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ORIGINAL ARTICLE

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Effects of sealed pressing and ozonization on properties of styrene-butadiene rubber and polyethylene glycol adhesive bonded board produced by two-stage pressing

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Abstract Boards were produced by using SP adhesive, which contains styrene-butadiene rubber and polyethylene glycol as major constituents. The use of polyethylene in place of clay, which is also a generally used constituent of SP adhesive, was confirmed to improve board properties. In general, the properties of boards are poorer when produced by two-stage pressing, in which mats are first processed by temporary adhesion and then processed into boards by permanent adhesion; however, the properties of boards produced by two-stage pressing were improved when polyethylene was added to the SP adhesive. In addition, internal bond strength and thickness swelling was greatly improved when boards were produced from ozonized wood and by sealed pressing. Thus, the properties impaired by two-stage pressing were improved by ozonization and sealed pressing.

Key words Board · Styrene-butadiene rubber · Polyethylene · Two-stage pressing · Sealed pressing

Introduction

The properties of boards (particleboard) produced by bonding with SP adhesive, which consists primarily of styrene-butadiene rubber (SBR) and polyethylene glycol (PEG), were reported in our previous paper.[1] The SP adhesive is advantageous particularly because posthardening does not progress at ambient temperature. Because general thermosetting resins gradually harden even when heat is not applied to them, wood raw materials with thermosetting resins need to be hot pressed and processed into boards as soon as the thermosetting resins have been sprayed. When the SP adhesive is used, however, wood raw materials can be left as they are for a long time because the SP adhesive remains plastic. This property allows adhesion in two stages, i.e., temporary adhesion followed by permanent adhesion, which is highly promising for developing wood-based materials for new applications. For instance, after processing wood raw materials to which adhesive has been sprayed into a mat, the mat is preliminarily pressed at a relatively low temperature (around 80°C) to attain a density of about 0.35 g/cm^3 (Fig. 1). This stage causes the raw materials to temporarily adhere, and the mat can then be transported. Subsequently, the mat is placed in a metallic mold to undergo permanent adhesion by final pressing at high temperature (around 180°C) to attain a density of about 0.7 g/cm^3. In this manner, wood-based materials of various shapes (molding materials) can be produced. In the present study, this method is called two-stage pressing, although so far, few wood-based materials have been produced by this method.

As reported in our previous paper,[1] we improved the SP adhesive to enhance internal bond strength by modifying the mix proportion of the constituents. In the present study, we developed adhesives suitable for producing molding materials by further improving the SP adhesive. According to Ehara et al.,[2] boards produced by two-stage pressing were inferior in properties to conventional boards produced by pressing once when bonding with the SP adhesive. This study attempted to find ways to prevent this reduction in properties. To produce molding materials by hot pressing, it is appropriate to use thermoplastic, which plasticizes when hot pressed and hardens when cooled. However, materials produced by bonding with only thermoplastic remain unstable in shape when removed from the hot press because the thermoplastic has softened. Nonetheless, the combined use of SP adhesive, which can harden quickly after hot pressing, and thermoplastic can overcome the problem of shape instability and thus is promising for developing molding materials. Thus, in this study, boards were produced by adding thermoplastic to the SP adhesive.

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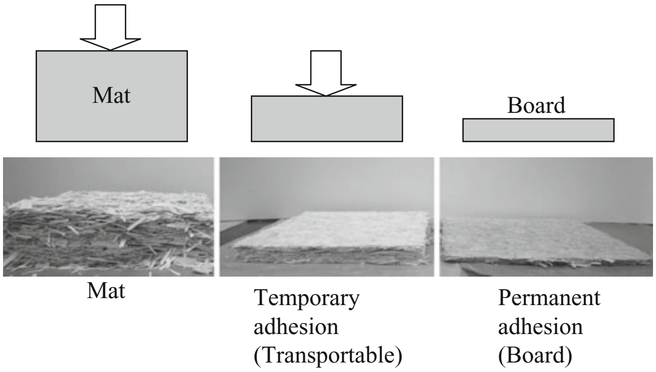

Fig. 1. Schema of mat, temporarily adhered mat, and permanent adhered mat (board) for two-stage pressing

