The Durability of Diphenylmethane Diisocyanate– and Phenol-Formaldehyde–Bonded Japanese Cypress Particleboard

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Abstract

Particleboard was fabricated using hinoki (Japanese cypress: *Chamaecyparis obtsusa* Endl.) particles and different amounts of diphenylmethane diisocyanate (MDI) resin (2%, 4%, 6%, and 9%) and phenol-formaldehyde (PF) resin (6% and 10.2%). Four accelerated aging treatment tests were conducted to compare the durability performance of MDI-bonded boards with that of PF-bonded boards: two Japanese industrial standard wet-bending tests, a European cyclic moisture test (V313), and the American Society for Testing and Materials (ASTM) six-cycle test (ASTM-6c). The bending strength of the wet MDI boards increased with increasing resin content, and the strength retention after the treatment was greater than 50 percent, which corresponded to that of high-moisture-resistant boards. After the V313 and ASTM-6c treatments, the thickness swelling of the MDI boards with 6 percent resin content was less than 5 percent, whereas that of the PF boards was about 10 percent. The thickness of the specimens varied according to the resin content and at each step of the treatment for both the V313 and the ASTM-6c tests. The resin content equivalence for the two types of resin was determined based on the internal bond (IB) strength after aging treatments of both types of board. The MDI board required only 50 to 70 percent of the resin required for a PF board with a comparable IB quality. The superior performance of the MDI-bonded board was clear after all the tests except for the boiling treatment, in which no significant difference was observed between MDI and PF.

The use of thinning from Japanese cypress (hinoki: *Chamaecyparis obtsusa* Endl.) trees for the production of particleboard is very important in Japan. The material properties of hinoki are better than or similar to that of Japanese cedar (sugi: *Cryptomeria japonica* D. Don). There have been several studies of the properties of wood-based panels made with sugi (Mallari et al. 1989, Suzuki et al. 1994); however, there are few published reports on wood-based materials constructed using hinoki (Kojima et al. 2009).

The practical importance of diphenylmethane diisocyanate (MDI) resin as a binder for mat-formed panel products is increasing because the formaldehyde emissions from amino-resin binders may cause sick house syndrome (Gallagher 1982, Nanami et al. 1998, Godish 2000). Attempts to improve MDI resin have been driven by the desire to reduce costs and develop new products requiring a stronger or more durable resin. Although some researchers revealed durability performance of polymeric MDI (pMDI)–bonded panels (Jackowski and Smulski 1988; Bernard et al. 1994a, 1994b; Charles and Okkonen 1998), the durability effect of pMDI-bonded boards used for residential construction is not widely recognized in Japan.

It has been shown that isocyanate groups can react with hydroxyl groups. Although recent work demonstrated that urethane linkage to wood occurs only under extreme conditions (Zhou and Frazier 2001), it is clear that pMDI-bonded boards have outstanding properties. Board performance equal to or better than that of PF-bonded boards is achievable at lower densities and lower resin levels by using somewhat faster press cycles (Deppe 1977, Johns et al. 1981, Loew and Sachs 1981). Even though some investigations of MDI for bonded boards have been carried out

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(Udvardy 1979; Saito et al. 1985; Grozdits et al. 1987; Hawke et al. 1992, 1993), MDI-bonded particleboard in Japan is not recognized as a P-type board, which is widely accepted as a durable or a waterproof binder for wood-based materials. Thus, a test to compare the durability of MDI and PF resin boards is required.

The objective of this study was to compare the durability of MDI- and PF-bonded boards to determine whether the bond quality of MDI and PF resins is equivalent. Particleboard was fabricated for this purpose using laboratory-made hinoki particles with different MDI and PF resin content (RC). To evaluate the durability of the two types of board, four accelerated aging treatments were conducted: two Japanese Industrial Standard (JIS) wet-bending tests (Japanese Industrial Standards Committee [JISC] 1994), European Standard V313 (European Committee for Standardization [CEN] 1993), and the American Society for Testing and Materials (ASTM) six-cycle test (ASTM 1993).

