the densities for higher rotational rate and longer pulverizing time were higher than the targeted density. In our previous report, the same phenomenon was observed for the CNF/wood flour mixed board [18]. Smaller LCNF particles mixed more closely with the wood flour, resulting in less vacant space in the pressed boards. Figure 4a and b show the bending properties of boards containing LCNF. For all conditions, the MOR and MOE of the boards containing LCNF were higher than for boards made from wood flour only, which indicated the binding effect of LCNF between the wood flour particles. In particular, the bending properties for the board with LCNF made from 200 rpm/32 h were about three times higher than for wood flour board without LCNF. Particle size of the LCNF for 200 rpm/32 h was 1/20 that of untreated wood flour (Table 1). In general, the quality of composite properties mixed with smaller size particles was not improved [20]. Nevertheless, in this study the properties of boards made with LCNF were improved compared to wood flour only. As a result, the nano-sized fibril effectively trapped wood flour particles. According to the JIS standard [21], the minimum requirement of MOR for MDF is 5 MPa, and the maximum value of the board in this study was over 10 MPa without resin. This indicates that LCNF could be used as an adhesive for wood-based materials. The mechanism of bond performance development is thought to be due to the mechanical entanglement between LCNF and wood flour associated with water evaporation during hot pressing. The other possibilities (ex: the thermomechanical flow of chemical components) were supposed, should be discussed in our next papers.

Figure 5 shows the internal bond (IB) strength of boards containing LCNF. The IB value of the board with 100 rpm LCNF was the same as the IB of a wood flour board. For 150 rpm and 200 rpm, IB values tended to increase with increasing pulverizing time. The IB values for boards with LCNF made from 200 rpm/16 h and 32 h were about five times greater than for wood flour board without adding LCNF. This is the reason that the nano-sized fibril formed on the fiber surface reinforced binding between wood flour and LCNF.

Figure 6 shows the degrees of thickness swelling (TS) and weight change (WC) in the wood flour/LCNF composites with water absorption. The TS and WC values for boards containing LCNF were lower than for boards fabricated with wood flour only. The board densities were almost the same as those shown in Table 2, indicating that the void ratios in the test samples were nearly identical. Nano-sized fibrils formed at the fiber surfaces during mixing of LCNF with wood flour resulted in close binding of the two components. As a result, water was not as easily able to enter the composite samples. Thus, incorporation of LCNF improved water resistance. Due to the lack of standards for swelling behavior of nonstructural panel, it is difficult to compare swelling behavior of LCNF board with other nonstructural panels. With the comparison of structural MDF provided by Japanese industrial standards (JIS) [21], thickness swelling and water absorption of LCNF boards are much higher than MDF. This is because no adhesion was added to LCNF board.

The effect of composition (LCNF/wood flour ratio) on physical and mechanical properties of wood flour boards was also studied. Note that only the LCNF for 200 rpm/4 h was used for this study. Table 3 shows the density of the manufactured boards. As indicated above, the board densities were 0.94-0.99 g/cm^3, which were very close to the targeted density of 1.00 g/cm^3. Figure 7 shows the bending properties (MOR and MOE) of the boards as a function of LCNF content. The bending properties of boards with LCNF had higher values than for wood flour only. Even if the LCNF content was 5%, the values were twice those for wood flour only. The highest values were found for the boards containing 10% LCNF and 90% wood flour, which was considered the optimal composition. Generally there is a positive correlation between additive amount of adhesion and mechanical properties of board because the resin flows and penetrates so effectively. However in this experiment, mechanical properties of LCNF board decreased from additive rate at 10% to 20%. LCNF reinforces the board property by the entanglement between wood flour and LCNF. The LCNF only exhibits localized flow and no penetration. It is suggested that part of LCNF does not become entwined with wood flour but with LCNF itself and then agglomerated when 20% of LCNF additive rate. In our previous report, the same phenomena were observed for CNF or LCNF/wood flour mixed board [18, 19]. Figure 8 shows IB strength as a function of LCNF content. IB strength of boards with LCNF was higher than for wood flour only. IB strength for 10% LCNF content was the same for 20% LCNF content. Figure 9 shows the thickness swelling (TS) and weight change (WC) results from the water absorption test. Both TS and WC decreased with increasing LCNF content, due to close binding of LCNF to the wood flour. The results for 10% LCNF content were the same as for 20% LCNF content.

