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# Dimensional Stabilization of Particleboard by Formaldehyde Treatment

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# ホルムアルデヒド処理によるパーティクルボードの寸法安定化

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## Résumé

A vapor phase treatment with formaldehyde (formalization) was applied to the dimensional stabilization of three types of commercial particleboard: urea-formaldehyde (UF), melamine-formaldehyde (MF), and phenol-formaldehyde (PF) types. Dimensional stability, hygroscopic property, and mechanical properties of the treated particleboard were compared with those of the other wood-based materials. By a sulfur dioxide catalyzed formalization up to 24hr at 120°C, thickness swelling and linear expansion decreased with reaction time irrespective of the types of particleboard. The antiswelling efficiency (ASE) reached 70 and 60% in thickness and linear directions, respectively. Even a low waterproofing UF type particleboard, the dimensional stability in water soaking at room temperature was enhanced to the same degree as that for UF type plywood. The restraint of swelling was revealed not only in water soaking state but also below fiber saturation point, and the equilibrium moisture content decreased to about a half of the untreated one. These results show that the decrease of moisture adsorptive property contributed effectively and largely to the dimensional stabilization of the particleboard. The modulus of rupture (MOR) in dry state decreased by 20 to 30% when ASE reached at the level of 70%, whereas the water saturated state MOR and the modulus of elasticity (MOE) was unchanged and increased by 20 to 40%, respectively. On the other hand, the loss of internal bond was much more serious than that of bending strength, especially in UF type particleboard.

## 要　旨

尿素－ホルムアルデヒド（UF）系，メラミン－ホルムアルデヒド（MF）系，及びフェノール－ホルムアルデヒド（PF）系の3種類の樹脂接着剤を用いて作られた，市販のパーティクルボードに対して，ホルムアルデヒド処理を適用し，寸法安定性の改良を試みた。処理試片について，寸法安定性，吸湿性，及び機械的性質を測定し，他の木質材料との性能の比較を行った。その結果，二酸化イオウを触媒とした，120°Cにおける24時間までの処理で，厚さ膨潤率，及び線膨潤率は，パーティクルボードの種類に関係なく，反応時間とともに低下し，抗膨潤能（ASE）は厚さ方向で70%，面方向で60%に達した。耐水性の低いUF系のボードでさえ，冷水浸せきにおける寸法安定性は，UF系樹脂接着剤で作られた屋内用合板に匹敵する程度にまで向上した。膨潤の抑制効果は，水浸せき時だけでなく，繊維飽和点以下においても発現し，平衡含水率は無処理の約半分にまで低下した。これらの結果は，水分吸着性の低下がパーティクルボードの寸法安定化に大きく寄与していることを示す。常態の曲げ強さは，70%のASEレベルで

20～30%低下したが，湿潤状態の曲げ強さは無処理とほとんど変わらず，また曲げ弾性係数は20～40%増大した。これに対して，剝離強度の低下は，とくにUF系ボードにおいて深刻であった。

## 1. Introduction

The demand for wood-based composites such as fiberboard and particleboard increases more and more because of the lack of timbers for plywood. However, those composites are inferior in mechanical properties as well as dimensional stability, and their use is limited to the furniture and interior materials. When the composites are used for floor and wall, their dimensional instability is troublesome, although mechanical strength is less important. For instance, the dimensional instability in the linear direction causes serious curvature and gap as a result of repeated moisture adsorption and drying. Moreover, the dimensional change in thickness direction, which is larger by one order than the linear direction, brings the warp and/or creak in furnitures. Therefore, it is indispensable to improve the dimensional stability of these wood-based composites to the comparable degree obtainable for the plywood, when we intend to use them as the alternative substances of the plywood.

The author1) has reported that the dimensional stability of medium-density fiberboard (MDF) increased remarkably by a formaldehyde treatment (formalization). The vapor phase reaction such as the formalization is advantageous for MDF, because it is produced in dry state throughout the manufacturing processes, and it has high permeability to the gaseous reagents. The same situation will be the case also for the particleboard.