Materials and Methods

Board fabrication

Hinoki particles were prepared in the laboratory and used to compare the effect of the two resin types. Hinoki logs with diameters greater than 20 cm were waferized by a disk flaker and then hammer milled to reduce the particle size. The particles were screened using a sieve (opening = 1 mm) to remove fines and dried to a uniform moisture content of 2 percent. Figure 1 shows the resulting hinoki particles. An emulsified pMDI adhesive (Cosmonate M-201, Mitsui Takeda Chemicals Co., Ltd.) was used as a binder, and wax (EJ80/147, Exxon Mobil Co., Ltd.) was applied as an additive. Commercial liquid PF resin (PL-222, Sunbake Co., Ltd.) was used for comparison.

The controlled conditions of the board manufacturing were as follows: target density, 0.65 g/cm^3; homogeneous single-layer panel construction; and wax dosage, 0.5 percent. The RC and wax dosage were based on the ovendry weight of the screened particle furnish. Four RC levels were set for the MDI: 2, 4, and 6 percent (control condition) and 9 percent (for solid basis). Only two RC levels were used for the PF resin (6% and 10.2% for solid basis). An RC of 6 percent for the PF resin was used for direct comparison with 6 percent MDI, and 10.2 percent PF (which was 1.7× 6 percent MDI) was applied to measure the ratio of the bond effectiveness. We selected the 1.7× value somewhat arbitrarily, but it is based on previous experience in our laboratory.

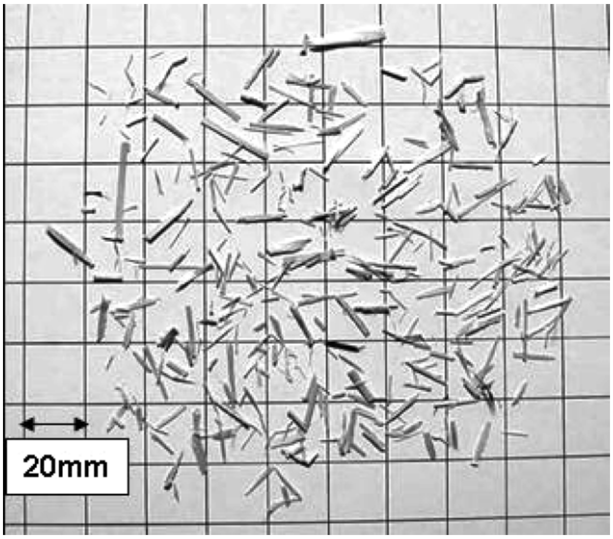

Figure 1.—Hinoki particles as particleboard furnish.

Adhesive was sprayed using an air-spray gun (ANEST IWATA Co., Ltd.) for blending purposes onto the dried particle furnish inside a laboratory scale rotary blender. Mat forming was performed over a metal caul plate by hand-felting in a 600 by 600-mm forming box in a ventilated area. The particles were oriented randomly. The mat moisture content before pressing was approximately 10 percent and achieved by adding water to the resin. The fabricated boards were 600 by 600 by 11.7 mm. Press stops were used to control the final thickness of the board. A laboratory scale hot press was used in this research. These processes were the same for pMDI and PF boards.

The press schedule was as follows. The platen temperature was 180°C, and the maximum pressure applied in the first step was 3.0 MPa for 1 minute. The pressure was reduced to 1.5 MPa during the second step, to 1.0 MPa during the third step, and to 0.5 MPa during the fourth step, each for 1 minute. The total press time was thus 5 minutes. Three panels were produced for each resin condition and resin type.

Mechanical tests

After conditioning the boards at 25°C and 65 percent relative humidity for more than 2 weeks, a bending test was conducted according to JIS-A 5908 (JISC 1994) using a 50 by 250-mm specimen. The modulus of elasticity (MOE) and the modulus of rupture (MOR) from bending were determined using four samples selected randomly under each condition. In addition, the internal bond (IB) strength was determined using 10 randomly selected samples of the JIS method (JISC 1994).

