Production of the High Density and Strength Moldings from Wood Flour without Binder by the WIP (warm isostatic pressing) Technology

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Wood flour of various particle sizes was milled from lumber waste with the fine grinder. After production of preliminary moldings from the air-dried wood flour by the press using a metal implement, wood moldings were cast using a fluoride rubber mold in a pressure of 50-500MPa and at a temperature of $90-140^{\circ}$ with WIP equipment. The density of wood moldings was about $1.4g/\text{cm}^3$ irrespective of particle size and casting conditions, being close to density of wood cell wall $(1.5g/\text{cm}^3)$. The Rockwell's hardness was quite large compared with the hardness of useful wood (about 60HRR), and was comparable to very hard wood (about 90HRR). The compressive strength was increased with the decrease in used particle size and the rise of casting temperature. The strength was larger than one of useful wood (40-50MPa), and the maximum strength was 90MPa or more. The wood moldings did not break up in boiling for 72 hours, though the moldings swelled.

Key words: wood flour, no-binder, warm isostatic pressing, high density, high strength

1. INTRODUCTION

The wood of about 5 million ton C per year is discharged as the scrap of wood products, such as lumber-manufacturing refuses, housing demolition and furniture, etc. Most (about 70%) is only burned or thrown away. This is the problem which must be immediately solved from not only a viewpoint of the reduction of natural resource utilization but also the construction recycling law. However, since the wood wastes are discharged in various forms, this becomes a barrier for effective recycling. It is required to develop the new manufacturing process which is not restrained by the form of wood waste. This should promote the recycling of wood wastes.

It was shown that the wood surface was thermal-plasticized by an allylation, a kind of chemical modification, and consequently was able to adhere tightly each other by pressing at a temperature of 120-180 °C and in a pressure of 0.5-1.4MPa without using binder ¹⁻³⁾. The allylation of wood should have caused the disintegration of a crystal structure in cellulose and the drop of thermal-softening temperature of lignin. As a result, it was considered that the contact area between two surfaces was increased and the molecules of surface came closer to each other during pressing the allylated wood. These would be a reason for tight adhesion between the surfaces of allylated wood.

On the other hand, it was reported that the moldings like plastics was able to be produced from the oven-dried wood flour by pressing at a temperature of 180-240°C and in a pressure of 100MPa for 10min without using binder⁴). This was considered to be because of the softening of matrix components in wood by the heating in high temperature and pressing in high pressure. But, the high temperature caused the discoloration of wood moldings. The produced moldings collapsed and took apart to pieces by immersion in water.

The heat-softening temperature of lignin isolated from

wood is $134-235^{\circ}$ in the drying state and is dropped to $77-128^{\circ}$ in the moisture content of $7.2-27.3^{\circ}$ ⁵⁾. This suggests that the natural lignin in wood is soften in air-dried state by heating at comparatively low temperature. The lignin is consequently expectable to function as a plasticizer of particles when the wood flour is pressed in an airtight vessel so that the flour does not lose the moisture.

In the new proposed process, the wood wastes are ground to fine particles and cast at low temperature $(140^{\circ}C)$ or less) and in super-high pressure (500MPa or less) without using binder with WIP (warm isostatic pressing) equipment to produce the wood moldings having high density and high strength.

The apparent density of wood is 0.10-1.31g/cm³ because of existence of opening called cell lumen, though the density of wood cell wall is about 1.50g/cm³. The compressive strength parallel to grain of wood is 30-110MPa, and the strength of WPC (Wood Plastic Composites) is 70-120MPa. These values are numerical targets in this research. The production of the wood products having high added-value should contribute to progress of effective utilization of wood wastes.

2. EXPERIMENTS

Wood (Douglas fir) flour was milled from lumber waste with the fine grinder and was sifted in various particle sizes (32-mesh (0.5mm) -pass to 350-mesh (44 μ m)-on) through a sieve. The particle sizes of used wood flour are shown in Table I.

At first, the air-dried wood flour (MC: 8-10%) was put into a metal implement and was pressed in a pressure of 9-12MPa and at a room temperature for 3-5 minutes. The preliminary moldings were produced through this procedure.

Then, these preliminary moldings were put into an airtight mold made of fluoride rubber. The rubber mold

was immediately put in WIP equipment, and was pressed in a pressure of 50-500MPa and at a temperature of 90-140 $^{\circ}$ for fixed time (1hr), to cast into the wood moldings (diameter of about 5cm, height of about 2cm)^{6,7}.

Table I Particle sizes of wood flour used in casting of moldings.

Number	Particle size
No.1	$32 \text{mesh}(0.5 \text{mm}) \text{ pass} - 60 \text{mesh}(250 \ \mu \text{ m}) \text{ on}$
No.2	$60 \text{mesh}(250 \ \mu \text{ m}) \text{ pass} - 100 \text{mesh}(149 \ \mu \text{ m}) \text{ on}$
No.3	$100 \text{mesh}(149 \ \mu \text{ m}) \text{ pass} - 120 \text{mesh}(125 \ \mu \text{ m}) \text{ on}$
No.4	$120 \text{mesh}(125 \ \mu \text{ m}) \text{ pass} - 170 \text{mesh}(88 \ \mu \text{ m}) \text{ on}$
No.5	$170 \text{mesh}(88 \ \mu \text{ m}) \text{ pass} - 250 \text{mesh}(63 \ \mu \text{ m}) \text{ on}$
No.6	$250 \text{mesh}(63 \ \mu \text{ m}) \text{ pass} - 350 \text{mesh}(44 \ \mu \text{ m}) \text{ on}$

Observation by SEM (Scanning Electron Microscopy) and measurements of X-ray diffraction, density, Rockwell's hardness (hardness unit: HRR, JIS Z 2245, K 7202 compressive strength (dimension: (Plastics)), 1cmx1cmx2cm, test speed: 0.2mm/min) were carried out on the produced wood moldings. X-ray diffractograms of wood flour and wood moldings were obtained with 20mm-diameter disks prepared. The equatorial diffraction pattern was measured over a range of $2\theta = 3^{\circ}$ to 40° at a rate of 0.02°/sec using Ni-filtered Cu-K α radiation generated at 40kV and 120mA. The slit widths used were 1-1-0.4-0.5°. The water-resistance of wood moldings was examined by putting in boiled water for 72 hours.