In this study, the formalization was applied to the particleboard which were prepared with various types of adhesive resins, and the dimensional stability, hygroscopic property, and mechanical properties were compared with those of plywood.

## 2. Experiment

### 2.1 *Materials and reagents*

Three types of commercial particleboard (thickness: 12㎜) produced by Eidai Co., Ltd., were used. They were assembled with urea-formaldehyde (UF) resin, melamine-formaldehyde (MF) resin, or phenol-formaldehyde (PF) resin adhesive, and their specific gravities in oven-dry were 0.71, 0.73, and 0.77, respectively. Water soaking tests and the determination of hygroscopicity were conducted with 50㎜×50㎜ test pieces, and mechanical properties were determined with the test strips of 200㎜×30㎜. A seven layer cross-laminated and 12㎜ thick plywood, which was adhered with UF resin adhesive by Eidai Co., Ltd. for indoor uses, was tested for the comparison of the properties with particleboard.

Technical grade of tetraoxane was used as the formaldehyde vapor source and sulfur dioxide catalyst was obtained from a commercial bomb.

### 2.2 *Procedure of formalization*

The formalization of the test specimens for water soaking and hygroscopicity tests was achieved by the similar method reported previously[2] in an about 3.5 liter glass vessel, and that for the strength tests was done in a 20 liter stainless steel vessel.

Prior to the treatment, the specimens were heated at 120°C for 20 min under vacuum. After evacuation of the vessel in which the specimens and tetraoxane had been placed, a given amount of sulfur dioxide was volumetrically introduced and the whole vessel was heated in an oven. The reaction was conducted at 120°C for up to 24hr. One reaction vessel contained two specimens and the reaction was repeated twice under the same conditions. Assuming that all parts of the added tetraoxane vaporizes and depolymerizes to the vaporous formaldehyde monomer, the concentration of formaldehyde in the reaction vessel corresponds to 28.6 mmol/dm$^3$, and that of sulfur dioxide is about 5.1 mmol/dm$^3$ in the standard state.

### 2.3 *Measurements after treatment*

#### 2.3.1 *Water soaking tests*

The specimens were dried for overnight at 60°C under vacuum, and their weights, thicknesses at four corners, and linear length at four edges were measured. Then the specimens were soaked in water at room temperature for 24hr (referred to cold water soaking); they were initially evacuated for about one hour so as to sink in water. After the measurements of thicknesses and linear lengths, the specimens were soaked in hot water at 70°C for 2hr (referred to hot water soaking), and followed by boiling in water for 2hr and oven drying at 105°C for several hours. The measurements of dimensions were repeated at each water soaking stage. The swelling and antiswelling efficiency (ASE) in thickness or linear direction were defined as follows:

$$\text{Swelling (\%)} = \{(L_w / L_a) - 1\} \times 100 \qquad (1)$$

$$\text{ASE (\%)} = \{1 - (S_u / S_t)\} \times 100 \qquad (2)$$

where, $L_a$ and $L_w$ are thickness (or edge length) in oven-dry state before water soaking test and that after water soaking state, respectively, and $S_u$ and $S_t$ are swelling of untreated and treated specimens in cold water soaking, respectively. Because the linear expansion did not significantly differ between machine direction and cross machine direction, the mean value was used for the swelling in linear direction. Also for the plywood, the grain direction of the surface layer was not considered, because the linear expansion did not differ significantly between two perpendicular directions.

#### 2.3.2 *Determination of hygroscopic properties*

Once oven-dried, the specimens were successively conditioned at 50, 65, and 90% R. H. and 20°C for every 3 weeks in a constant temperature and humidity chamber, and the weight, thickness, and edge length were determined. The swelling and moisture excluding efficiency (MEE) were calculated by Eq. (1) and the following equation, respectively.

$$\mathrm{MEE}\ (\%) = \{1 - (M_u / M_t)\} \times 100 \qquad (3)$$

where, $M_u$ and $M_t$ are equilibrium moisture content of untreated and treated specimens, respectively.