Conclusions

Wet ball milling of recycled wood flour resulted in the formation of nano-structured fibers with nano-sized surface fibrils. Uniformity of the processed wood flour was controlled by the ball mill settings (i.e., pulverizing time and rotational rate). A reliable and safe method for mixing wood flour and LCNF was established using the compounding machine. The physical and mechanical properties of the resulting wood flour boards were significantly improved with the addition of LCNF, due to close binding between LCNF and wood flour particles.

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Figure and Table Captions

Table 1

Median particle size after ball milling.

Figure 1

Particle size distributions shown for wood flour, 200 rpm/1h, 200 rpm/8h, and 200 rpm/32h.

Figure 2

SEM micrographs of the surface morphology of (a) untreated wood flour, (b) LCNF (100 rpm/4h) and (c) LCNF (200 rpm/4h).

Figure 3

Manufactured board.

Table 2

Board densities: effect of changing pulverizing conditions.

Figure 4

Bending properties of wood flour boards containing LCNF.

(a) MOR; (b) MOE.

Vertical bars indicate standard deviations.

Figure 5

Internal bond (IB) strength of wood flour boards containing LCNF.

Vertical bars indicate standard deviations.

Figure 6

Thickness swelling and weight change with the water adsorption test.

(a) Thickness swelling (TS); (b) Weight change (WC).

Vertical bars indicate standard deviations.

Table 3

Board densities: effect of changing composition.

Figure 7

Bending properties of wood flour boards containing LCNF.

(a) MOR; (b) MOE.

Vertical bars indicate standard deviations.

Figure 8

Internal bond (IB) strength of wood flour boards containing LCNF.

Vertical bars indicate standard deviations.

Figure 9

Thickness swelling and weight change with the water adsorption test.

(a) Thickness swelling (TS); (b) Weight change (WC).

Vertical bars indicate standard deviations.

Table 1

Rotational rate (rpm)	Pulverizing time (hour)					
	1	2	4	8	16	32
100	139.8	111.8	78.9	54.3	44.9	32.0
150	92.1	61.0	28.5	16.0	10.9	7.5
200	65.9	29.8	16.0	11.2	7.3	5.8
Control (wood flour) :219.2 μm						unit: μm

