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Improvement of dimensional stability and weatherability of composite board made from water-vapor-exploded wood elements by liquefied wood resin impregnation

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Abstract High-density and high-resin-content boards were produced by phenolic resin impregnation into board materials prepared by the water-vapor-explosion process (WVE) to develop high-durability wood composite boards for exterior use. Wet–dry cyclic tests and accelerated weathering tests were conducted, and the fundamental properties were determined to examine the effect of resin impregnation on board qualities. Bending and internal bond strength of resin-impregnated boards (I-board) satisfied the criterion for 18-type particleboard described in JIS A 5908. Thickness swelling (TS) after 24-h water immersion was approximately 2%. Resin impregnation improved the dimensional stability of the boards. In wet–dry cyclic testing, TS of I-board was the same as that of plywood. The retention ratio of modulus of rupture of I-board was large; thus, I-board had high bond durability. Color change of I-board was less than that of ordinary particleboard after a 500-h accelerated weathering test. I-Board had lower surface roughness than boards produced by a spray application method (S-board) and higher water repellency, although the difference in resin contents of the face layer was small. Thus, it is suggested that the surface properties and weatherability of I-board were improved by impregnation of phenolic resin. High-density and resin-impregnated boards made from the WVE elements are expected to withstand actual exterior use.

Key words Exterior board · Water-vapor-explosion process · Liquefied wood resin impregnation · Wet–dry cyclic test · Accelerated weathering test

Introduction

The dimensions of wood composite boards change easily when soaked with water. Thus, these boards are hardly used in outdoor environments in Japan. New demands for recycled waste wood could be generated if wood composite boards were made suitable for use in outdoor environments.

The water-vapor explosion (WVE) process is one of the methods used to recycle waste wood from house demolitions.[1,2] In this process, large timbers such as pillars are used as raw materials, and water-saturated wood is exploded from inside due to high water vapor pressure and temperature by hot pressing. The WVE process can produce elements of various sizes from solid wood without using wood cutting tools such as axes, saws, chisels, and drills.

In our previous study,[3] we investigated wood composite boards made from raw materials prepared by the WVE process for exterior use. High-density and high-resin-content, single-layer and three-layer boards were produced to obtain high durability, and the fundamental properties of these boards were examined. High-density boards showed high decay resistance and satisfied the criterion of bending strength for 18-type particleboard described in JIS A 5908. However, thickness swelling (TS) after 24-h water immersion was relatively large. Therefore, it was necessary to improve the dimensional stability of these boards.

There are numerous reports on improving the dimensional stability of wood by impregnation of phenolic resin.[4–8] However, few studies have reported on the durability of phenolic-resin-impregnated boards.[5,8] In the present study, high-density (1.1 g/cm^3) boards were produced from sugi (*Cryptomeria japonica* D. Don) elements that were prepared by the WVE process and were impregnated with phenolic resin synthesized from liquefied wood in order to improve the properties, dimensional stability, and weatherability for exterior use. It was expected that penetrability of sugi elements could be improved by high-temperature steam treatment during the WVE process. The effect of impregnation treatment on the properties of these boards

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was determined by comparing these boards with the three-layer boards prepared by spray application. The changes in bending properties and surface properties were also evaluated by a wet–dry cyclic test and an accelerated weathering test, respectively.

Materials and methods

High-density and high-resin-content boards were produced by the following two methods.

1. Phenolic resin was impregnated into the elements of the face layer and sprayed onto the elements of the core layer (I-board).
2. Phenolic resin was sprayed onto the elements of both surface and core layers (S-board).

Preparation of WVE elements

The elements were prepared by the WVE process from water-saturated sugi wood (moisture content 100%–200%) as raw materials I-board and S-board. The length and width of the WVE elements were less than 200 and 10 mm, respectively. The conditions of explosion were the same as those described in a previous report.[3]

In the case of I-board, the elements of the face and core layer were sieved through a 2.24-mm mesh. The elements that passed through the sieve were used for the face layer, and the retained elements were used for the core layer. In the case of S-board, elements less than 5 mm in width were used for the face layer and those with widths in the range of 5–10 mm were used for the core layer.

