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ORIGINAL ARTICLE

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Effects of low bondability of acetylated fibers on mechanical properties and dimensional stability of fiberboard

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Abstract Fiberboards were prepared from acetylated fibers with various weight gains: 0, 4.7, 9.4, 18.5, and 24.8 weight percent gain (WPG). The effects of low bondability of acetylated fibers on mechanical properties and dimensional changes were determined. The decreased mechanical properties of acetylated fiberboard are mainly due to low bondability. To improve bending strength, high face density is also needed. The thickness swelling according to JIS and the linear expansion under relative humidity changes decreased with increasing WPG. As for accelerated weathering and the outdoor exposure test, the thickness changes in 4.7–18.5 WPG boards were much higher than those in 0 WPG board and 24.8 WPG board. The high thickness change in 4.7–18.5 WPG boards is due to low bondability. Although 24.8 WPG board also has low bondability, the thickness change of 24.8 WPG board decreased. The high dimensional stability of acetylated fibers, caused by high WPG, probably outweighs the dimensional change caused by low bondability. On the other hand, during the boiling test the thickness changes in 24.8 WPG board and the 4.7–18.5 WPG boards were higher than those in 0 WPG board. The effect of the boiling test on the boards is more severe than that seen with the accelerated weathering and outdoor exposure test; therefore, the effects of the low bondability probably cancel the effects of the high WPG. It is necessary to increase the bondability of acetylated fibers to improve the dimensional stability and the mechanical properties.

Key words Fiberboard · Acetylation · Bondability · Density profile · Dimensional stability

Introduction

Acetylation is an effective method for making wood-based composites dimensionally stable,[1–6] but the mechanical properties of the composites are decreased by this treatment.[4] Most previous studies showed that these decreases in strength are caused mainly by low bondability due to low wettability,[5,6] the loss of a substantial amount of wood per unit mass,[6] and the deterioration of fibers by acetylation.[2]

With regard to mechanical properties, although the density profile obviously plays an important role,[7] it has been ignored in most of the studies on acetylated wood-based composites so far. If density profiles are taken into account, the reasons for the decreases in mechanical properties are made clearer. The density profiles of wood-based composites are formed by thermoplastic behavior of wood during hot pressing.[8] In general, acetylation increases the thermoplastic nature of wood,[9] but it has not been reported whether this phenomenon influences the formation of the density profile. When discussing the mechanical properties of acetylated wood-based composites, it is necessary to determine the density profile.

On the other hand, acetylated wood-based composites are likely to be used under severe conditions because of their high dimensional stability. In particular, higher bondability is required if the composites are used under severe conditions for a long time.[10] If bondability is poor, the composites' dimensional changes may increase over time. However, the effects of low bondability on increasing dimensional changes under severe conditions have hardly been mentioned in most studies so far. In addition, most studies focus attention on the thickness change due to moisture content changes, and little work has been done to evaluate composites' linear expansion, which is an important property.[11] For practical purposes, thickness changes under severe conditions over a long time and linear expansion should be evaluated with respect to the low bondability.

In this study, fiberboards were prepared from acetylated fibers with various weight gains (0%, 4.7%, 9.4%, 18.5%,

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24.8%). The mechanical properties and density profiles were determined, and the effects of the low bondability on mechanical properties were studid. Moreover, a boiling test, an accelerated weathering test for 1000h, an outdoor exposure test for about 1 year, and a linear expansion test were done. The relations between acetyl weight gain and dimensional change during these tests were studied, and the effects of the low bondability on these dimensional changes are discussed.

Experimental

Raw material of fiberboards

Wood fibers from yellow cedar (*Chamaecyparis nootkatensis* (D. Don) Spach) were the raw material. Its air-dried density was 0.51 g/cm^3 according to Sudou.[12] Fibers were acetylated with vapor-phase acetic anhydride. The acetyl weight percent gains (WPG) were controlled by changing the times of acetylation. The WPGs were 0% (untreated fibers), 4.7%, 9.4%, 18.5%, and 24.8%. After acetylation the fibers were dried in an oven resulting in moisture contents of 2.96%, 2.62%, 2.32%, 1.96%, and 1.70%, respectively.