Accelerated aging treatments

Specimens 50 by 50 mm were conditioned for pretreatment in a dryer at 60°C for 24 hours. Four accelerated aging tests were selected for evaluation. The ASTM-6c method, specified in ASTM D 1037 for mat-formed panel products (ASTM 1993), consists of six cycles of the following treatment sequence: immersion in 49°C water for 1 hour, steaming at 93°C for 3 hours, freezing at −12°C for 20 hours, drying at 99°C for 3 hours, steaming at 93°C for 3 hours, and drying at 99°C for 18 hours. The V313 method is a European standard cyclic moisture test (CEN 1993). The test specimens were exposed to three cycles of the following treatment: immersion in water at 20°C for 72 hours, freezing at between −12°C and −20°C for 24 hours, and drying at 70°C for 72 hours. The JIS-A treatment is defined as hot-water immersion for 2 hours followed by water immersion for 1 hour, and the JIS-B treatment is boiling-water immersion for 2 hours and then water immersion for 1 hour (JISC 1994). Seven replicate samples selected at random from each board type were tested. The specimen thickness was measured after each step for each aging treatment cycle. The thickness swelling (TS) was calculated on the basis of the original dimensions.

After treatment, the specimens were dried at 60°C for 24 hours followed by a week of reconditioning, after which the

IB tests were conducted. Three randomly selected samples were tested after the JIS-B treatment using 50 by 250-mm specimens.

Results and Discussion

Bending properties

The average density of all boards made in this research was 0.656 g/cm^3 (SD = 0.018 g/cm^3). When we compared both 6 percent resin contents, the MOR of MDI-bonded boards was about 40 MPa, while that of PF-bonded boards was about 32 MPa. These means are statistically different ($P < 0.01$). This result indicates that the bending property of MDI-bonded boards is superior to that of PF-bonded boards. The bending properties of MDI- and PF-bonded boards were compared under wet and dry conditions. Figure 2 shows the effect of resin type and RC. The MOR of MDI-bonded boards with 9 percent RC was about 45 MPa, while that of 2 percent RC boards was greater than 30 MPa. These values were far larger than bending strength of JIS requirement for 18-type particleboard (JISC 1994). The mean values of the MOE for these boards (data not shown) were 4.3 and 3.6 GPa for 9 and 2 percent RC, respectively. An elastic constant of 4.3 GPa is comparable to the MOE specified for structural Class 2 plywood in the Japanese Agricultural Standard (Ministry of Agriculture, Forestry and Fisheries 2000). The bending properties of PF-bonded boards were relatively smaller than those of MDI boards, even though the values were sufficient to meet the JIS requirement for particleboard (JISC 1994). The MOR of MDI-bonded boards after the boiling treatment of the JIS-B method was in the range of 18 to 24 MPa for 2 to 9 percent RC. Figure 2 shows that the MOR retention, which is the ratio of the measured MOR to the MOR of the control condition, was between 50 and 60 percent. These results indicate that hinoki could be used for high-performance particleboard. One of the reasons for these higher bending properties was the shape and quality of the hinoki elements, waferized from logs at a thickness of 0.5 mm followed by hammer milling to ensure a thin and relatively uniform configuration. The average length and width of the particles were 15 and 3.2 mm, respectively. Using this type of furnish is not common in commercial particleboard production. The average length and width of the particles typically used in commercial particleboard were 10 and 2.0 mm, respectively. Thus, improving particle quality could improve the mechanical properties of the board.

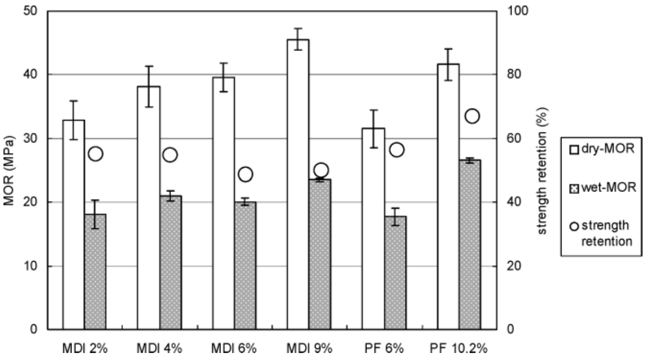

Figure 2.—Effect of resin content on the modulus of rupture (MOR) and strength retention after the JIS-B treatment for MDI- and PF-bonded hinoki particleboards.