Synthesis of adhesive

Liquefied wood resin was used as the adhesive. The synthesis method was as follows. First, the mixture of wood meal and phenol was heated at 150°C for 1 h with sulfuric acid catalyst, and the wood meal was liquefied into phenol. In the next stage, formaldehyde and phenol-liquefied wood were reacted, and the initial condensation product was obtained.[9] This resin was called liquefied wood resin. The characteristics of liquefied wood resin were: pH, 10.4; viscosity, 93.0 mPa·s (25°C); solid content, 43.0%.[3] There are few reports on the bond durability of phenol–formaldehyde resin from liquefied wood.

Manufacture of boards

The dimensions of boards were 150 × 150 × 8 mm. The conditions of hot pressing were as follows: pressing pressure, 3.0 MPa; pressing time, 15 min; plate temperature, 200°C. These conditions were the same as those described in the previous report.[3]

In the case of I-board, the target density of the boards was 1.1 g/cm^3. The weight ratio of the face layer to the core layer was 1:4 (face:core:face = 1:8:1), and the three-layer mats were randomly formed. The elements of the face layer were dipped into the liquefied wood resin (43.0% solid content), and vacuumed at 20 kPa for 30 min. They were then dried in air for 1 day. Mean weight percent gain (WPG) of the face-layer elements by impregnation was approximately 45%. The target resin content of the core layer was 30% by spraying.

In the case of S-board, the target densities of the boards were 0.7 and 0.9 g/cm^3. The weight ratio of the face layer to the core layer was 1:1 (face:core:face = 1:2:1), and the elements were used to randomly form the three-layer mats by hand. The target resin content of the face and core layers were 40% and 20%, respectively. The boards were hot-pressed and then conditioned for 1 week at room temperature.

Measurement of vertical density profile

The vertical density profiles of the boards were measured by a density profiler (DA-X, GreCon). The dimensions of the specimens were 40 × 50 × 8 mm, and the densities were measured at intervals of 0.02 mm across the board thickness.

Measurement of board properties

The fundamental properties of the I-board were measured to estimate the effect of impregnation of the resin. The mechanical properties [bending properties and internal bond strength (IB)] were measured in accordance with JIS A 5908 by using a Tensilon 5-TMC (Orientec). Modulus of rupture (MOR) and modulus of elasticity (MOE) were measured on 130 × 40-mm specimens at a 120-mm span. The measurements were conducted for 12 replications. IB was measured on 20 × 20-mm specimens for 6 replications. TS and water absorption (WA) were assessed on 40 × 40-mm specimens after 24-h immersion in distilled water, and 6 replications were conducted. Moreover, the changes in TS and WA of I-board by immersion in water were measured every 3 days until attainment of the saturated state.

Wet–dry cyclic tests

A wet–dry cyclic test was conducted based on the accelerated aging test method of the West Coast Adhesives Manufacturers Association (WCAMA)[10,11] using specimens with dimensions the same as those used for the bending test to estimate the bond durability. Motoki et al.[12] investigated the properties of flakeboards by a similar wet–dry cyclic test. One cycle of the wet–dry cyclic test comprised immersion in water at 80°C for 3 h followed by drying in an oven at 80°C for 3 h. Thirty cycles of the cyclic test were conducted, and the changes in bending properties, thickness, and weight were measured. Commercial structural softwood plywood (JAS for Structural Plywood, 2003.2: bending property, class 2; bonding quality type, special) and oriented strand board (OSB) (JAS for Structural Panel, 2003.2: bending strength, class 4) were also tested as controls. Thickness and density of both panels were 10 mm and 0.7 g/cm^3, respectively.

Accelerated weathering tests

An accelerated weathering test was conducted using specimens with the same dimensions as those used for the bending test; the weathering test was performed in accordance with JIS K 5600 7–7 by using a Xenon Weatherometer Ci4000 (Atlas). The conditions of the test were: irradiance, 550 W/m^2 in the wavelength range of 290–800 nm; black panel temperature, 65°C; chamber temperature, 40°C; one test cycle, 102 min of light exposure followed by 18 min of light exposure with water spray. The test cycle was repeated for up to 500 h. The changes in color difference (ΔE^*), water repellency (WR), and surface roughness (SR) were measured at 0, 100, 200, and 500 h.