Manufacture of fiberboard

Fiberboards were made from acetylated fibers (0, 4.7, 9.4, 18.5, and 24.8 WPG). The adhesive used was melamine formaldehyde resin (U-816, P type resin according to A 5908 1994; Mitsui Chemical Co.) that had a solid content of 65%. Ammonium chloride, 1% by weight, was added to the resin as a hardener. A 10% aqueous solution of ammonium chloride was prepared and was added to the adhesive. The target resin content was 15% based on the oven-dried weight of the fiber. The adhesive solution was sprayed onto the agitating fibers, and a fiber mat was formed. Before and after spraying the fibers were disentangled by a single disk refiner. The mat was hot-pressed under 4.0MPa at 160°C for the first 1min, 2.0MPa for the next 2min, and finally 1.0MPa for 3min. Two distance bars of 10mm height were placed between plates of the press to regulate thickness. The dimensions of the boards were 23 × 22 × 1cm. The target board density was 0.7 g/cm^3. Three boards were made under each experimental condition.

Properties tested

Specimens were conditioned under 20°C and 65% relative humidity for about 1 week. The modulus of rupture (MOR), internal bond strength (IB), and thickness swelling (TS) were measured according to JIS A 5908 1994. Six replications for each series were performed.

After the TS test the boiling test was done, and the thickness change (TC_B) of the TS specimens was measured. The test order was as follows: TS (according to JIS A 5908 1994), air-drying at 20°C and 65% RH for about 1 month, boiling for 2h, cold-water soaking for 1h, and air-drying at 20°C and 65% RH for about 1 month.

The thickness change (TC_A) during the accelerated weathering test was measured on 50 × 60 × 10mm specimens for 1000h. A weather meter (35A xenon weather-o-meter; Atlas) was used; the black panel temperature was 63° ± 3°C, and the water spray interval was 18min/2h. The thickness change (TC_O) during the outdoor exposure test was performed on 100 × 55 × 10mm specimens. These specimens were exposed outdoors facing south and at an angle of 45° to the horizontal in Tsukuba, Japan for approximately 1 year (June 1999 to May 2000).

The thickness was measured after each stage. The TC_B was calculated based on the board thickness at the initial air-dried condition, and the TC_A and the TC_O were calculated based on the board thickness at the initial oven-dried condition. When the thickness of the accelerated weathering and outdoor exposure specimens was measured, the specimens were oven-dried at each stage.

After the boiling test and the accelerated weathering test, the IB of the specimens was measured. The surfaces of the specimens were damaged by these tests,[10] and so their surfaces were sanded off (3–5mm depth), and 50 × 50 × 3–4mm specimens were prepared. Three replications were performed for each series.

Linear expansion (LE) and thickness change (TC_L) of wet bending specimens before the wet bending test were measured by calipers. The specimens were first oven-dried and then conditioned under each relative humidity until the boards attained equilibrium moisture content. The LE and TC_L were calculated based on the oven-dried length and thickness. The relative humidity was changed from 0% (oven-drying), to 45%, 65%, 75%, 90%, 75%, 65%, and 45% and then oven-drying. Finally, after the LE test, the specimens were subjected to the wet bending test (test B according to JIS A 5908 1994). Three replications for each series were performed for the LE and wet bending test.

Density profile

Density profiles were obtained by sanding 0.5–1.0mm from the surface of the nondestructive portion of bending test pieces. The face and core in the density profile were defined[13] as follows: The face was defined as 0.5–1.5mm depth from the surface; the first 0.5 mm of thickness from the surface was excluded.[7] The core was defined as the innermost 0.5mm of the depth of the boards. In this study, density profiles are depicted by each density in the depth divided by the mean board density; it is defined as "relative density."