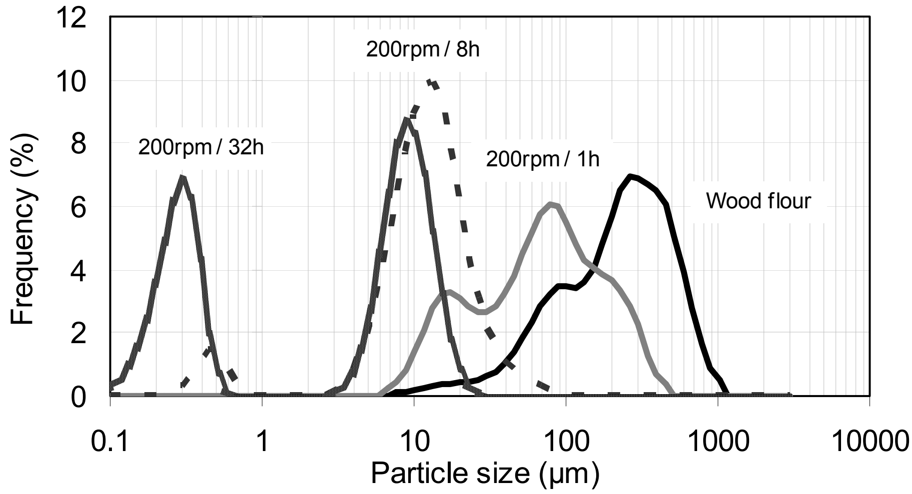
Table 2

Samples	Density (g/cm^3)	Standard deviation
Wood flour	0.94	0.06
100rpm/1h	0.91	0.04
100rpm/2h	0.94	0.05
100rpm/4h	0.98	0.07
100rpm/8h	0.99	0.06
100rpm/16h	0.97	0.03
100rpm/32h	0.96	0.05
150rpm/1h	0.92	0.02
150rpm/2h	0.92	0.02
150rpm/4h	0.92	0.02
150rpm/8h	0.93	0.06
150rpm/16h	0.95	0.04
150rpm/32h	1.01	0.04
200rpm/1h	1.00	0.04
200rpm/2h	1.03	0.02
200rpm/4h	0.99	0.04
200rpm/8h	0.97	0.04
200rpm/16h	1.09	0.05
200rpm/32h	1.19	0.06

Table 3

Samples	Density (g/cm^3)	Standard deviation
Wood flour (WF)	0.94	0.06
WF : LCNF = 95 : 5	0.99	0.07
WF : LCNF = 90 : 10	0.97	0.06
WF : LCNF = 80 : 20	0.99	0.04

Figure 1



12
10
8
6
4
2
0
Frequency (%)
200rpm / 8h
200rpm / 32h
200rpm / 1h
Wood flour
0.1
1
10
100
1000
10000
Particle size (μm)

Figure 2

(a)

(b)

(c)