Change of thickness during accelerated aging treatments

The ASTM-6c test is one of the most severe accelerated aging treatments and has been used for high-moisture-resistant types of mat-formed panel products. Each cycle consists of six steps: immersion in water at 49°C for 1 hour (hot-water soaking step), steaming at 93°C for 3 hours (first steaming step), freezing at −12°C for 20 hours (freezing step), drying at 99°C for 3 hours (first drying step), steaming step at 93°C for 3 hours (second steaming step), and drying at 99°C for 18 hours (second drying step).

Figure 3 shows the thickness changes during this treatment. In Figure 3, to simplify the data view, no error bars are shown. The coefficients of variation (COV; %) were 5.9 (MDI 2%), 8.8 (MDI 4%), 9.4 (MDI 6%), 9.5 (MDI 9%), 8.7 (PF 6%), and 9.4 (PF 10.2%).

All types of boards fabricated for this study survived the ASTM-6c treatment. The TS value was the largest at the first steaming step in each treatment cycle, and the order of TS was as follows: at the freezing step, the second steaming step, the hot-water soaking, the first drying step, and the second drying step. The test results were similar to those obtained by Kajita et al. (1991) for commercial boards. The TS of the MDI board at the first steaming step was in the range 10 to 35 percent, and the TS after drying was about 3 to 20 percent. The TS of PF-bonded boards with 6 percent RC was between the TS of MDI 2 and 4 percent boards. Also, the TS of PF 10.2 percent boards were between that of MDI 4 and 6 percent boards.

The V313 test method was originally developed in Europe and was submitted to the International Organization for Standardization (ISO) as method for testing the moisture resistance of wood-based panels (Suzuki 2001, ISO 2003). Each cycle consists of three steps: immersion in water at 20°C for 72 hours, freezing at between −12°C and −20°C for 24 hours, and drying at 70°C for 72 hours. All the steps and cycles required 3 weeks to complete; the ASTM-6c test took about 2 weeks.

Figure 4 shows the TS values during the V313 treatment for all the board types. After the 3 days of immersion in water, the thickness increased with the amount of water and decreased slightly with the freezing treatment for 1 day. In Figure 4, to simplify the data view, no error bars are shown. The COV (%) were 9.1 (MDI 2%), 6.3 (MDI 4%), 15.2 (MDI 6%), 10.8 (MDI 9%), 7.6 (PF 6%), and 10.1 (PF 10.2%).

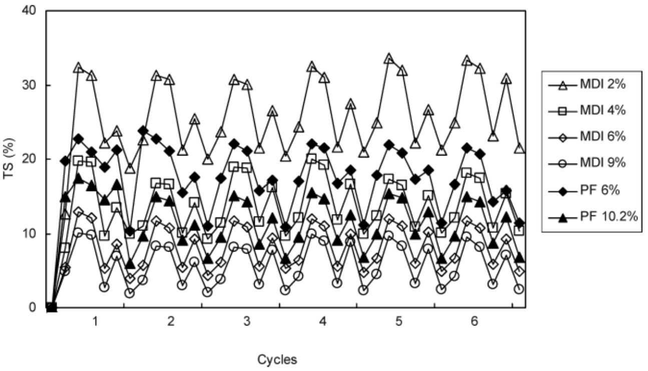

Figure 3.—Change in thickness of the hinoki particleboard for each step of the ASTM-6c accelerated aging treatment.

