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Binding Mechanism of Binderless Boards Fabricated by Compressively Molding with High-Pressure Steam

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Fabrication of binderless boards by compressively molding with high-pressure steam has been developed recently. The purpose of this study is to clarify its binding mechanism. The significant improvement in the physical properties of boards by converting its raw material from sawdust to wood-fiber suggests that fabric entanglement is deeply involved in its binding mechanism. Physical properties of boards made from extractive-free sawdust indicates that the presence of extractives in wood plays a role of supporting its dimensional stability, but this may not be a major binding factor. Furthermore, structural stability of fabricated boards by removal of hemicelluloses and lignin with hot water extraction and dioxane extraction indicates that the main binding factor in board fabrication is cellulose, even though lignin and hemicelluloses contribute to the additional strength. The increase in cellulose crystallinity and reconstruction of its crystal structure by high-pressure steaming creates entanglement of sawdust in the board. In addition, the successful fabrication of binderless boards with sufficient strength from the materials which contain infinitesimal amount of lignin, such as paper, cotton and fabric emphasizes that cellulose plays a major role in the binding mechanism of binderless boards.

Key words: Binderless Board, Compressively Molding, Binding Mechanism, High Pressure Steam

1. INTRODUCTION

Large volumes of wood residues, such as sawdust, chips and forest thinnings are wasted from the sites of lumbering and forestry. Even though decline in natural resources and emission of greenhouse gas are major world concerns, most of them are disposed by burning or buried landfills without utilizing as woody resources. To respond the requirement of ecological and sustainable technologies of utilizing those wood resources, fabrication of binderless board by compressively molding with high-pressure steam has been developed recently [1-4]. By using this technique, boards can be fabricated from woody residues without adding any chemical adhesives. Since the boards fabricated by this technique do not contain any synthetic chemicals, they are expected to be utilized as biodegradable and eco-friendly materials. In previous work on this technique, investigations of the effect of particle sizes [2], application of this method to agricultural residues [3] and investigation of better steaming conditions [4] were conducted. However, the binding mechanism of this binderless board has not been elucidated yet. The purpose of this study is to clarify the binding mechanism by varying its raw materials and analyzing its physical properties.

2. MATERIAL AND METHODS

2.1 Sample preparations

To avoid affections on mechanical properties of fabricated boards by discrepancy of tree species, red lauan (*Shorea negrosensis)* was selected as material species for this experiment. Sawdust screened to less than 0.84 mm (20 mesh pass) and wood-fiber provided from Hokushin Co., Ltd were used as raw materials for fabricating the boards.

To investigate the effect of the presence of extractives in wood, extractive-free sawdust was prepared by 24hours of soxhelet extraction with ethanol-toluene 1:2 mixture.

Shredded copier paper (Sunace R100, Japan Pulp and Paper Co., Ltd) and cotton (provided from KURABO Industries LTD.) were prepared to investigate the physical properties of boards made from materials which contain infinitesimal amount of lignin and hemicellulose.

2.2 Board fabrication

The high-pressure apparatus (Hisaka made HTP-50/250) for board fabrication consists of an airtight autoclave with six compressing cylinders. An amount of 600g of the raw materials were placed inside metallic frames (190 × 290 × 230mm) , and a metallic plate was put on each material. The brief board fabrication process is listed below:

(1) Vacuuming stage

Air in the apparatus and the materials is vacuumed for 10min prior to steam injection.

(2) Softening stage

High-pressure steam is injected into the autoclave, and inner temperature of the autoclave is raised up to 120°C within 10min. The temperature is then held constant for 20min.

(3) Compressing stage

Softened materials are compressed to a target density of approximately 1g/cm^3.

(4)Fixation stage

More steam is injected into the autoclave while the materials are being compressed, and the inner temperature of the autoclave is raised up to 180°C within 10mins. The temperature is then held constant for 10min.

2.3 Testing

2.3.1 Physical property testing

The fabricated binderless boards were of dimensions of 190 × 290 mm and 10mm thick, and they were dried and cut to the various specifications for physical property tests. (in accordance with JIS A 5908-2003) A multi-strength testing machine (Imada Seisakusho Co., Ltd.SV55) was used for measuring internal bonding (IB) and bending strength (modulus of rupture, MOR). The dimensions of samples for each test were 30mm × 30mm and 10mm thick for IB and 30mm × 290mm and 10mm thick for MOR. Five samples were prepared for each test to obtain accurate data.