Contact angle

Solid wood of hinoki (*Chamaecyparis obtusa* Endl) was prepared for measuring the contact angle. The wood was acetylated by liquid-phase acetic anhydride and xylene heated to 120°C for 6h. Acetic anhydride was diluted with xylene to obtain several WPGs. The acetic anhydride/xylene volume ratios were 10:0, 3:7, 2:8, and 1:9; and the mean WPGs were 18.9%, 13.6%, 7.4%, and 4.8%, respectively. The contact angles of water-drops about 35s later were measured by a face contact-angle meter (Kyowa Kaimenkagaku Co.) in the axial direction; 50 water-drops were measured.

Results and discussion

Density profile

In general, the thermoplastic behavior of wood is enhanced by acetylation,[9] but this probably is not the case for acetylated fibers during hot pressing.[6] To measure the thermoplasticity of wood, the rate of temperature increase must be slow, and a long measuring time is necessary.[9] Meanwhile, when a fiberboard is made, a mat is pressed directly by high-temperature plates during a short hot pressing time. Moisture is an important factor for increasing thermoplasticity[14]: Water behaves as a plasticizer. According to our previous study,[15] the plasticity of acetylated wood is not increased by moisture. Therefore, untreated fibers are probably plasticized much more easily than acetylated fibers.

The locus of thermoplasticity during hot pressing may be observed in the density profile, as shown in Fig. 1. As the WPG increased, the slope of the density profile became more gentle, that is, the lower the WPG, the steeper the density profile. It is likely that the acetylated fibers are more difficult to plasticize by heat and vapor than untreated fibers during hot pressing. The degree of thermoplasticity of acetylated fibers depends on the WPG. Thus, the fibers with low WPGs are compressed more easily than those with high WPGs, and therefore the density profiles of low WPG become steeper.

Internal bond strength

One of the disadvantages of acetylated wood is its low wettability,[6] which probably leads to low IB. The relation between WPG and IB is shown in Fig. 2. The IB was highest at 0WPG, decreased considerably at 4.7WPG, and was nearly constant at values over 4.7WPG. This shows that even a low WPG (e.g., 4.7WPG) leads to low bondability.

The IB is generally related to core density.[16] In this study the density profile of high WPG boards had a gentle curve, and the gentle density profile implied high core density. However, owing to the different mean board densities, almost all the cores had about the same value, as shown in Table 1. With respect to the value of the core density, the IB of acetylated board was much lower, which may have been due to the low bondability between acetylated fibers caused by poor wettability.[6]

Wettability is closely related to the contact angle, which is affected by acetylation, as shown in Table 2. The contact angle at 0WPG was significantly lower (at the 5% level according to the Tukey test) than that of acetylated wood across the range 4.8–18.9WPG. The fact that the contact angle of wood at 7.4WPG is lower than 4.8 and 13.6WPG probably results from experimental anomalies and is anticipated to be of no practical significance.

Table 1. Mean board density, face density, and core density of experimental boards

WPG	Mean board density (g/cm^3)	Face density (g/cm^3)	Core density (g/cm^3)
0	0.77	0.95	0.66
4.7	0.75	0.92	0.66
9.4	0.73	0.85	0.66
18.5	0.72	0.80	0.66
24.8	0.69	0.77	0.63

The face was defined as 0.5–1.5mm depth from the surface of the boards, and the core was defined as the innermost 0.5mm of the thickness of the boards
WPG, weight percent gain