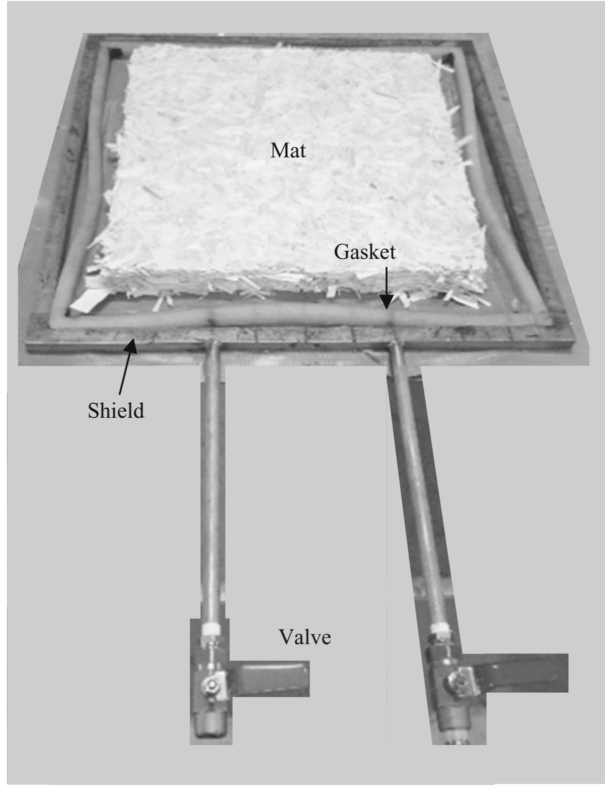

Fig. 2. Shield for sealed pressing for high pressure and high temperature

As shown in our previous paper,[1] the dimensional stability of boards bonded with SP adhesive was low. To enhance dimensional stability, it is important to reinforce internal bond strength by concomitantly using thermoplastic. To improve dimensional stability, it is also important to avoid accumulation of stresses inside the boards during pressing. Wood raw materials plasticize under high temperature and pressure, so if pressed while they are plastic, stress does not accumulate and compressive deformation can be corrected. Accordingly, wood raw materials were processed also by sealed pressing at high temperature and pressure in this study. With sealed pressing, wood raw materials are sealed between shields on the top, bottom, and sides of the press, and high temperature and pressure are then applied (Fig. 2).[3,4]

As reported in our previous paper,[1] ozonization was confirmed to improve the internal bond strength of boards bonded with the SP adhesive. Accordingly, ozonization was also implemented in this study, and the properties of boards produced using ozonized wood raw materials sprayed with SP adhesive, to which thermoplastic had been added, were examined, and the effect of ozonization was evaluated for developing molding materials.

Thus, in this study, the SP adhesive was improved by adding thermoplastic; boards were produced by bonding with the improved SP adhesive, ozonized wood raw materials, and sealed pressing; and the properties of boards were analyzed as basic research for developing molding materials.

Experiment

Improvement of the SP adhesive

In our previous study,[1] the optimal mix proportion of SBR, PEG, and clay, which are the major constituents of the SP adhesive, was examined and determined to be SBR:PEG:clay:water = 50:20:20:10 (weight ratio). With the amount of PEG (PEG-400; Dai-ichi Kogyo Seiyaku) set at 20%, an adhesive was produced using SBR (Nipol LX-430, with solid content of 49%; ZEON) and clay (ST Kaolin Clay; Takehara Kagaku Kogyo). For the tests, a new adhesive was produced adding polyethylene (High-Density Polyethylene abifor 1300/20; Abifor), which is a thermoplastic, to replace clay. The control was produced using the SP adhesive containing clay. The mix proportion of polyethylene or clay was adjusted based on the amount of SBR; i.e., when the proportion of SBR was set at 50, either polyethylene or clay was added in the proportion of 20 or 25 (Table 1). The mix proportion of PEG was fixed at 20 when the proportion of SBR was 50. In this study, four types of adhesives were produced with the mix proportion of either polyethylene or clay being either 20 or 25, respectively, when the amount of SBR was set at 50. These adhesives are called PE 20, PE 25, Clay 20, and Clay 25, respectively; the boards produced by bonding with these

Table 1. Mix proportion (weight ratio) of SBR, PEG, clay, and water

Adhesive	PE 20	PE 25	Clay 20	Clay 25
SBR[a]	50	50	50	50
PEG[b]	20	20	20	20
Polyethylene	20	25	0	0
Clay	0	0	20	25
Water	10	5	10	5
Viscosity (Pa)	10	10	7.5	7.6

[a] Styrene-butadiene rubber
[b] Polyethylene glycol

adhesives were correspondingly called PE 20 board, and so forth. The viscosity of each adhesive was measured.