### 2.3.3 *Mechanical property tests*

The bending test was conducted in two states: equilibrated at 20°C, 50% R. H. (referred to dry state), and water swollen for 24hr at room temperature (referred to wet state). The internal bond was determined only in dry state by the residual fragment of the bending test. The mechanical properties of untreated particleboard and plywood are summarized in Table 1.

Table 1 Mechanical properties of untreated particleboard and plywood in dry (20°C-50% R. H.) and wet states (Mean values of three specimens).

| Material (Type) | MOR (MPa) | | MOE (GPa) | | IB (kPa) |
|---|---|---|---|---|---|
| | Dry | Wet | Dry | Wet | Dry |
| Particleboard | | | | | |
| (UF) | 18.8 | 11.1 | 2.84 | 1.82 | 824 |
| (MF) | 20.6 | 14.8 | 3.33 | 2.40 | 873 |
| (PF) | 23.7 | 16.1 | 3.43 | 2.26 | 892 |
| Plywood | 39.2 | 30.6 | 3.62 | 3.43 | — |

## 3. Results and Discussion

### 3.1 *Physical characteristics and waterproofing properties*

Table 2 shows the changes in weight, dimensions, and specific gravity of the particleboard after treatment. The formalization caused up to about 5% of weight increase, 2% of thickness swelling, 0.2-0.3% of linear expansion, and resultant increase in apparent specific gravity in oven-dry state. The dimensional changes during water soaking tests were estimated on the basis of the dimensions in oven-dry state after treatment.

Figure 1 shows the swelling of samples in cold water soaking test against the reaction time. The swelling in thickness and linear directions decreased with an increase in the reaction period, and it took one day to level off the swelling. In this reaction system, the reaction rate may be controlled by the three main steps: (1) vaporization and cleavage of

Table 2 Physical property changes as the results of formaldehyde treatment for 24hr (Mean values and their 95% confidence limits of four specimens).

| Type of particleboard | Weight increase (%) | Thickness swelling (%) | Linear expansion (%) | Increase of specific gravity (%) |
|---|---|---|---|---|
| UF | 4.70 ± 0.48 | 1.80 ± 0.45 | 0.212 ± 0.090 | 2.41 ± 0.65 |
| MF | 4.71 ± 0.24 | 2.06 ± 0.20 | 0.232 ± 0.084 | 2.12 ± 0.16 |
| PF | 5.62 ± 0.35 | 2.19 ± 0.62 | 0.268 ± 0.015 | 2.80 ± 0.35 |

tetraoxane to formaldehyde monomer, (2) diffusion of the formaldehyde into reaction site, and (3) real reaction between formaldehyde and wood components. Among them, the former two steps seem to proceed rather slowly, that is, the conversion of tetraoxane to formaldehyde is not always rapid even at 120°C under reduced pressure, and thus it takes initial several hours to attain a sufficient concentration of formaldehyde. Further, though the reagent molecules reach easily to the surface of particles, the diffusion into inner reaction sites, which is not generally accelerated even at elevated temperature, is not a rapid step. Consequently, although the bondings between the surfaces of particles complete within a short period, it is required a long period to spread the reaction into inner surface of particles and to attain high dimensional stability.

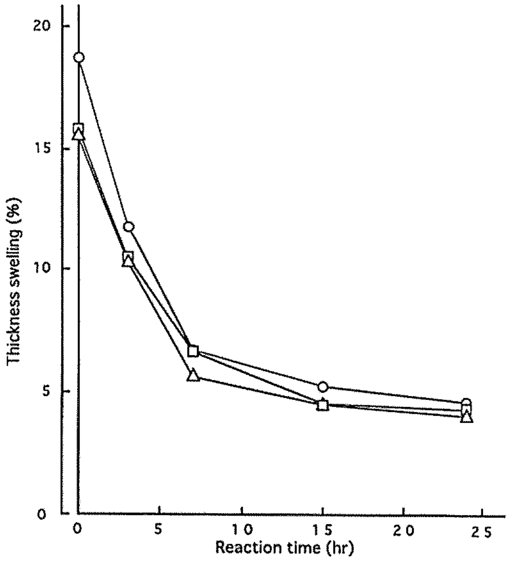

Fig. 1 Relationship between thickness swelling in cold water soaking and formalization time.
Legends: ○ UF type △ MF type □ PF type