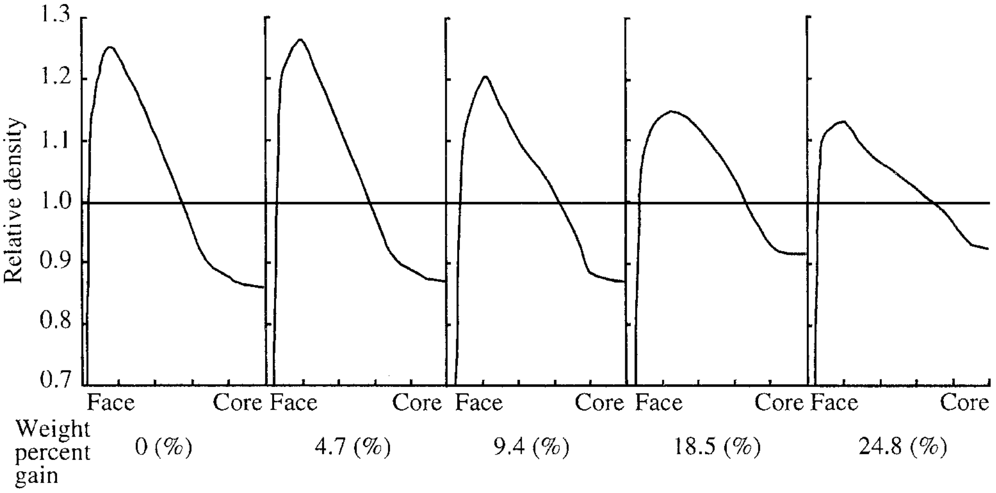

Fig. 1. Relative density profiles of experimental boards. Relative density was the density for each thickness divided by the mean board density

Table 2. Contact angle of acetylated wood with various weight percent gains

WPG	Contact angle (°)
0	24 (4.1)
4.8	33 (3.8)
7.4	28 (5.3)
13.6	33 (5.2)
18.9	31 (4.8)

The numbers in parentheses indicate standard deviations
WPG, weight percent gain

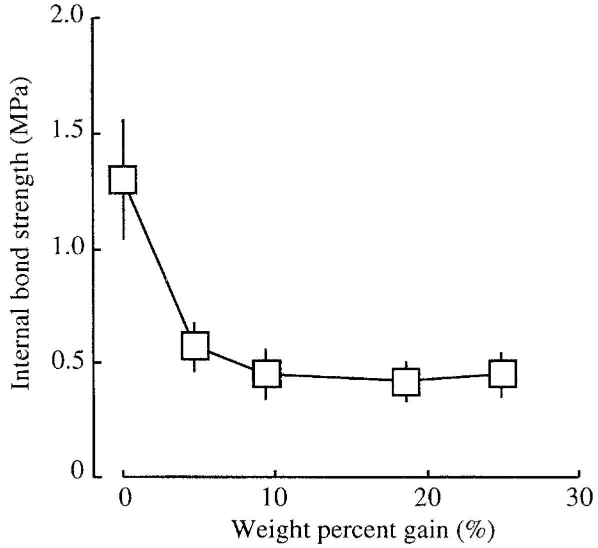

Fig. 2. Relation between weight percent gain and internal bond strength. *Vertical lines* denote standard deviations

When wood fibers are acetylated, the acetylation probably progresses gradually from the surface toward the inside. Comparing the surfaces of low WPG fibers and high WPG fibers, the degree of actylation may be almost the same, and both surfaces presumably have low wettability. Thus, even low WPG interferes with bondability.

The IB after the boiling test and accelerated weathering test is shown in Figs. 3 and 4. The manner in which IB decreased considerably at 4.7 WPG was similar to that in Fig. 2, but there was a difference in the residual ratios (IB in Figs. 3 or 4/IB in Fig. 2). The residual ratios of 0 WPG board and 4.7–24.8 WPG boards were about 80% and 17%, respectively. As the former residual ratio was much higher than the latter, it is obvious that the bondability of boards from acetylated fibers is much lower under severe conditions than that of boards from untreated fibers.