Production of boards by two-stage pressing

Hinoki (*Chamaecyparis obtusa* Endl.) wood was chipped into small strands using a disk-type knife ring flaker. The strands measured about 25.4 (3.23) mm in length in the axial direction, 7.19 (7.19) mm in width, and 0.45 (0.20) mm in thickness on average (standard deviation in parentheses) with 50 replications. These strands were dried, and the moisture content after drying became approximately 4%. The strands were put in a rotary drum device, where adhesives were applied using a spray gun at a ratio of 15% solid content to the weight of oven-dry strands. After spraying adhesives, the strands were manually formed in a forming box measuring 30 cm × 30 cm to be processed into mats. Employing an ordinary pressing method, the mats were hot pressed at a hot plate temperature of 180°C for 15 min. The moisture content of the mats was set at 8%. This method is called one-stage pressing because pressing is implemented once. The board thickness was set at 1 cm with the target board density of 0.70 g/cm^3.

Boards were also produced by two-stage pressing. Strands were formed into mats in the same manner, which were then hot pressed at a hot plate temperature of 80°C for 20 min to become transportable with a thickness of 2 cm (board density of 0.35 g/cm^3) (temporary adhesion) (see Fig. 1). Subsequently, the mats were further hot pressed at a hot plate temperature of 180°C for 15 min to be processed into boards with a thickness of 1 cm. The final board dimensions were 30 cm × 30 cm × 1 cm with the target board density of 0.7 g/cm^3 (permanent adhesion). Several boards were produced under the conditions described above, one board specimen per condition. The boards produced by one-stage pressing and two-stage pressing are called one-stage board and two-stage board, respectively, and depending on the adhesive used, the board produced by bonding with the PE 25 adhesive by two-stage pressing, for example, is called a two-stage PE 25 board. The moisture content of mats produced by one-stage pressing was 8%; however, that of the mats produced by two-stage pressing became 4%, which was lower than 8% because the mats had already been hot pressed by the temporary adhesion in the first pressing process.

Production of boards by sealed pressing

Boards were also produced by sealed pressing at a hot plate temperature of 180°C for 15 min. The valves were shut for the first 10 min to maintain high pressure, and subsequently the valves were opened for another 5 min to release vapor. In contrast to sealed pressing, the conventional unsealed pressing is called open pressing. The predetermined amount of water was sprayed onto strands that were processed into mats with the moisture content being adjusted to three levels, i.e., approximately 8%, 12%, and 16%. Then, the mats were further processed into boards by bonding with the PE 25 adhesive by one-stage sealed pressing and one-stage open pressing. The board dimensions were planned as 30 cm × 30 cm × 1 cm with the target board density of 0.70 g/cm^3. Several boards were produced under the conditions described above, one board specimen per condition. Boards produced by open pressing are called open board whereas those produced by sealed pressing are called sealed board.

Production of boards from ozonized wood raw materials

Following our previous work,[1] boards were also produced from wood raw materials that had been treated in advance by ozonization at an ozone charge rate of 0.5%. The predetermined amount of water was sprayed onto strands that were processed into mats with the moisture content being adjusted to approximately 8%, 12%, or 16%. Then, boards were produced by bonding with the PE 25 adhesive by one-stage sealed pressing and one-stage open pressing. The board dimensions were 30 cm × 30 cm × 1 cm with the target board density of 0.70 g/cm^3. Several boards were produced under the conditions described above, one board specimen per condition. Boards produced from ozonized raw materials are called Oz-board, whereas those produced from untreated (no ozonized) raw materials are called U-board. The names of boards are further differentiated by the pressing method used, e.g., Oz-boards produced by sealed pressing are called sealed Oz-board.

Production of two-stage board from ozonized wood raw materials by sealed pressing

Two-stage boards were produced form ozonized wood raw materials by sealed pressing, and the effect of this method on board properties was verified. The dimensions of the boards were 30 cm × 30 cm × 1 cm with the target board density of 0.70 g/cm^3. Several boards were produced under the conditions described above, one board specimen per condition.