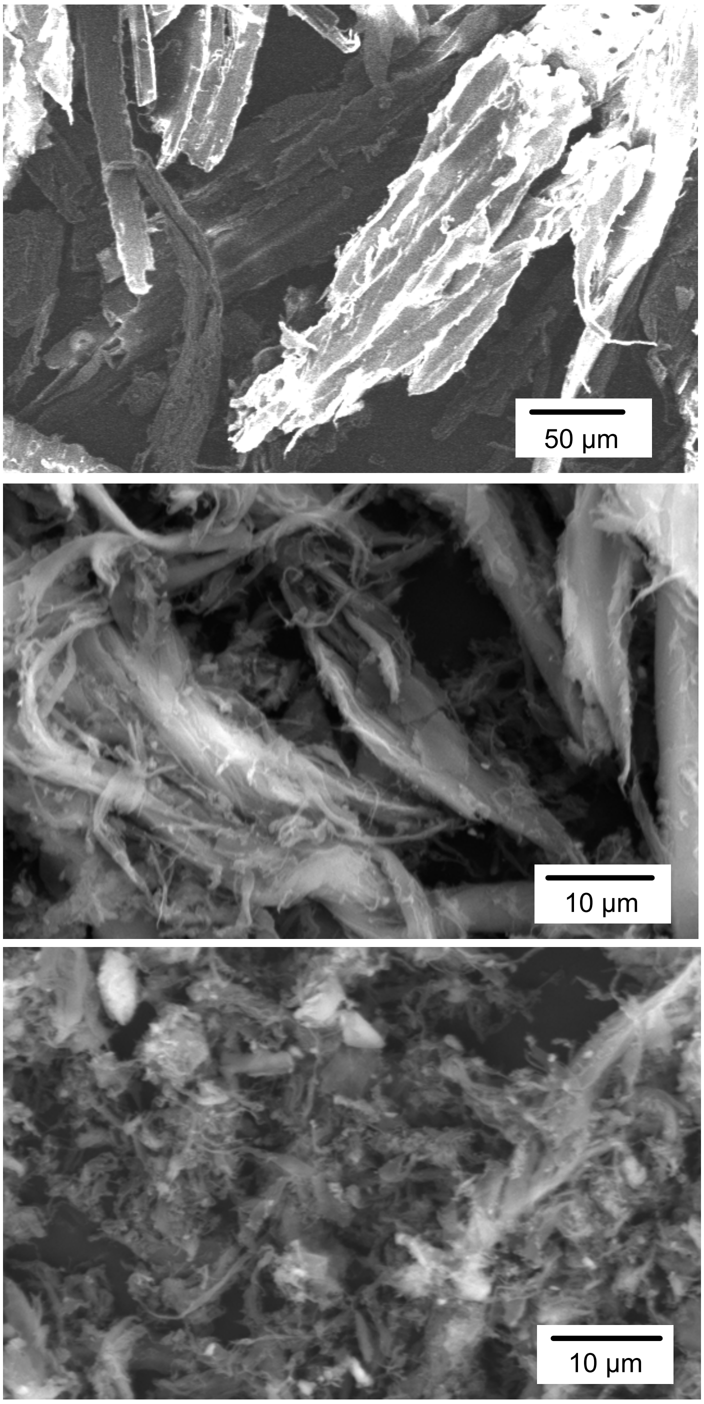