2.3.2 Hot water and dioxane extraction

To remove hemicelluloses and partially degraded low molecular weight lignin fractions in fabricated board, (1)hot water extraction, (2)dioxane extraction and (3)extraction consisting of both procedures were conducted. Oven-dried binderless board made from sawdust was cut to dimensions of 30mm × 30mm and 10mm thick. Method (1) and (2) were conducted by 48hours of soxhelet extraction with each solvent. Extraction (3) was conducted by 96hours of soxhelet extraction consisting of extraction (2) after extraction (1).

For each extraction, the percentages of decreases in weight and increases in thickness were measured after drying the sample.

2.3.3 X-ray diffraction

X-ray diffraction of untreated saw-dust and slush powder which was filed down from fabricated saw-dust board were measured by using X-ray diffractometer (Rigaku Rint 2000/PC) with vertical goniometer. The wavelength of X-ray was 0.154 nm (Cu $K_{\alpha 1}$ line). The degree of crystallinity index (*CrI*) was calculated by the following Segal's equation [5].

$$CrI = (I_{020} - I_{am}) / I_{020} \times 100\ (\%)$$

I_{020}: the intensity of (020) cellulose crystal plane

I_{am}: the intensity of amorphous region of cellulose

3. RESULTS AND DISCUSSION

3.1 Comparison between sawdust and wood-fiber

Comparison of physical strength of each fabricated boards is shown in Table 1. By converting fabrication material from sawdust to wood-fiber, the physical strength of fabricated board was improved, and its IB and MOR increased by 30.6%, and 255% respectively. Especially, the improvement of MOR was remarkable, and even though the fabricated board does not contain any chemical adhesives, this value satisfies JIS specification for MDF type30 boards which is the strictest standards for MDF.

Digital microscopic observation of fabricated boards represented complex entanglement of fabric on wood-fiber board surface while sawdust board had rather flat surface (Fig 1). It indicates this entanglement is occurring in lengthwise and crosswise directions and consolidating in board's physical strength. This observation suggests that fabric entanglement is deeply involved in the binding mechanism of board fabrication.

Table 1 IB and MOR comparison of each board

Material	IB(MPa)	MOR(MPa)
Sawdust	0.980	11.5
Wood-fiber	1.28	40.8
Extractive-free sawdust	1.31	12.0

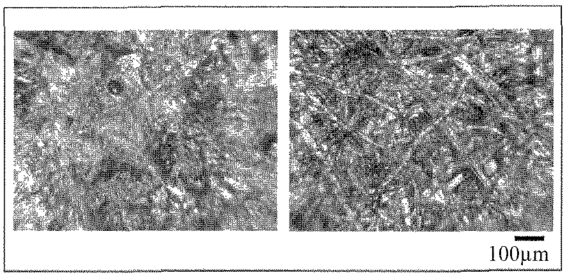

Fig.1. Digital microscopic images of fabricated boards
Left: sawdust board, Right: wood-fiber board

3.2 Comparison between sawdust and extractive-free sawdust

The physical strength of extractive-free sawdust board outpaced sawdust board (Table 1). Significant increase of IB by 33.7% indicates that extractives in wood are not involved in its binding mechanism or rather inhibiting its adhesion. Subtle increase of MOR by 4.34% can be considered as the result of particle size and its shape. By removal of extractives which seem to inhibit the adhesion, the particles could form robust entanglement to compressive direction which enhanced IB. However, it did not produce the same effect to crosswise direction because its size and shape are the same as regular sawdust while wood-fiber formed complex entanglement because of its fibrous structure. Since this experiment was conducted with only single specie, depending on extractive components, the possibility of existence of which possess adhesive-like effect on fabrication remains. However, this result emphasizes that the effect caused by extractives is not the main binding factor but an additional strength, even if there are some extractives possess adhesive-like effect.

Results of percentage increases in weight and thickness (thickness swelling, TS) when samples were immersed in cold water for 24 hours are shown in Fig. 2. Since the percentage of weight increase indicates the percentage of water absorption, the result shows that extractive-free sawdust board absorbed water more efficiently. As a consequence, TS value of extractive-free sawdust board was raised by 37.4%. This thickness swelling is mainly attributed to the swelling of hygroscopic cell wall components and partly caused by some other factors. Frequent swelling and shrinking of woody particles which make up the board increase the risk of deterioration of internal bonding. Thus, extractives in woods play a role in supporting board's dimensional stability by their water repellant-like characteristics. In addition, extractives which exhibit antibacterial activity are not negligible, such as thujic acid from Western Red Cedar (*Thuja plicata*) [6, 7]. Depending on raw materials of fabrication, resistance against biodegradation can be expected by containing these components in the board.