The thickness swellings and linear expansions of various types of particleboard were plotted against water soaking steps (Fig. 2). Among the types of particleboard, the degree of thickness swelling and linear expansion of untreated particleboard did not differ significantly in the cold water soaking stage, but differed remarkably in the hot water soaking stage. Especially, the UF type particleboard swelled to the immeasurable extent. On the other hand, after the formalization for 24hr, the dimensional stability of every type of particleboard was considerably improved: their appearances were kept almost unchanged even after 2hour's of boiling and their original dimensions were recovered after final oven-drying. The ASE reached to values of more than 70 and 60% in thickness and linear directions, respectively, irrespective of the types of particleboard.

Figure 3 shows the relationship between the square of ASE and weight increase. Similar to the case for formalization of wood[3], a fairly good linear relation was observed. The ASE .went over 70% by merely about 5% of the weight increase, although the ASE of the PF type particleboard was significantly lower than those of the other types at the same level of weight increase.

The results of the water soaking test were compared among three kinds of wood-based composites which were prepared with UF resin adhesive (Fig. 4). In the thickness direction, the swelling of the particleboard formalized for 24hr was lower than that of the MDF treated with formaldehyde for 24hr[1], and was comparable to that of the plywood. On the other hand, the linear expansion of either particleboard or MDF was lower than that of the plywood.

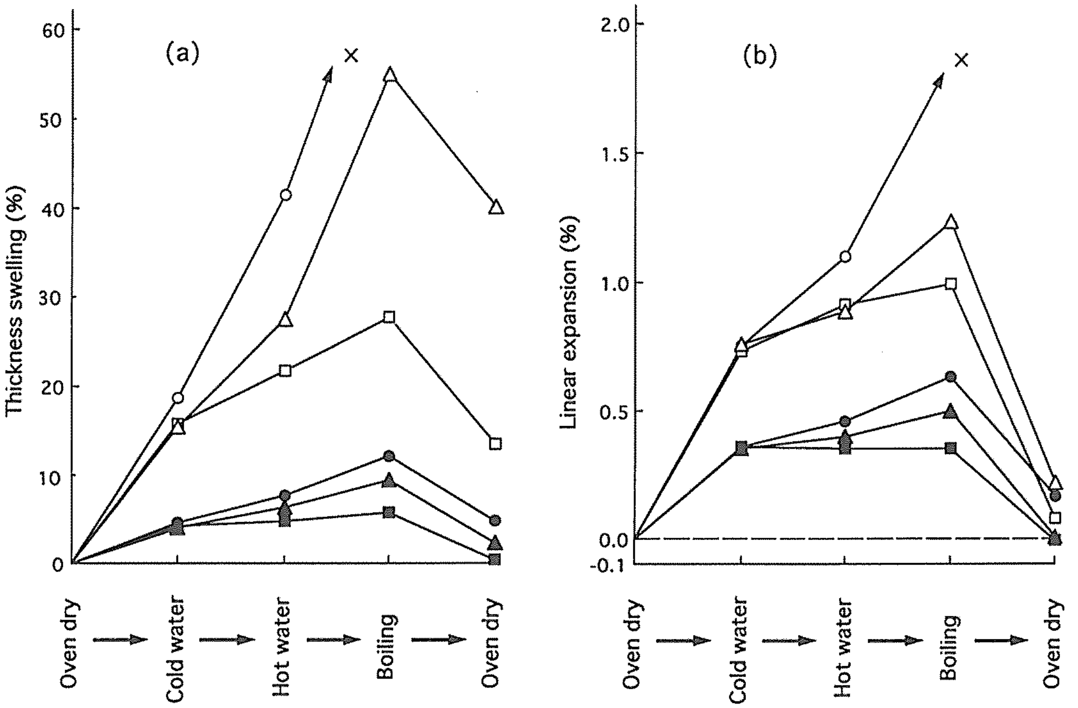

Fig. 2 Thickness swelling (a) and linear expansion (b) in successive water soaking steps for three types of particleboards.