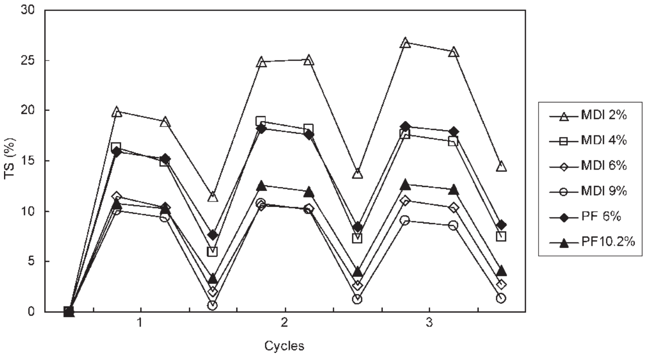

Figure 4.—Change in thickness of the hinoki particleboard for each step of the V313 cyclic test.

The TS values of these boards increased gradually during successive cycles. The TS of MDI-bonded boards under wet conditions ranged between 10 and 25 percent, which was less than that in the ASTM-6c treatment. The TS of PF 6 percent boards was found to be almost the same as for MDI 4 percent boards, and the TS of PF 10.2 percent boards was close to that of MDI 6 percent boards. These results show that MDI-bonded boards are more stable than PF-bonded boards with the same RC level.

Comparison between MDI and PF resin-bonded boards

The effect of RC on TS was evaluated to compare the durability performance of MDI and PF resins. Figure 5 shows the relationships after ASTM-6c and V313 treatments using the largest TS under wet conditions during the last cycle of the treatments. The TS of MDI-bonded boards decreased from around 30 to 10 percent for both aging treatments with increasing RC, whereas the TS of PF boards was significantly larger than that of MDI boards with the same RC level. The TS of PF boards with 6 percent RC was about 20 percent under the wet condition of the V313 treatment; this value was close to that of MDI boards with 4 percent RC. Thus, as far as TS under wet conditions is concerned, PF 6 percent boards and MDI 4 percent boards are comparable. The TS of a PF 6 percent board after ASTM-6c treatment was 22 percent, and this value corresponded to a board that would be made with 3.6 percent MDI resin. Similarly, an MDI RC equivalent to a PF 10.2 percent RC was obtained to about 5 percent by projecting the TS value of a PF board on the TS curve of an MDI board. Figure 6 shows the TS at each RC level to confirm the aging effects of the two different treatments and shows the relationship between the TS results after the V313 and ASTM-6c treatments under wet conditions. A linear relationship was observed for MDI boards with different RC values. The TS values for PF boards were on the same line so that the position of PF RC corresponding to MDI resin was easily determined.

Figure 7 shows the effects of RC on TS caused by the JIS-A and JIS-B treatments. The equivalence of the bond quality between MDI and PF resins was determined in the same manner as before. PF with a 6 percent RC after JIS-A corresponded to MDI of about 2 percent based on a simple comparison. PF with a 10.2 percent RC was projected to an MDI value of about 3 percent. The JIS-A treatment TS values clearly demonstrated the superiority of MDI resin. However, this was not clear for the JIS-B treatment. A simple arithmetic calculation shows that PF RC values of 6 and 10.2 percent corresponded to MDI RC values of 5.3 and 8.4 percent, respectively. The advantage of MDI resin was not as significant as after the JIS-A treatment. A similar dependency was observed in the graph of the relationship between the TS results after the JIS-A and JIS-B treatments shown in Figure 8. Although the TS of MDI- and PF-bonded boards appeared on the same straight line in Figure 5 for the V313–ASTM-6c relationship, the TS values of PF-bonded boards after the JIS treatments formed a different line from the MDI line (open symbols). One of the reasons for this