Single-layer particleboards made of commercial wood particles were used as control. The dimensions of particleboards were 150 × 150 × 8 mm, the target density was 1.1 g/cm^3, and the target resin content was 30%. Liquefied wood resin was sprayed and the mats were formed. The mats were hot-pressed under the same conditions used for I-board.

The color difference was measured using spectrophotometers (CM-508i, Minolta; NF-333, Nippon Denshoku). ΔE^* was calculated from L^*, a^*, and b^* in accordance with JIS K 5600 4-6. WR was measured by using the method reported by Kiguchi et al.[13] WR indicates the degree of water penetration, so the higher the value of WR, the more difficult it is for water to penetrate the boards. The measurement of SR was conducted using a CCD laser sensor LK-080 (Keyence). The conditions for measurement were: distance between sensor and specimens, 80 mm; diameter of laser spot, 70 µm; and scan speed, 5 mm/s. The SR was measured at 25-µm intervals across the board width. The data was processed using SPCANA version 4.71 with a band pass range of 0.5–75 Hz.

Results and discussion

Bending properties and IB of I-board

The fundamental properties of I-board and S-board are shown in Table 1. The MOR of I-board was almost the same as that of S-board (target density 0.9 g/cm^3), although the density of I-board was 0.2 g/cm^3 higher than that of S-board. Unlike MOR, the MOE of I-board was smaller than that of S-board (0.9 g/cm^3). The IB of I-board was significantly higher than that of S-board (0.9 g/cm^3). On the basis of these results, the strength properties of I-board satisfied the criterion for 18-type particleboard described in JIS A 5908 (standard for particleboard).

Vertical density profiles of boards

The difference in strength properties between I-board and S-board was investigated with the vertical density profiles of boards (Fig. 1). The mean densities of I-board and S-board were 1.08 and 0.89 g/cm^3, respectively. The densities of the face layer and core layers of I-boards were 1.23 and 1.03 g/cm^3, respectively. The density ratio of the face layer to the core layer was 1.19. On the other hand, the densities of the face layer and core layers of S-board were 1.08 and 0.74 g/cm^3, respectively. The density ratio was 1.46. It is considered that the density ratio of I-board was smaller because the target density of I-board was higher and the effect of increasing the density by hot pressing was relatively lower.

The difference in the face-layer density between I-board and S-board (0.9 g/cm^3) was approximately 0.1 g/cm^3. Therefore, it is considered that the MOR values of the boards were similar. The increase in the ratio of the face-layer density was relatively smaller for high-density boards than for low-density boards. It is assumed that the difference in MOR is hard to induce as the target density becomes higher, especially for high-density boards with density over 1.0 g/cm^3.

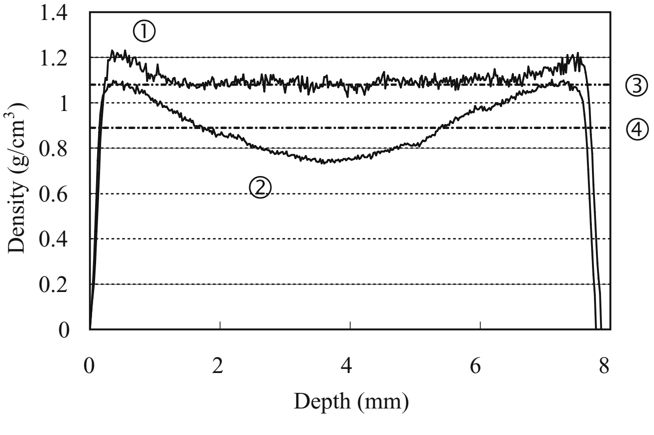

Fig. 1. Vertical density profile of I-board and S-board. *1*, I-Board; *2*, S-board (0.9 g/cm^3); *3*, mean density of I-board; *4*, mean density of S-board