Property tests

The boards were conditioned in a temperature- and humidity-controlled chamber at 20°C and relative humidity of 65%, and the respective tests were conducted after the mass of the boards became constant. According to JIS A 5908 2003,[5] the modulus of rupture (MOR), internal bond strength (IB), and thickness swelling (TS) were measured, with the number of replications of 5, 8, and 7, respectively. Before conducting IB tests, the density profile (DP) of the board specimens was measured using a DP analyzer (DA-X; GreCon). In consideration of the variance of the board density of the specimens, density distribution was plotted based on the relative density, which was obtained by dividing the density in the thickness direction by the mean board density of the specimens.

Results and discussion

Properties of boards produced by two-stage pressing

Modulus of rupture

Figure 3 shows the results of MOR measurements of the boards produced by bonding with four types of adhesives by one-stage pressing and two-stage pressing. The MOR of the one-stage boards was higher than that of the two-stage boards for all the adhesives bonded. Figure 4 shows the values of the density profile, which affects the MOR. As no DP variance caused by the difference in the types of adhesives bonded was observed, the DPs of the one-stage and two-stage boards bonding with PE 20 are plotted in Fig. 4 as examples. The surface layer density of the one-stage board is higher than that of the two-stage board, which is one of the reasons why the MOR of the former was higher.

The retention was almost invariably around 80% for all boards irrespective of the types of adhesives bonded, showing that the adhesives scarcely affected the retention. Specifically, the addition of polyethylene did not lead to an increase in the retention. Nevertheless, the MOR of the two-stage PE 20 board and the two-stage PE 25 board was around 33 MPa, whereas that of the one-stage Clay 20 board and the one-stage Clay 25 board was 30.5 MPa and 34.8 MPa, respectively. This result shows that the addition of polyethylene raised the relative values of MOR, and the MOR of the two-stage PE 20 board and the two-stage PE 25 board became almost the same as that of the one-stage Clay 20 board and the one-stage Clay 25 board. Accordingly, the addition of polyethylene served to increase the MOR.

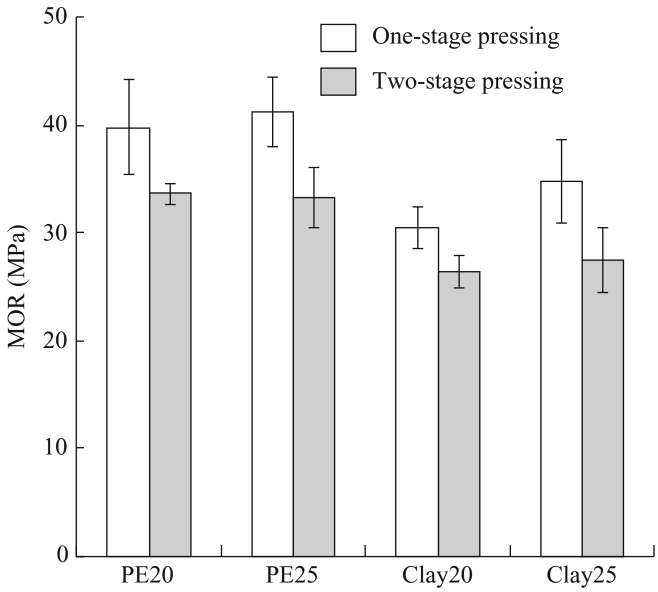

Fig. 3. Effects of pressing method and adhesive on modulus of rupture (*MOR*). *Vertical bars* denote standard deviations. PE 20, PE 25, Clay 20, and Clay 25 are shown in Table 1