Legends: ○ UF type (untreated) △ MF type (untreated) □ PF type (untreated)
● UF type (treated for 24hr) ▲ MF type (treated for 24hr) ■ PF type (treated for 24hr)

As stated above, though the linear expansion of the particleboard is not always absolutely large, it causes serious curvature and gap in the jointed area when the particleboard is exposed to the humidity change. Therefore, it can be said in certain sense that the dimensional stability in linear direction is more important than that in thickness direction. At present, it is satisfactory that the same level of dimensional stability that was found for the plywood was attained also for the particleboard by the formalization.

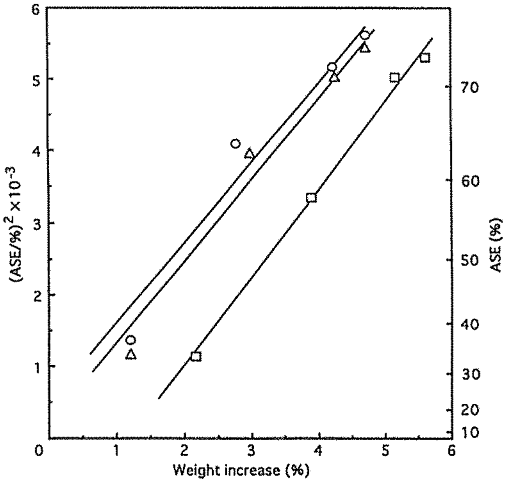

Fig. 3 Relationship between square of ASE and weight increase.

Note: Legends are same as in Fig. 1.