Table 1. Properties of I-board and S-board

Board type	Mean density (g/cm^3)	MOR (MPa)	MOE (GPa)	IB (MPa)	TS (%)	WA (%)
I-Board	1.07 (0.03)	22.6 (2.7)	3.9 (0.3)	1.12 (0.44)	1.9 (1.0)	14.6 (2.5)
S-Board 0.9 g/cm^3	0.86 (0.01)	22.7 (4.4)	4.4 (0.6)	0.16 (0.05)	13.5 (0.9)	23.5 (1.2)
S-Board 0.7 g/cm^3	0.67 (0.02)	12.8 (3.9)	2.5 (0.6)	0.21 (0.09)	13.2 (1.5)	33.2 (1.9)

Values in parentheses are standard deviations

MOR, Modulus of rupture; MOE, modulus of elasticity; IB, internal bond strength; TS, thickness swelling; WA, water absorption

On the other hand, I-board had superior IB to S-board, because the core-layer density of I-board was over 1.0 g/cm^3 and higher than that of S-board. Moreover, it was thought that I-board had more bonding points at the core layer than S-board because the resin content of the core layer of I-board was higher.

Change in TS and WA after water soaking

The TS of I-board was 1.9% after 24-h water immersion (Table 1). Kajita[5] reported that TS of particleboards produced by impregnation of phenolic resin was also low. Our present findings are in accordance with these previous results. It was reported that the low molecular weight phenolic resin impregnated into wood fibers and cured, the resin closed off –OH groups of wood from water molecules, and decreased the dimensional change of wood.[4] In addition, in our preliminary experiment, the WPG of the WVE elements by impregnation was about 10% higher than that of the wood particles for commercial particleboards under the same impregnation condition. This suggested that the WVE elements had high penetrability, which induced a significantly small value of TS of I-board. The WA of I-board was also small (14.6%). On the other hand, TS of S-board was over 12%.

It is necessary to examine the effect of moisture over a long term. Figure 2 shows the change in TS and WA of I-board during continuous immersion in water until the boards were saturated with water. TS and WA of I-board under the water-saturated condition were 4.8% and 28.6%, respectively. These values were less than those of both the OSB and plywood. TS and WA of I-board reached the water-saturated condition after 9 days of immersion. This result indicates that the dimensional change of I-board was small for long-term immersion. Thus, I-board had greater dimensional stability than S-board, and the resin-impregnation treatment was considered to be an effective technique to improve water resistance.

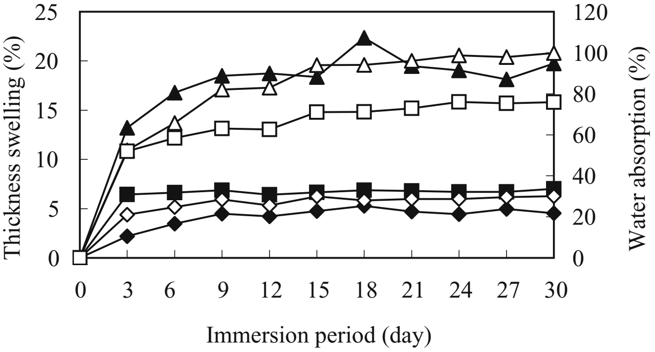

Fig. 2. Changes in thickness swelling and water absorption of specimens by continuous water immersion. *Filled diamonds*, thickness swelling (TS) of I-board; *filled squares*, TS of plywood; *filled triangles*, TS of oriented strand board (OSB); *open diamonds*, water absorption (WA) of I-board; *open squares*, WA of plywood; *open triangles*, WA of OSB

Change of board properties during wet–dry cyclic tests

I-Board is shown to have high water resistance. However, it is also necessary to consider the bond durability under repetitive wet and dry conditions to evaluate the durability for exterior use. The changes in TS and MOR during the wet–dry cyclic test were investigated and shown in Figs. 3 and 4. TS of I-board was the same as that of the plywood and less than that of the OSB. Because wood composite boards were hot-pressed, springback generally occurred to absorb moisture, and the bonding points between the elements were destroyed; subsequently, high TS of the boards was induced. Because I-board showed the same TS as the plywood, I-board had high water resistance.