Bending strength

Figure 5 shows the relations between WPG and the specific MOR (MOR/mean board density)[17] in the air-dried and wet bending tests. In general, bending strength is directly related to face density,[7] as shown in Table 1. The air-dried specific MOR and wet specific MOR of the untreated board were the highest of all boards owing to the high bondability

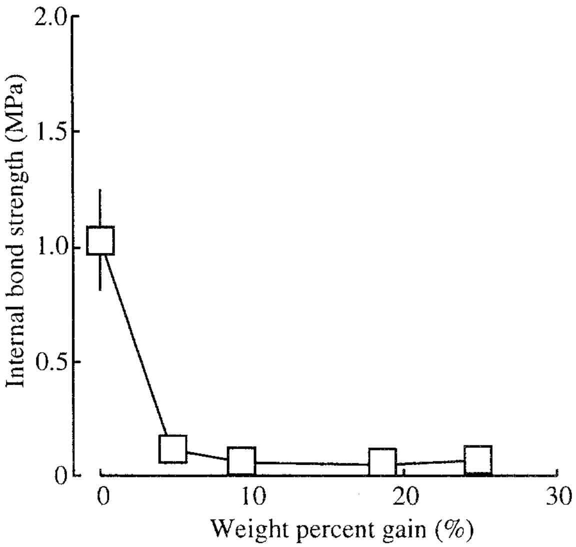

Fig. 3. Relation between weight percent gain and internal bond strength after the boiling test. *Vertical lines* denote standard deviations

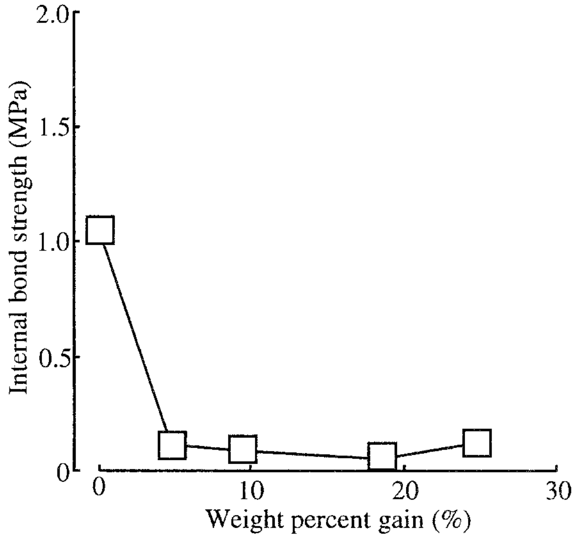

Fig. 4. Relation between weight percent gain and internal bond strength after accelerated weathering test. The standard deviations of each internal bond strength are too low to be shown on the graph

and the high face density. Both specific MORs drastically decreased, particularly at 4.7 WPG, although a marked decrease in face density was not observed. The residual ratios (wet MOR/air-dried MOR) of 0 WPG board and 4.7–24.8 boards were about 47% and 28%, respectively; and the residual ratios of 4.7–24.8 WPG boards were much lower. This is also mainly due to low bondability. At 24.8 WPG the air-dried specific MOR decreased significantly. This decrease is caused by the substantial amount of wood lost per unit mass,[6] deterioration of fibers by acetylation,[2] and low bondability.[5,6] In addition, the lower face density proves to

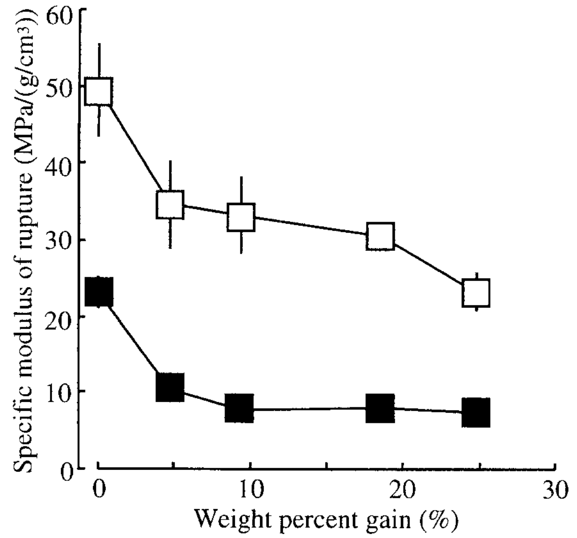

Fig. 5. Relations between weight percent gain and the specific modulus of rupture of air-dried and wet bending tests. *Open squares*, air-dried bending test; *closed squares*, wet bending test. *Vertical lines* denote standard deviations