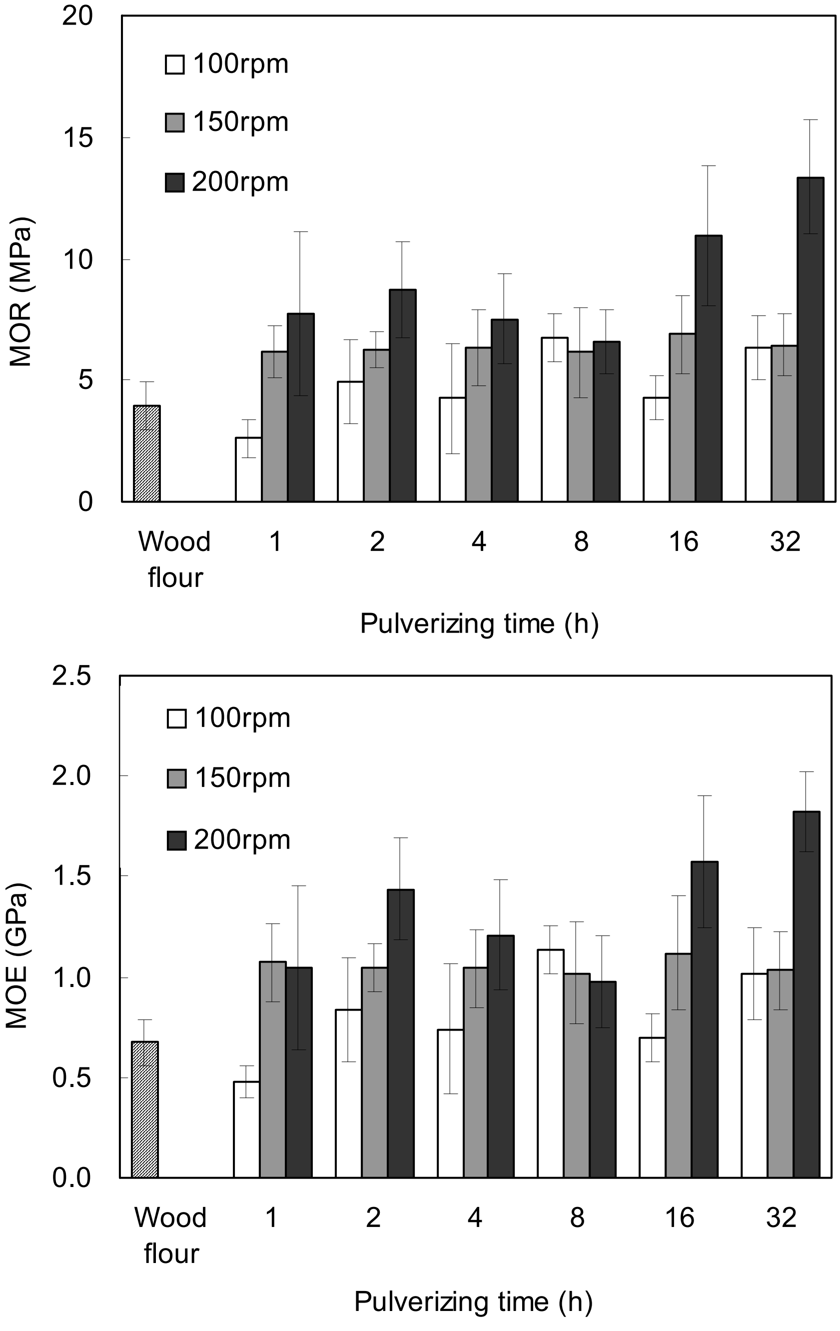