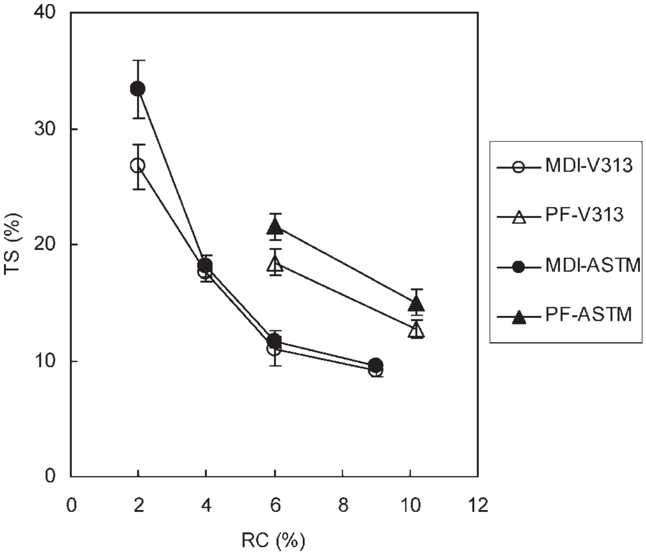

Figure 5.—Effect of resin content (RC) on thickness swelling (TS) during V313 and ASTM-6c treatments. Each TS value was the largest TS under wet conditions in the last cycle of each aging treatment.

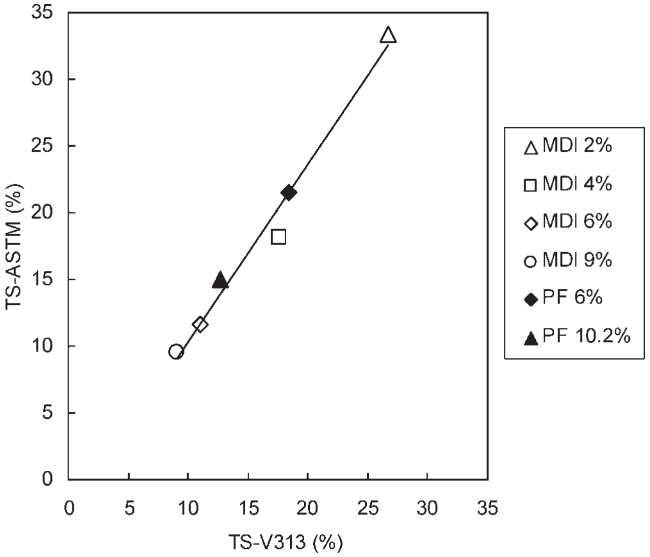

Figure 6.—Relationship between thickness swelling (TS) under wet conditions of two aging tests. TS-V313: TS under wet conditions in the third cycle of the V313 treatment; TS-ASTM: largest TS in the sixth cycle of the ASTM-6c treatment.

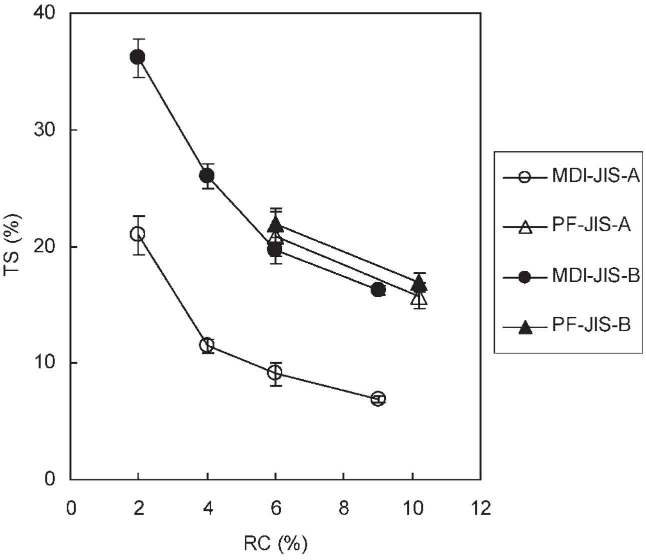

Figure 7.—Effect of resin content (RC) on thickness swelling (TS) under wet conditions of the JIS-A and JIS-B treatments.