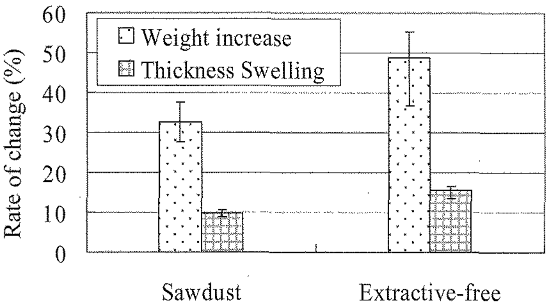

Fig.2. Weight increase and TS comparison between sawdust board and extractive-free sawdust board

3.3 Hot water and dioxane extraction

The values of percentages of decrease in weight (WD) and increase in thickness (TE) caused by each extraction are shown in Table 2 and Table 3, respectively. Hemicelluloses are easily hydrolyzed to lower molecular weight products such as oligosaccharides by high-pressure steam treatment while lignin degradation is rather slower [8]. The result of hot water extraction showed a larger weight decrease by removal of hemicelluloses, reflecting this phenomenon. Some sugars seem to be extracted by dioxane according to the difference of WD value between the extraction (2) and dioxane extraction of extraction (3). The values of WD and TE for dioxane extraction observed in extraction (3) indicate the actual alteration caused by the loss of partially degraded low molecular weight lignin. Since each value was measured after drying the samples, WD value indicates the amount of component which is extracted by each procedure, and TE value indicates the thickness expansion caused by the deterioration of the internal bonds as a result of losing its constituents. The occurrence of thickness expansion suggests that these constituents are involved in its binding mechanism. Especially, larger value of TE caused by removal of low molecular weight lignin shows that this component possesses adhesive-like activity to the board fabrication. TE value caused by removal of sugars was not significant. However, its value is not still negligible by hypothecating that volume shrinkage occurs by loss of board component. The board appearance before and after extraction are shown in Fig. 3. Even though little expansion of thickness was observed, its cubic structure was perfectly maintained. This emphasizes that the board structure is maintained by other components while lignin and hemicelluloses are providing additional strength.

Table 2 Percentages of decrease in weight (WD) and thickness expansion (TE) for each extraction procedure

	WD (%)	TE (%)
Hot water (1)	11.4	3.63
Dioxane (2)	5.04	9.77

Table 3 Percentages of WD and TE for extraction (3) HW: hot water extraction DX: additional dioxane extraction

WD (%)			TE (%)		
HW	DX	Total	HW	DX	Total
11.4	1.85	13.3	3.63	6.22	9.85

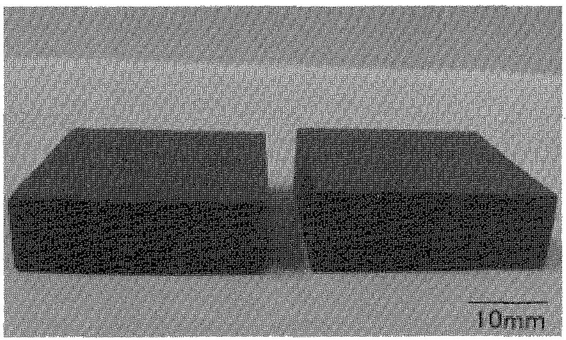

Fig.3. Board sample appearance before and after extraction (3) Left: before, Right: after

3.4 Board fabrication from copier paper

To specify the main binding factor of board fabrication, copier paper was chosen as a material which is predominantly comprised of cellulose and containing infinitesimal amounts of lignin and hemicelluloses. The board appearance and physical properties of fabricated paper board are shown in Fig.4 and Table 4, respectively. Fabrication of boards from paper was possible, and it was containing certain physical strength. Compared with the boards fabricated from sawdust, its physical strength was drastically decreased. These decrements can be considered as the result of loss of woody components such as lignin, hemicelluloses and extractives. The significant increase in TS value indicates that the paper boards contain a high water absorption rate which is a crucial disadvantage to dimensional stability against water. Even though a significant decrease in physical properties was observed, the paper boards were still strong enough to satisfy the MDF type 5 board requirements of JIS standard. In addition, boards can be fabricated from cotton (Fig.4). The successful fabrication of boards from materials virtually consisting of only cellulose emphasizes the fact that cellulose is playing a major role in the binding mechanism of binderless board fabrication.