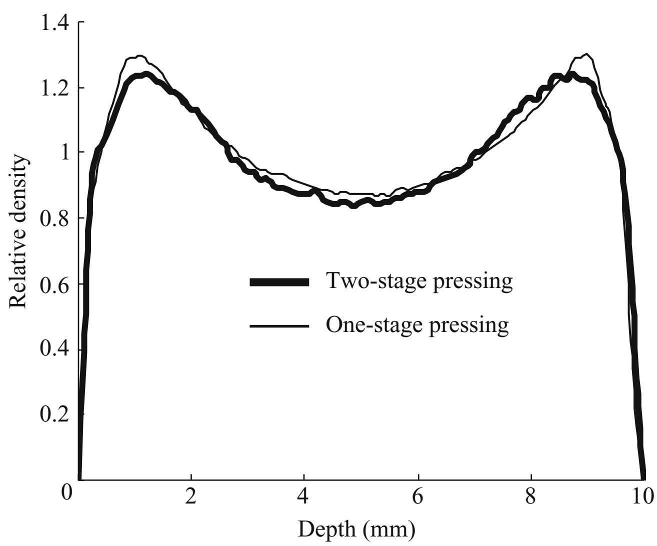

Fig. 4. Effects of pressing method on density profile. Relative density is the density at each depth divided by the mean board density. Boards were produced by one-stage pressing (*fine line*) or two-stage pressing (*heavy line*)

Internal bond strength

Figure 5 shows the results of IB measurements of the boards produced by bonding with the four types of adhesives by one-stage pressing and two-stage pressing. Although MOR retention was around 80%, IB greatly decreased by two-stage pressing. The IB retention of the two-stage Clay 25 board was around 53% whereas that of boards produced by bonding with the other three adhesives was around 62%. As with MOR, the addition of polyethylene was not shown to be effective in increasing the retention. The IB of the

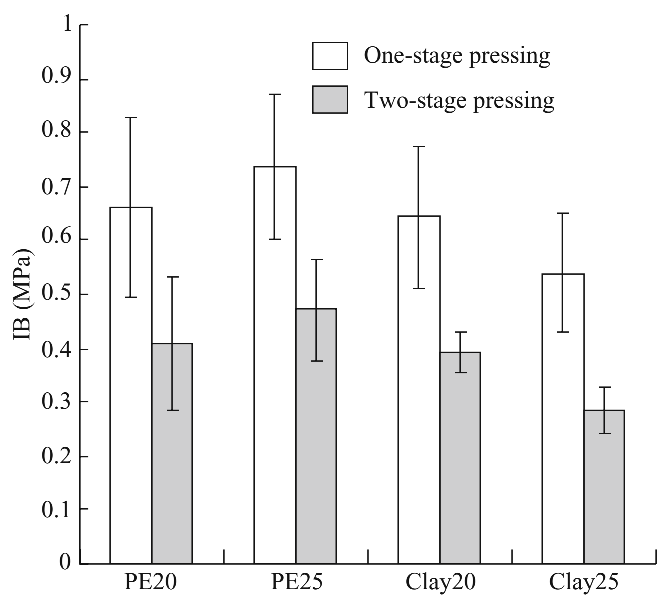

Fig. 5. Effects of pressing method and adhesive on internal bond strength (*IB*). *Vertical bars* denote standard deviations. PE 20, PE 25, Clay 20, and Clay 25 are shown in Table 1

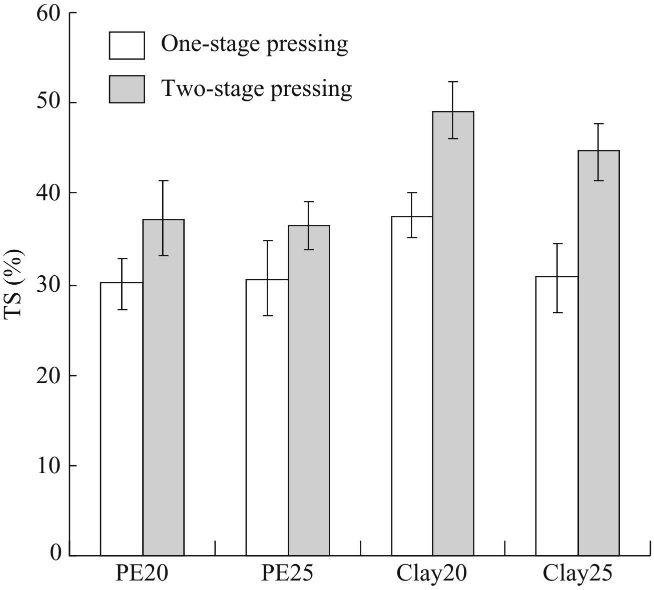

Fig. 6. Effects of pressing method and adhesive on thickness swelling (*TS*). *Vertical bars* denote standard deviations. PE 20, PE 25, Clay 20, and Clay 25 are shown in Table 1