Furthermore, because the increase in TS was small from the completion of the 1st treatment cycle to the 30th treatment cycle, the deterioration of bond quality was less than that of the other boards and bond durability was superior. Kajita[5] showed that TS of the particleboards made from chips impregnated with low molecular weight phenolic resin was half of that of control boards after the ASTM accelerated aging test. The results of the study of Kajita were in agreement with the results of our wet–dry cyclic test. Moreover, the change in WA during the wet–dry cyclic test showed a similar tendency to that of TS.

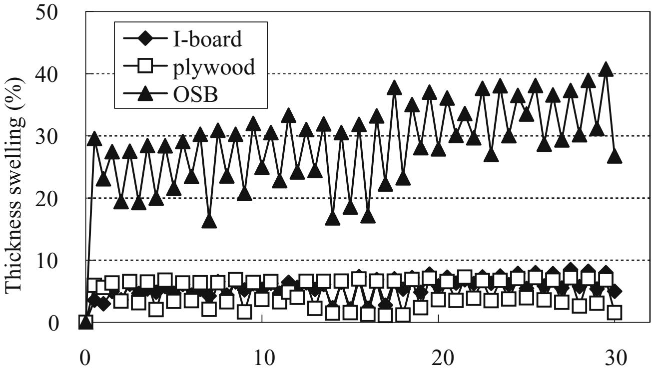

Fig. 3. Changes in thickness swelling of specimens during wet–dry cyclic treatment

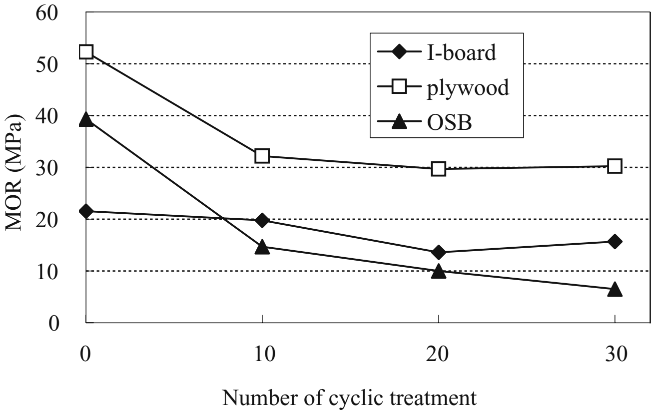

Fig. 4. Changes in modulus of rupture (*MOR*) of specimens during wet–dry cyclic treatment

It is thought that the reduction in bending strength after the cyclic test was small because of the small dimensional change of I-board. The initial MOR of the OSB was greater than that of I-board. However, the MOR of OSB reduced considerably and was less than that of I-board after 10 treatment cycles. The retention ratio of MOR at the 30th cycle treatment was 70%, 20%, and 60% for I-board, OSB, and plywood, respectively. The MOR of the OSB with large TS (Fig. 3) reduced considerably, while the MOR of I-board and plywood with small TS (Fig. 3) reduced slightly. It is assumed that the deterioration of bond quality was less and the MOR was hardly reduced with decreasing TS during the cyclic test. A similar tendency was observed for the change in MOE during the cyclic test. Saito et al.[14] reported that the bending properties of plywood had higher retention ratios than OSB. The changes in TS, MOR, and MOE of I-board during the wet–dry cyclic test, indicate that I-board has greater bond durability and might withstand exterior use.

Change in board surface properties by accelerated weathering treatment

As described above, it appears that I-board was superior in terms of bond durability and the bond strength of I-board was difficult to reduce. However, it is important to evaluate surface properties and durability if I-board is intended for exterior use. Therefore, an accelerated weathering test was conducted using water and ultraviolet (UV) radiation, and the changes in surface properties were evaluated.

The color of weathered specimens was changed after exposure to UV radiation, and cracks were formed on the surface of the specimens because of swelling and shrinking. The change in color difference is shown in Fig. 5. The initial color of I-board was darker than that of particleboard (PB) because of the presence of resin-rich elements on the surface. Because the photooxidative products in the resin-rich portion were difficult to leach out with water, the change in the color difference of I-board surface might be smaller than that of PB. The same results were obtained for S-board. It is thought that the color difference of S-board (0.7 g/cm^3) was larger than those of other S-boards or I-boards because of a small absolute quantity of resin. Because there was less resin on the surface of PB, the photooxidative products could easily leach out. Therefore, the color difference of PB increased with increasing weathering time.