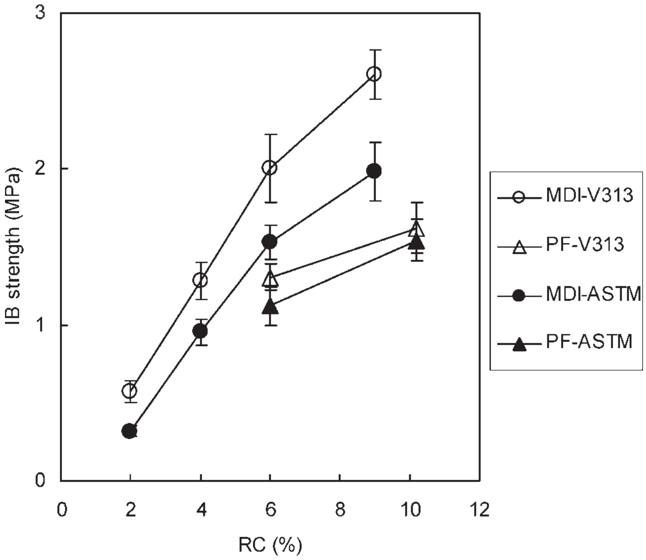

Figure 9.—Effect of resin content (RC) on the internal bond (IB) strength after the V313 and ASTM-6c treatments.

phenomenon was the effect of the boiling treatment in JIS-B. The effect of hot water temperature on the TS of the MDI resin bonds in particleboard or fiberboard will be a topic for future research.

IB strength is one of the most important indexes for the bond quality of mat-formed panel products. Figure 9 shows the effect of RC on the IB strength of the MDI and PF boards after the V313 and ASTM-6c treatments. The IB strength increased dramatically with increasing RC for MDI-bonded boards and slightly increased for PF-bonded boards. The IB strength after the ASTM-6c treatment was slightly smaller than that after the V313 treatment for both resin types at each RC level. The equivalence was determined in the same manner as before. The IB strength of a PF 6 percent board after V313 treatment was comparable to that of an MDI 4 percent board. After the ASTM-6c treatment, the IB strength of a PF 6 percent board corresponded to that of an MDI 4.6 percent board. The estimated IB equivalent of a PF 10.2 percent was MDI of 4.9 and 6.1 percent after the V313 and ASTM-6c treatments, respectively.

Figure 10 shows the relationships between IB strength and RC for the two resin types in the two water immersion treatments. Similar to what occurred in the V313 and ASTM-6c treatments, the IB strength increased with increasing values of RC, and the IB strength of MDI boards was larger than that of PF boards. The equivalence of the two resin types was deduced from this graph in the same manner. The IB values after JIS-A and JIS-B for MDI boards were close to each other, even though the TS values were quite different as shown in Figure 7. This means that the boiling water in JIS-B caused swelling in the MDI-

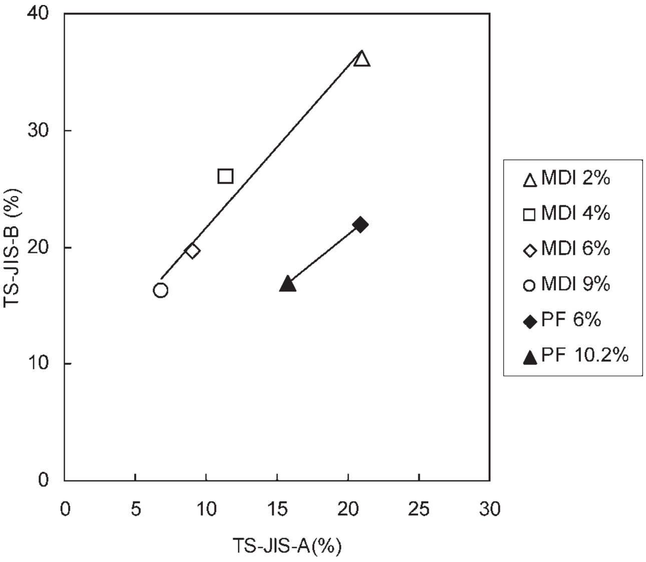

Figure 8.—Relationship between thickness swelling (TS) due to the JIS-A treatment and JIS-B treatment under wet conditions.