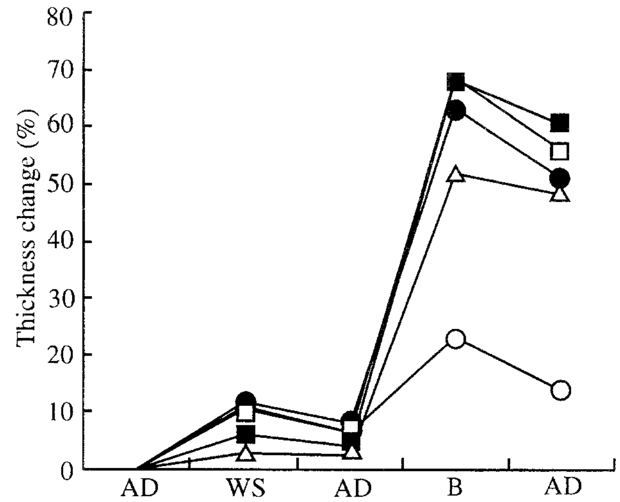

Fig. 7. Relations between weight percent gain (WPG) and thickness change (TC_B). Test order is as follows: 24h water soaking (*WS*), air-drying (*AD*), 2h boiling and 1h water soaking (*B*), air-drying (*AD*). *Open circles*, 0WPG; *closed circles*, 4.7WPG; *open squares*, 9.4WPG; *closed squares*, 18.5WPG; *open triangles*, 24.8WPG

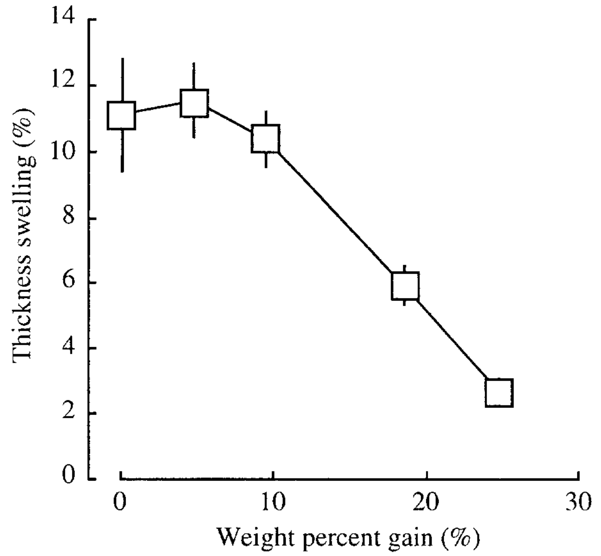

Fig. 6. Relation between weight percent gain and thickness swelling according to JIS. *Vertical lines* denote standard deviations