The SR profiles of I-board, S-board (0.9 g/cm^3), and PB (1.1 g/cm^3) are shown in Fig. 6. SR at weathering times of

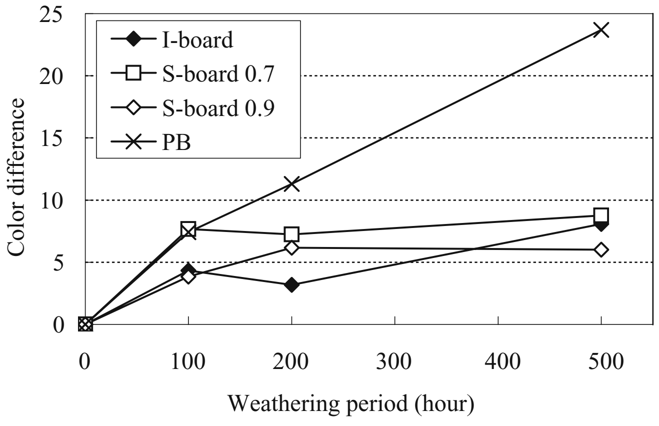

Fig. 5. Changes in color difference of accelerated weathering specimens. *PB*, Particleboard

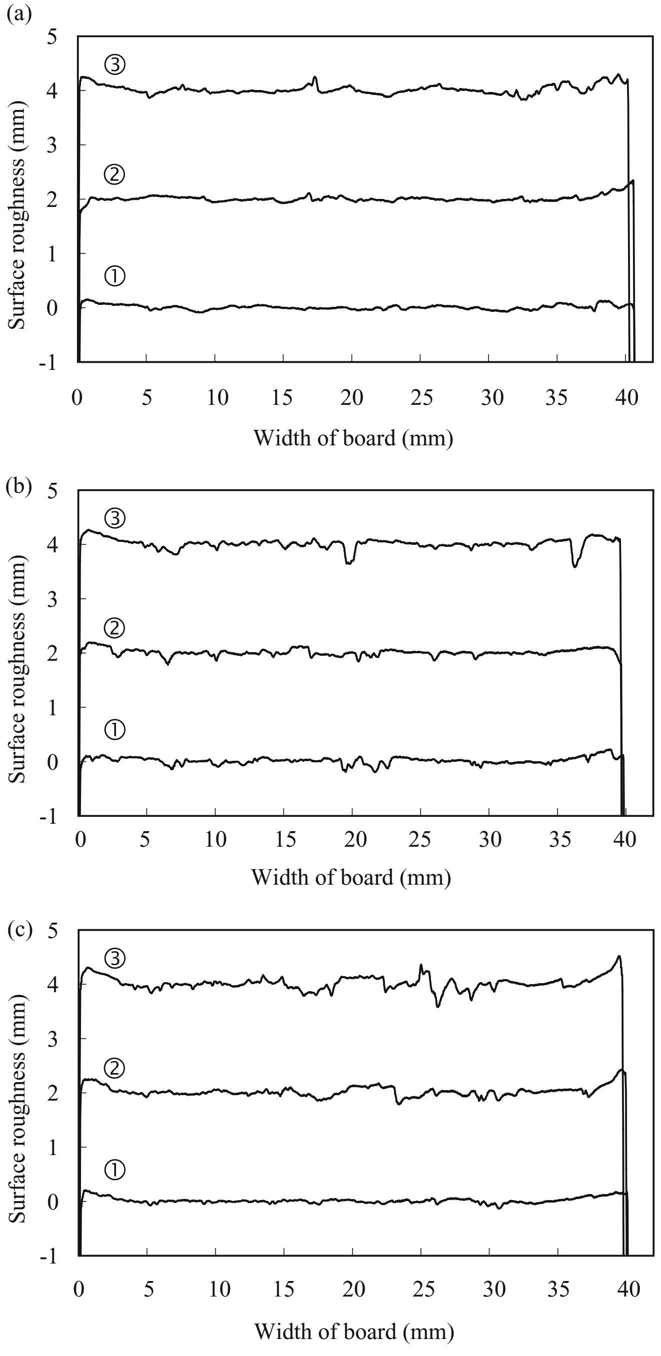

Fig. 6a–c. Surface roughness of accelerated weathering specimens of **a** I-board, **b** S-board, and **c** PB. *1*, No treatment; *2*, 200-h treatment; *3*, 500-h treatment