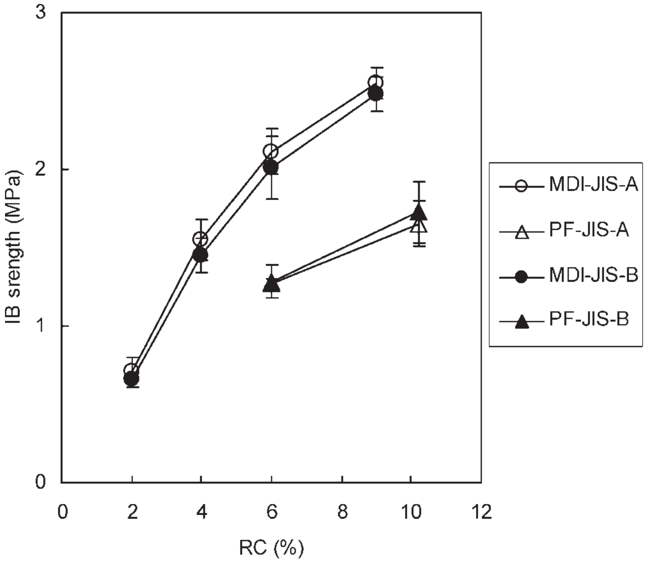

Figure 10.—Effect of resin content (RC) on the internal bond (IB) strength after the JIS-A and JIS-B treatments.

Table 1.—Equivalent MDI resin content compared with PF resin.

	RC of PF resin (%)	Control (%)[a]	MDI RC for each aging treatment (%) JIS-A	JIS-B	V313	ASTM-6c
IB	6	3.3	3.3	3.6	4.1	4.6
	10.2	4.5	4.4	5.0	4.9	6.1
TS	6	—	2.0	5.3	3.8	3.6
	10.2	—	3.1	8.4	5.5	5.0

[a] Control = without aging treatment.

bonded board but did not decrease the IB strength compared with the IB value after the JIS-A treatment with 70°C water immersion. No clear conclusion could be made as to what occurred in the binder and element in the MDI-bonded board during the treatments. The effect of water temperature is one of the key items to be discussed in later research because the only difference between the two methods was the treatment temperature. It might be said that the JIS-A and JIS-B treatments are used to degrade panel products as a pretreatment before bending tests, whereas the V313 and ASTM-6c methods were developed for moisture resistance tests. The TS strongly affects the results in the bending test so that the JIS-A and JIS-B treatments can differentiate the various types of the panel even if the sample specimens have the same IB strength.

Equivalence of bond quality

The equivalence was evaluated on the basis of TS and IB strength. Table 1 shows the estimated MDI RC corresponding to the RC of PF-bonded boards for each RC and each variable. Control is the IB strength without aging treatment. All values in this table (except for control) were introduced in Figures 5, 7, 9, and 10.

Approximately half of the MDI resin dosage was required to obtain the comparable IB strength in a PF board. According to the severity of the aging treatment from JIS-A to ASTM-6c, larger amounts of MDI seemed to be necessary to obtain the same IB strength. However, a different trend was evident in the results for TS. The superiority of MDI resin was not as clear for TS after the JIS-B test, which included the boiling treatment. Thus, the IB may be a better indicator than TS for the comparison, and cyclic aging treatments such as V313 and ASTM-6c are appropriate test methods for determining the equivalency. The ratio of PF RC to the estimated MDI RC for both the V313 and ASTM-6c treatments ranged from 1.30 to 2.08, and the simple mean of these eight values was 1.71. This value is very close to the ratio of 1.7, which was used as the bond effectiveness of the two resins.

Conclusions

Under dry conditions, the MOR of MDI-bonded boards with 9 percent RC was about 45 MPa, and of boards with 2 percent RC was greater than 30 MPa. These values are far larger than the bending strength of commercial particleboard. The bending properties of PF-bonded boards were relatively smaller than those of MDI boards, although both were sufficient to meet the JIS requirements for particleboard (JISC 1994).

The IB strengths of MDI-bonded boards after aging treatments were significantly higher than those of PF-bonded boards. It was possible to achieve equivalent IB strengths using a considerably lower dosage of MDI resin compared with that required in PF boards. MDI boards were also more stable than PF boards for the same RC level. The superiority of the MDI resin as a binder was confirmed via various aging treatments using TS and IB as parameters, except that the advantage of the MDI resin was not clear for TS after the JIS-B treatment, which included the boiling procedure.

Acknowledgment

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