Figure 3

Figure 4



20
15
10
5
0
MOR (MPa)
100rpm
150rpm
200rpm
Wood flour
1
2
4
8
16
32
Pulverizing time (h)
2.5
2.0
1.5
1.0
0.5
0.0
MOE (GPa)
100rpm
150rpm
200rpm
Wood flour
1
2
4
8
16
32
Pulverizing time (h)

Figure 5

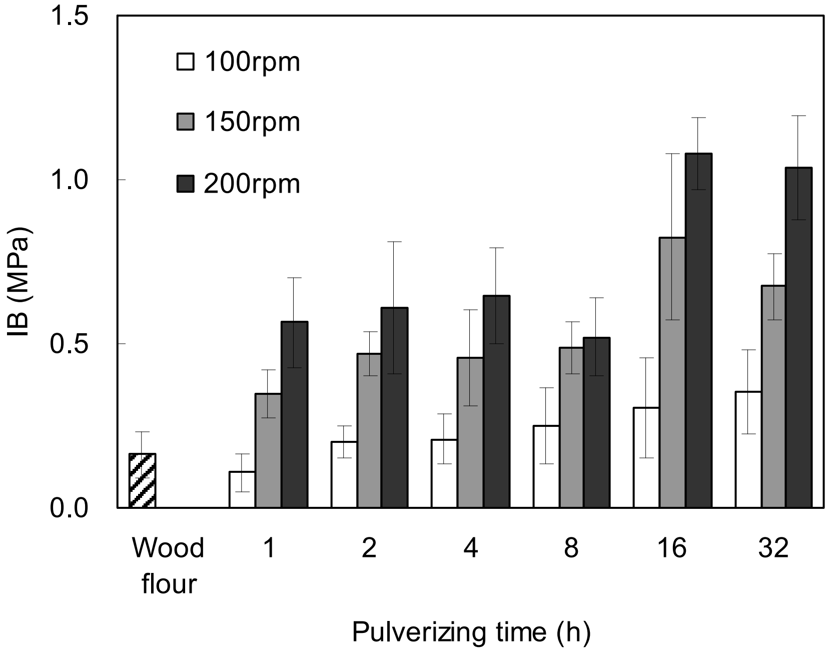

1.5
1.0
0.5
0.0
IB (MPa)
100rpm
150rpm
200rpm
Wood flour
1
2
4
8
16
32
Pulverizing time (h)