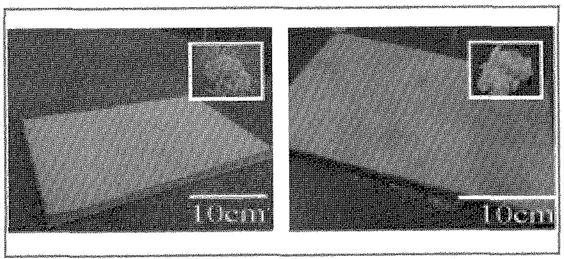

Fig.4. Fabricated boards from copier paper and cotton
Left: paper board, Right: cotton board
The small pictures at the upper right corner of each board shows respective raw materials.

Table 4 Physical properties of fabricated paper board and MDF type 5 JIS standard for each property
MOE: modulus of elasticity

	IB (MPa)	MOR (MPa)	MOE (GPa)	TS (%)
Paper board	0.36	5.53	0.81	47.9
MDF type5 JIS standard	≥ 0.2	≥ 5.0	≥ 0.8	-

3.5 Changes in cellulosic crystalline structure

X-ray diffraction patterns of each sample are shown in Fig 5. The calculated degrees of crystallinity index from this data were 48.2% for untreated sawdust and 58.0% for slush powder from the fabricated board. This result indicates that cellulose crystallinity increased during the board fabrication process.

It has been reported that high-pressure steam treatment results in the increase in cellulose crystallinity and the transformation of its crystal structure. [9-11]. This phenomenon permits the permanent fixation of transformed cellulosic structure, and the technology has been applied to compressed wood and fabric materials [12, 13]. Structural fixation of compressed wood is caused by this phenomenon as a result of high-pressure steam treatment while the wood is being compressed.

The increase in cellulose crystallinity observed in the board samples strongly emphasizes that the permanent fixation of transformed cellulosic crystalline structure caused by high-pressure steam is deeply involved in its fabrication mechanism. The deformation and the entanglement of particles or fibers caused by compression are permanently fixed by high-pressure steam. Consequently, the entanglement of particles would be fixed so that it causes adhesion between particles.

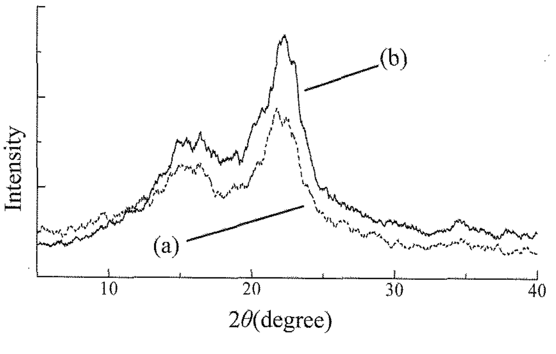

Fig5.X-ray diffraction patterns of each sample
(a):untreated sawdust (b):slush powder from board

4. CONCLUSION

The significant increase in physical properties of fabricated board by converting its raw material from sawdust to wood-fiber and successful fabrication of board from materials virtually consisting of only cellulose indicate that cellulosic structural fixation is deeply involved in its fabrication mechanism.

Physical properties of the board made from extractive-free sawdust indicated the importance of extractives presence in the boards and providing dimensional stability even though they are not directly related to the binding mechanism. Hemicelluloses and lignin appear to have adhesive-like activity as shown from the results after extracting them. However, the fact that the structure of the boards remained unchanged even after extraction shows that the fixed entanglement of cellulose was maintaining its cubic structure.

As the fabrication mechanism of binderless board, fixed cellulosic entanglement between particles forms the framework of board structure, and the other wood components provide additional strength to the board with their adhesive-like activity or water repellant-like activity. In other words, fixation of cellulosic entanglement caused by increase of its crysrallinity and transformation of crystal structure is the main binding factor of binding mechanism. Even though it is possible for the binderless board to be fabricated from cellulose alone, the combination of these additional factors allows the fabrication of further robust and dimensionally stabilized boards which can have a wider range of application.

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