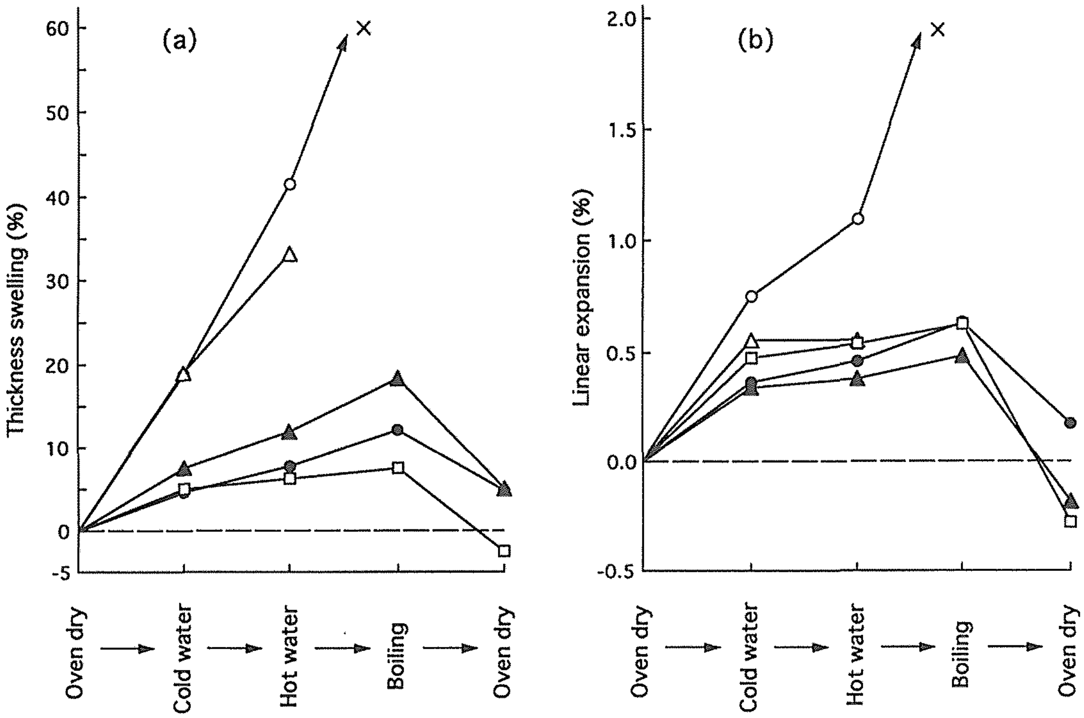

Fig. 4 Comparison of thickness swelling (a) and linear expansion (b) among three kinds of wood-based composites prepared with UF resin adhesive.

Legends: ○ UF type particleboard (untreated) ● UF type particleboard (treated for 24hr)
△ UF type MDF (untreated, cited from reference 1) ▲ UF type MDF (treated for 24hr, cited from reference 1)
□ UF type plywood

### 3.2 *Hygroscopic properties below fiber saturation point*

So far as the results of water soaking test, it was found that the dimensional stability comparable to the plywood was attained for the particleboard by the formalization. However, in practice the particleboard is rarely used in a water soaking state, and rather exposed to the repeating moisture adsorption and desorption below fiber saturation point. Therefore, it is necessary to investigate the dimensional behavior under the usual atmospheric humidity.

In Fig. 5, thickness swelling, linear expansion, and equilibrium moisture content of the UF type particleboard and plywood are plotted against relative humidity. The plots at 100% relative humidity show the values in water soaking state. The thickness swelling of untreated particleboard was about two times larger than that of the plywood, while that of particleboard treated for 24hr decreased to the same extent as for the plywood irrespective of the types of adhesive resin use.

After formalization for 24hr, the equilibrium moisture content of all types of particleboard became about a half of untreated particleboard and plywood, namely the MEE value reached about 50%. This indicates that the dimensional stabilization of the parti-

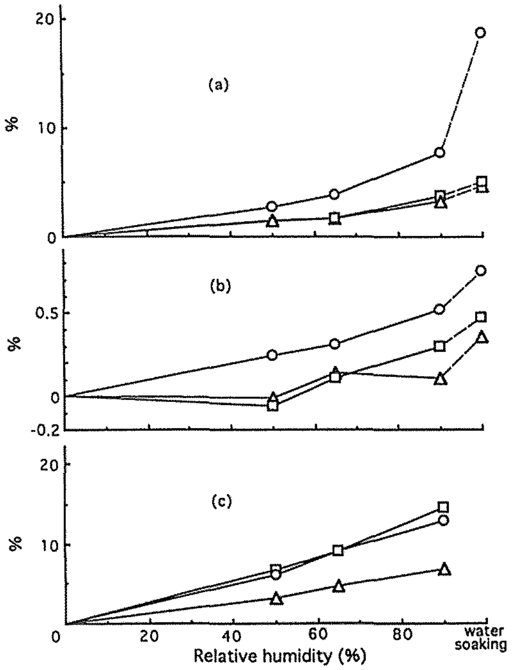

Fig. 5 Thickness swelling (a), linear expansion (b), and equilibrium moisture content (c) exposed to moisture at lower than fiber saturation point.

Legends: ○ UF type particleboard (untreated)
△ UF type particleboard (treated for 24hr)
□ UF type plywood