Figure 6

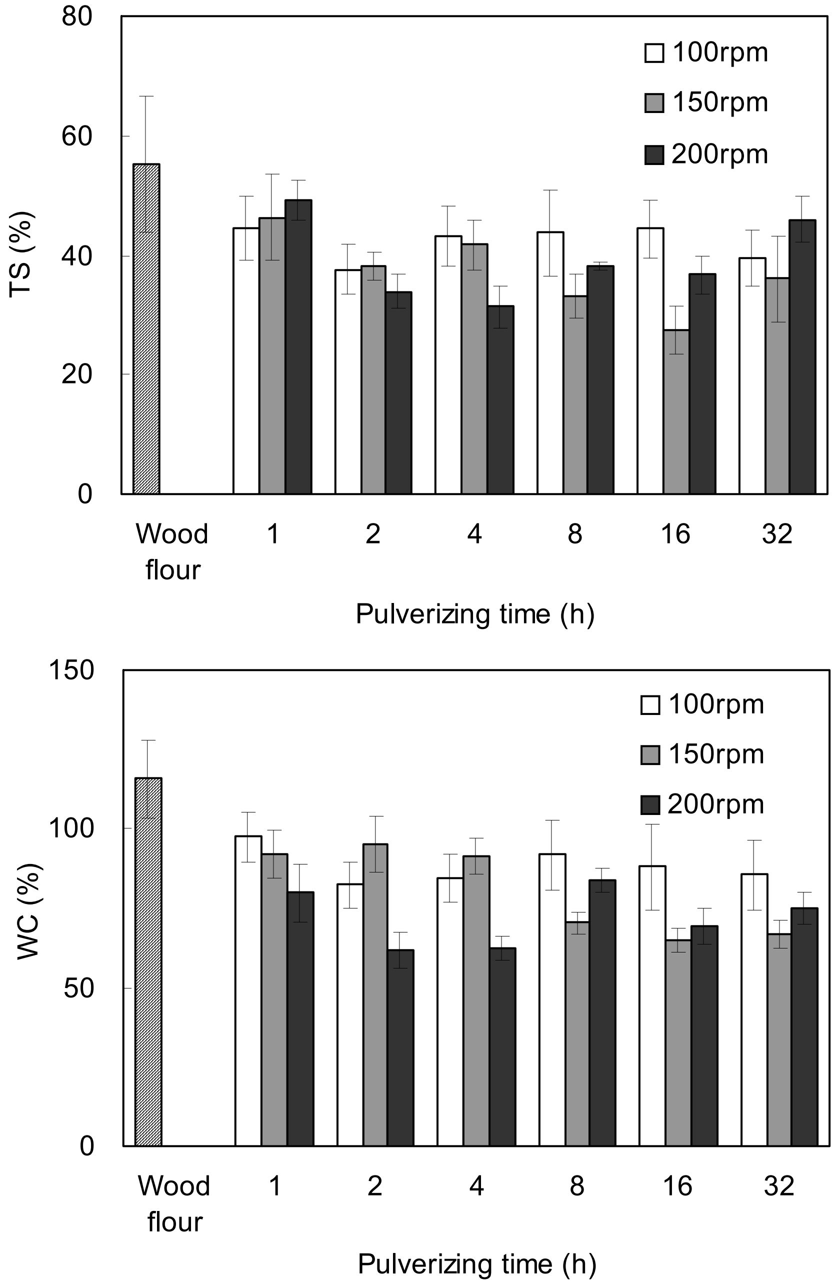
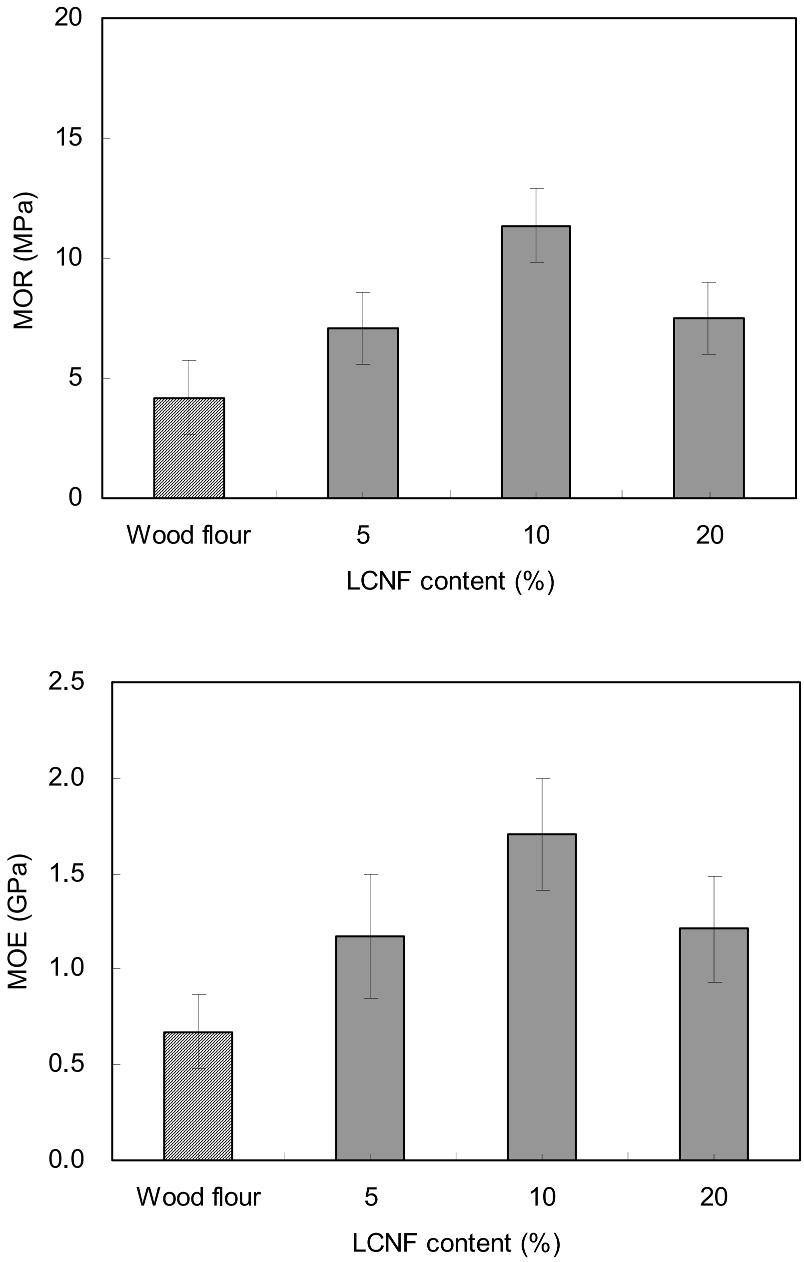