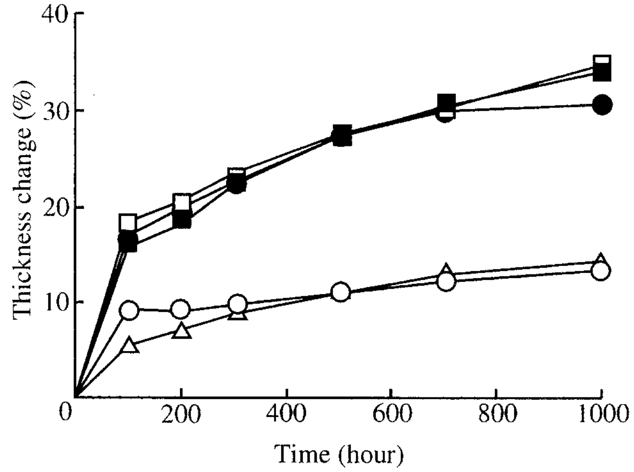

Fig. 8. Relations between weight percent gain (WPG) and thickness change (TC_A) during the accelerated weathering test for 1000h. *Open circles*, 0WPG; *closed circles*, 4.7WPG; *open squares*, 9.4WPG; *closed squares*, 18.5WPG; *open triangles*, 24.8WPG

be one of the important causes for the decrease in MOR in this study.

Thickness change

Figure 6 shows the relation between WPG and TS according to JIS. At more than 4.7WPG, the TS decreased linearly with increasing WPG, and the trend for TS was different from that of IB and MOR. The good effect of high WPG on TS is shown.

The TC_B of the boiling test is shown in Fig. 7. The TC_B of the 4.7–24.8WPG boards was much higher than that of the 0WPG board after the boiling test. This is due to the effect of low bondability; some of the fiber-to-fiber bondings may be easily cleaved by boiling (Fig. 3). Numerous studies have emphasized the high dimensional stability of acetylated board. However, acetylated fiberboard showed more extensive thickness changes especially under severe conditions such as the boiling test in this study.

The TC_A of the accelerated weathering test and TC_O of the outdoor exposure test are shown in Figs. 8 and 9, respectively. The trends of the accelerated weathering test and outdoor test were different from those of TS according to JIS. The TS decreased linearly with increasing WPG, but the TC_A and TC_O were not linearly related to the WPG. The TC_A and TC_O of 24.8WPG board were similar to those of 0WPG board, and the values were low. Despite the considerable decrease in IB (Fig. 4), the low TC_A and TC_O values for 24.8 WPG board are due to the high dimensional stability of the acetylated fibers following the high WPG; the high dimensional stability of the acetylated fibers outweighs the dimensional change caused by low bondability. Although 24.8 WPG board had low TC_O for 9 months, it may swell further because the IBs decreased drastically under severe conditions over time. In fact, the TC_O of 24.8 WPG board became higher than that of 0 WPG board over 10 months. Whether this trend continues over the longer term is currently unknown, but the experiment is being continued. On the other hand, the TC_A and TC_O of 4.7–18.5 WPG boards were higher than those of 0 WPG board and 24.8 WPG board owing to the low bondability. The effect of low bondability overcomes the effects of acetylation. Note that the specimens were oven-dried, and then TC_A and TC_O were determined. If TC_A and TC_O were determined in humid and wet conditions, 0 WPG board would probably swell more than 24.8 WPG board would swell. Although acetylation is effective in achieving high dimensional stability, the low bondability should be improved to maintain high dimensional stability.

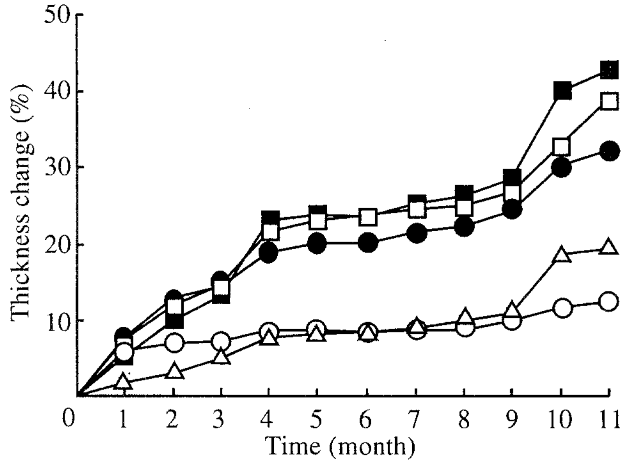

Fig. 9. Relations between weight percent gain (WPG) and thickness change (TC_O) during the outdoor exposure test for 1 year. *Open circles*, 0 WPG; *closed circles*, 4.7 WPG; *open squares*, 9.4 WPG; *closed squares*, 18.5 WPG; *open triangles*, 24.8 WPG