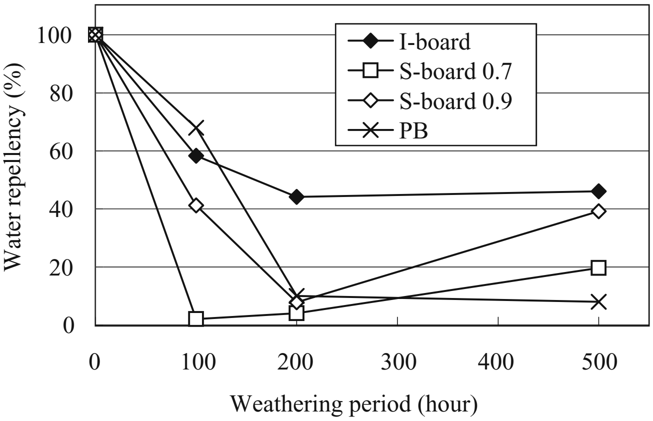

Fig. 7. Changes in water repellency of accelerated weathering specimens

0, 200, and 500 h are shown as baselines $y = 0$, $y = 2$, and $y = 4$ mm, respectively. The results showed that SR increased with increasing trea tment time in all specimens. In untreated specimens, the difference in the altitude of the surface was 0.22, 0.42, and 0.26 mm for I-board, S-board, and PB, respectively. After 500 h of treatment, SR changed to 0.42, 0.60, and 0.78 mm, respectively. The increase in SR of I-board was smaller than that for the other boards. In addition, the dimensional change of I-board by the wet–dry cyclic test was small (Figs. 2, 3), which made progression of surface deterioration difficult. On the other hand, because thickness swelling of S-board and PB was larger, cracks on the surface formed easily and then progressed. Moreover, the differences in the surface density and resin content also induced differences in SR.

The results for WR are shown in Fig. 7. WR values of I-board and PB were larger than that of S-board for 100-h treatment. It is considered that low SR makes water penetration into the board difficult. Hence, WR of I-board and PB might be larger than that of S-board. After 200 h of treatment, WR of S-board and PB reduced considerably. It is thought that the decrease in WR of I-board was less than that of S-board, because SR of I-board was smaller. Although the resin content of the face layer of I-board and S-board was 45% and 40%, respectively, the surface performances showed a large difference between the I-board and S-board. Therefore, it is suggested that the liquefied wood resin penetrated into the WVE elements by impregnation treatment and improved the properties. Because there was little change in the properties of I-board after the accelerated weathering test, it is expected that the durability of I-board is high and its properties are maintained for the long term in actual outdoor environments.

Conclusions

High-density and high-resin-content wood composite boards for exterior use were produced from WVE elements by impregnation of liquefied wood resin to improve dimensional stability. The durability of boards was evaluated by some accelerated aging tests. The following conclusions are drawn from the above experimental results:

1. The initial MOR of I-board satisfied the criterion of mechanical strength for 18-type particleboard described in JIS A 5908. I-Board showed small TS and saturated WA values. Hence, the boards were subjected to an impregnation process to improve their fundamental properties in order to withstand outdoor use.
2. In the wet–dry cyclic test, TS of the I-board was lower than that of OSB, and the retention ratio of MOR of I-board was large. Thus, the bond durability of I-board was excellent.
3. The change in color of I-board was less than that of ordinary particleboard after a 500-h accelerated weathering test. Furthermore, the SR of I-board was less than that of S-board, and the WR of the former was higher. Although the resin content of the face layer of I-board and S-board was 45% and 40%, respectively, the surface performances of the boards were very different.
4. It is suggested that the liquefied wood resin penetrated into the WVE elements by impregnation treatment and improved the properties. It is expected that the high-density and high-resin-content boards produced by resin impregnation will withstand actual exterior use.

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