80
60
40
20
0
TS (%)
100rpm
150rpm
200rpm
Wood flour
1
2
4
8
16
32
Pulverizing time (h)
150
100
50
0
WC (%)
100rpm
150rpm
200rpm
Wood flour
1
2
4
8
16
32
Pulverizing time (h)

Figure 7



20
15
10
5
0
MOR (MPa)
Wood flour
5
10
20
LCNF content (%)
2.5
2.0
1.5
1.0
0.5
0.0
MOE (GPa)
Wood flour
5
10
20
LCNF content (%)

Figure 8

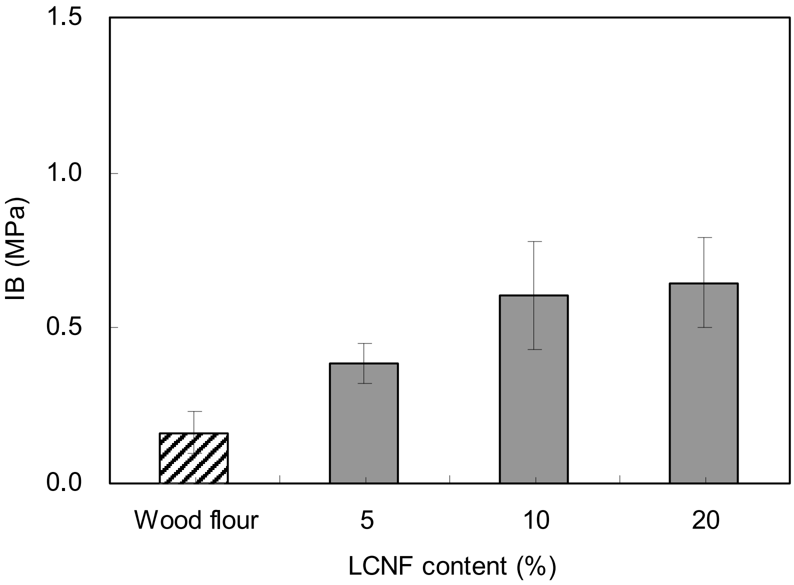
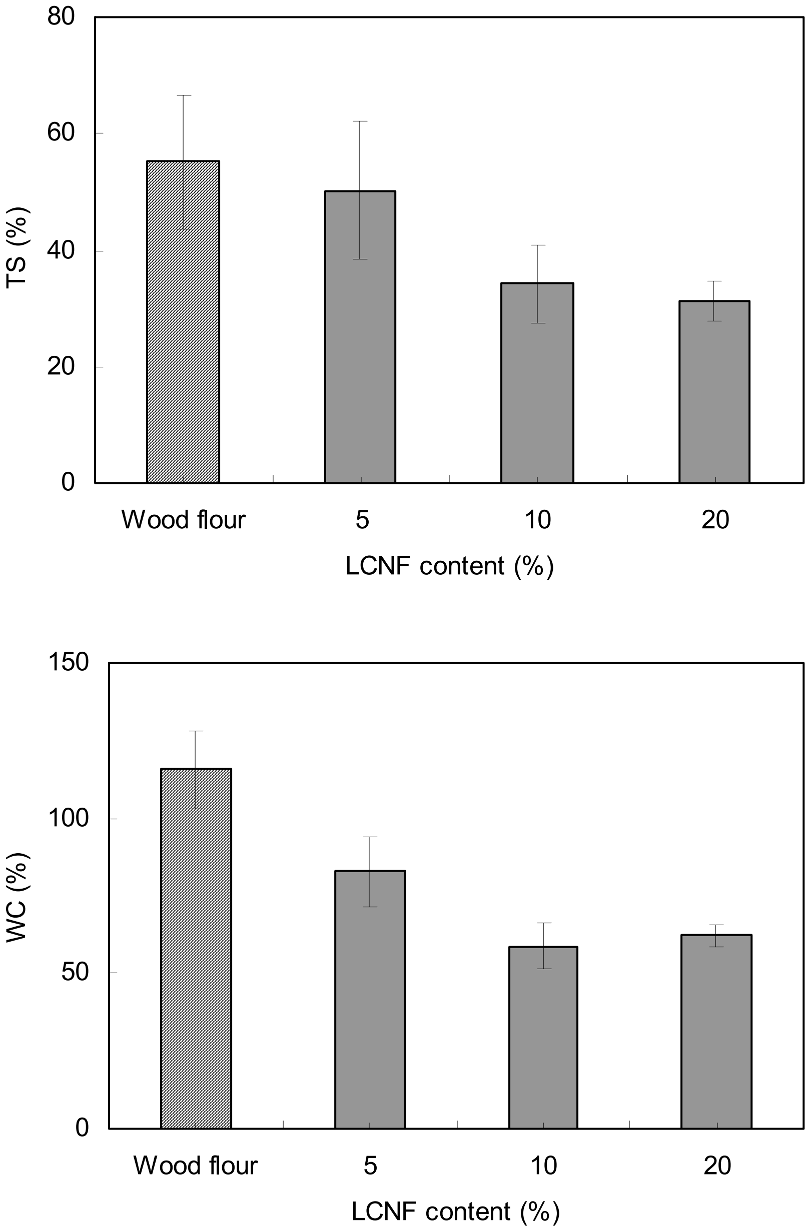


Figure 9



80
60
40
20
0
TS (%)
Wood flour
5
10
20
LCNF content (%)
150
100
50
0
WC (%)
Wood flour
5
10
20
LCNF content (%)