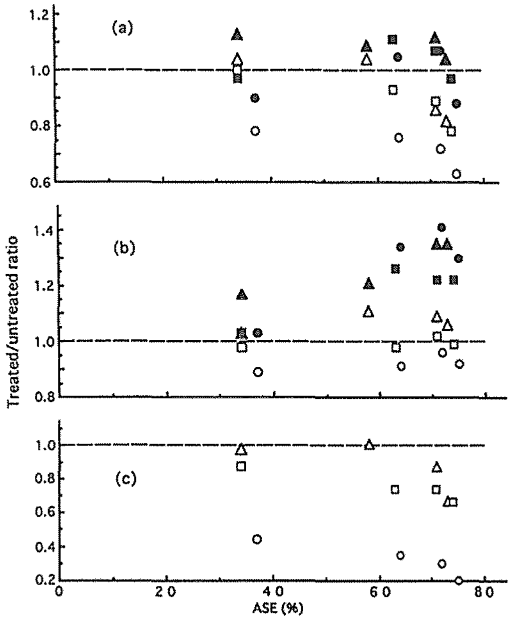

Fig. 6 Dependencies of MOR (a), MOE (b), and IB (c) in dry (open symbols) and wet state (solid symbols) on ASE.

Legends: ○ UF type □ MF type △ PF type

cleboard by the formalization is mainly resulted from the decrease in moisture adsorptive property. Though the figure does not include the result of the other types of particleboard, the moisture content of the PF type particleboard was somewhat higher than those of the other resin type ones especially in high relative humidity region. This may be due to the alkalinity of the PF resin adhesive.

### 3.3 *Mechanical properties*

In Fig. 6, the modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) were plotted against ASE. The MOR in dry state decreased by about 20% for MF and PF type particleboard and by about 30% for UF type one at 70% level of ASE compared with those for the untreated. Especially, the MOR of the UF type particleboard decreased considerably even at low ASE level. On the other hand, the MOR in water-swollen state was slightly higher than untreated irrespective of the resin types.

The MOE in dry state did not decrease significantly except for the UF type particleboard, and those in water swollen state were 20 to 40% higher than untreated at above 70% ASE level for any types of particleboard.

The internal bond decreased severely by formalization: only about 30% and 60% were retained for UF type and other types of particleboard, respectively, at 70% ASE level. The particleboard is usually overlaid by wood veneer or printed sheet, when it is used as the wall and floor. However, if the dimensional stability of overlaid material differs from the based one, internal stress generated during the drying and humidifying is probably resulted in the separation of the two materials. The decrease in internal bond will cause severely such separation of the overlaid material and/or curvature of the edge of the particleboard as a whole. Because the board is rarely used as the structural material, the decrease in bending strength is not critical, but the curvature of the surface or gap is not preferable from the viewpoint of the appearance. The restraint of the occurrence of those defects is important subject for the further use of particleboard.

## 4. Conclusions

Three types of particleboard were treated with vaporous formaldehyde under the catalysis of sulfur dioxide at 120°C. By the treatment for 24hr, the thickness swelling and linear expansion decreased remarkably, and all types of the particleboard resisted to the boiling in water for 2hr. The ASE went over 70 and 60% in thickness and linear directions, respectively. Even the UF type particleboard, the dimensional stability in the cold water soaking was comparable to a UF type plywood, although the dimensional stabilities in hot water soaking and boiling were inferior to the plywood.

By this formalization, the dimensional stability of particleboard at atmospheric humidity were also improved to the same level as that of the plywood, and the MEE reached about 50%.

The MOR in dry state decreased by the values of 20 to 30% at above 70% of ASE, whereas that in water saturated state increased to some extent when compared with that for untreated particleboard. The MOE in dry state was unchanged and that in wet state increased by 20 to 40%. On the other hand, the loss of internal bond was serious; especially that of UF type particleboard was about 70% at 70% level of ASE. The retention of internal bond is rather important subject than that of bending strength.

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## References

1) Minato, K., Kubo, N., Norimoto, M., Sasaki, H., Sawada, M., and Yamamoto, T. (1992) Dimensional stabilization of medium-density fiberboard by formaldehyde treatment. *Mokuzai Gakkaishi.* **38** (1). 67-72

2) Minato, K., Yasuda, R., and Yano, H. (1990) Improvement of dimensional stability and acoustic properties of wood for musical instruments with cyclic oxymethylenes II. *Mokuzai Gakkaishi.* **36** (11). 990-996

3) Stamm, A. J. (1959) Dimensional stabilization of wood by thermal reactions and formaldehyde cross-linking. *TAPPI.* **42** (1). 39-44