3. RESULTS AND DISCUSSION

3.1 Density of wood moldings

The density of the obtained moldings is shown in Fig.1. The high-density wood moldings $(1.39-1.43 \text{ g/cm}^3)$ were able to be produced by this process. The density didn't depend on the particle size of wood flour, molding temperature and pressure. The density of obtained moldings was much bigger than one of used wood materials, which was $0.35-0.40 \text{g/cm}^3$. This indicates that the wood cells of flour in moldings were crushed by pressing in super-high pressure and the form of flour particles was largely changed.

However, the density of produced moldings was unable to reach the value of 1.50 g/cm^3 which is the density of wood cell wall. This indicates that the opening or gap should have remained in wood flour particles or among particles.

The lignin should have been softened during the casting, because the casting was carried out in the higher temperature than the softening temperature of lignin in air-dry state, as mentioned above. It is thought that the softening of lignin brought about the adhesion among particles. The adhesion may be because of van der Waals forces between the molecules in wood particle surface.

3.2 SEM observation of surface of wood moldings

The SEM photograph of the surface of wood molding is



Fig.1 The density of wood moldings produced in various casting conditions.

and the folding fibers can be seen in the photograph. This fact indicates that the wood cells were softened during the casting with WIP equipment. The reason for softening of wood cell would be that the non-crystalline components in wood cell, such as lignin and hemicellulose, were easily softened by pressing in the applied pressure and temperature. It is considered that the softening of wood cell caused the adhesion among the wood flour particles.



Fig.2 The SEM photograph of the wood molding cast in a pressure of 100MPa and at a temperature of 140℃ (sample No.1).

3.3 X-ray diffraction curve of wood moldings

Fig. 3 shows the X-ray diffraction curve of the used wood flour and the produced wood moldings. In the wood flour, a

shown in Fig. 2. The melting-like parts (arrows in figure)

peak height was lower in the particle having smaller size (No.3), though the peak position didn't shift with the difference of particle size. This shows that crystallinity of wood was dropped with reduction in particle size, that is, a part of crystal in wood was destroyed during the milling by the fine grinder. The extent of destruction of crystal would have been larger in the case of particles having smaller size.

On the other hand, in the wood moldings the peak position shifted to higher degree compared with one of wood flour regardless of the particle size and the casting pressure, as clearly seen in the solid line in figure. This shows that the pressing in applied pressure and temperature brought about a certain change in the high order structure of the amorphous region in wood. This result may have deep relation with the softening of wood components observed in the SEM photograph.



Fig. 3 X-ray diffraction curves of the used wood flour and the wood moldings cast in a pressure of 100 and 400MPa and at a temperature of 140° C.

No.A:16mesh (1.19mm) pass – 32mesh (0.5mm) on No.B:60mesh (250μ m) pass – 120mesh (125μ m) on No.6: shown in Table I



Fig.4 The Rockwell's hardness of wood moldings produced in various casting conditions.

3.4 Hardness of wood moldings

The Rockwell's hardness of wood moldings is shown in Fig. 4. The hardness tended to increase with the reducing particle size of wood flour. The influence of casting pressure on the hardness was small. The hardness of moldings cast at a temperature of 140° C was 90-110HRR. These values were larger than the hardness in cross-section of not only medium hard wood (about 60HRR) but also hard wood (about 90HRR). This indicates that the very hard moldings were able to be produced by this process.

The casting temperature had large effect on the hardness of wood moldings cast in a pressure of 100MPa. The hardness of moldings cast at 90°C was almost same or a little larger compared with density of the medium hard wood, followed by the increasing with the rising temperature. The hardness differed with the temperature, though the density was almost same, as shown in Fig.1. This indicates that the tighter adhesion among the particles was brought about in the higher temperature, because the degree of softening of wood was larger.

3.5 Compressive strength of wood moldings

The compressive strength of wood moldings is shown in Fig. 5. The compressive strength tended to increase with the reducing particle size of wood flour. The influence of casting pressure on compressive strength didn't have a definite tendency in moldings cast at a temperature of 140° C. The strength of all moldings was larger than the strength parallel to grain of medium hard wood (40-50MPa). The some moldings had larger strength than hard wood (about 70MPa). The maximum strength of moldings was 90MPa.



Fig.5 The compressive strength of wood moldings produced in various casting conditions.

On the other hand, the compressive strength was increased with the rising temperature in the wood moldings cast in a pressure of 100MPa. The reason for this increase is considered that the softening of wood components became easier with the rise of temperature and the adhesion among particles became tighter.

3.6 Water-resistance of wood moldings

The wood moldings swelled during boiling in water for 72 hours, the same as wood swells in boiled water. However, the moldings never broke up in boiled water. The moldings returned to the original shape after drying, though the size was slightly larger. These indicate that the adhesion among particles in wood moldings is water-resistant.

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(Received October 10, 2003; Accepted October 31, 2003)