Linear expansion

The LE and TC_L in relation to relative humidity are shown in Tables 3 and 4, respectively. The LE and TC_L decreased with increasing WPG. The LE and TC_L of 24.8 WPG board were particularly low. The hysteresis of the 0–18.5 WPG boards was more pronounced than that of the 24.8 WPG board. The residual TC_L of the former boards was higher than that of the latter, and the former length shrunk more than the latter after absorption and desorption processes. As for the former boards, the swelled thickness caused shrinkage of the length via the Poisson effect, so the dimensional stability of the latter was higher than that of the former.

Conclusions

With respect to the density profile, it is obvious that the decreased mechanical properties of acetylated fiberboards are mainly due to low bondability. To improve bending strength, a high face density is also needed. On the other hand, low bondability may also increase the dimensional changes over time. Although acetylation is effective in achieving high dimensional stability, improved bondability could make acetylation even more effective.

Table 3. Thickness change (TC_L) of fiberboard in relation to relative humidity

WPG	Thickness change (%), by relative humidity							
	45% →	65% →	75% →	90% →	75% →	65% →	45% →	0%
0	2.08 (0.32)	3.23 (0.38)	3.71 (0.49)	7.13 (0.68)	6.34 (0.66)	5.48 (0.40)	4.43 (0.48)	0.99 (0.53)
4.7	1.66 (0.32)	2.84 (0.45)	3.48 (0.52)	6.66 (0.52)	5.90 (0.44)	5.32 (0.49)	4.20 (0.49)	1.01 (0.37)
9.4	1.28 (0.36)	2.63 (0.45)	3.18 (0.24)	6.39 (0.25)	5.71 (0.23)	4.90 (0.26)	3.77 (0.12)	0.85 (0.27)
18.5	0.96 (0.14)	1.74 (0.06)	2.04 (0.11)	4.25 (0.17)	3.98 (0.18)	3.32 (0.12)	2.13 (0.04)	0.48 (0.09)
24.8	0.50 (0.14)	1.06 (0.09)	1.46 (0.10)	2.59 (0.24)	2.28 (0.40)	1.87 (0.28)	1.32 (0.19)	−0.05 (0.05)

The numbers in parentheses indicate standard deviations
WPG, weight percent gain

Table 4. Linear expansion of fiberboard in relation to relative humidity

WPG	Linear expansion (%), by relative humidity							
	45% →	65% →	75% →	90% →	75% →	65% →	45% →	0%
0	0.39 (0.01)	0.52 (0.02)	0.57 (0.02)	0.67 (0.02)	0.62 (0.02)	0.54 (0.02)	0.41 (0.02)	−0.11 (0.01)
4.7	0.35 (0.01)	0.46 (0.01)	0.51 (0.01)	0.63 (0.01)	0.57 (0.01)	0.52 (0.06)	0.35 (0.01)	−0.10 (0.01)
9.4	0.32 (0.01)	0.43 (0.01)	0.48 (0.01)	0.61 (0.01)	0.56 (0.01)	0.47 (0.01)	0.34 (0.01)	−0.07 (0.02)
18.5	0.21 (0.02)	0.31 (0.02)	0.35 (0.02)	0.47 (0.03)	0.42 (0.03)	0.37 (0.03)	0.25 (0.02)	−0.06 (0.02)
24.8	0.16 (0.02)	0.25 (0.02)	0.30 (0.02)	0.40 (0.01)	0.36 (0.02)	0.30 (0.01)	0.21 (0.01)	−0.03 (0.01)

The numbers in parentheses indicate standard deviations
WPG, weight percent gain

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