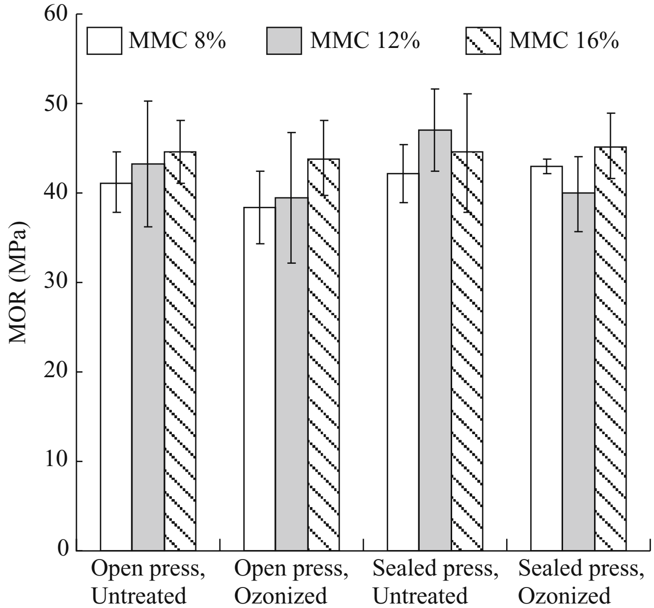

Fig. 7. Effects of pressing method, treatment by ozonization, and mat moisture content (*MMC*) on modulus of rupture (*MOR*). *Vertical bars* denote standard deviations

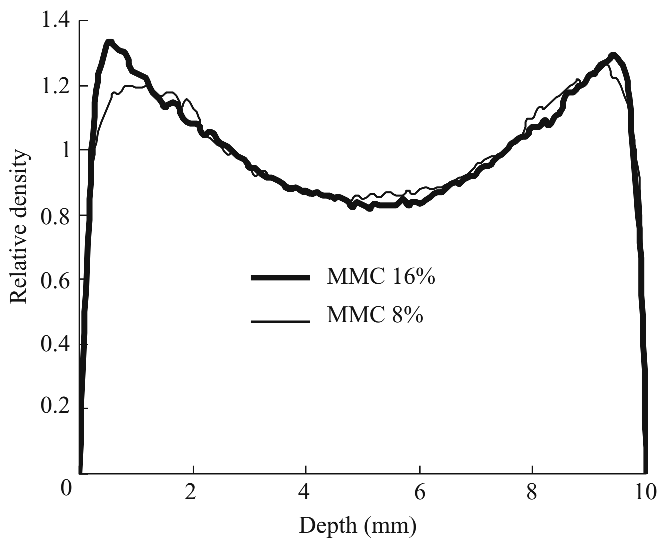

Fig. 8. Effects of mat moisture content (*MMC*) on density profile. Relative density is the density at each depth divided by the mean board density. Boards were made from untreated wood by open pressing

two-stage PE 20 board and the two-stage PE 25 board was 0.41 MPa and 0.47 MPa, respectively, whereas that of the one-stage Clay 20 board and the one-stage Clay 25 board was 0.64 MPa and 0.54 MPa, respectively. In contrast to the results of MOR, the relative values of IB did not increase when polyethylene was added, and the IB of the two-stage PE 20 board and the two-stage PE 25 board became lower than that of the one-stage Clay 20 board and the one-stage Clay 25 board.

Thickness swelling

Figure 6 shows the results of TS measurements of the boards produced by bonding with the four types of adhesives by one-stage pressing and two-stage pressing. The TS of the two-stage boards was higher than that of the one-stage boards, with the TS of the PE 20 board and the PE 25 board increasing by approximately 20%, the Clay 20 board by approximately 30%, and the Clay 25 board by approximately 45%. In contrast to MOR and IB, the addition of polyethylene was confirmed to be effective to reduce TS.

Thus, the properties of PE 20 board and PE 25 board mainly were higher than those of Clay 20 board and Clay 25 board. Because there is no significant difference between PE 20 board and PE 25 board, boards were produced by bonding with PE 25 in this study hereafter.

Effect of sealed pressing and ozonization on the board properties

Modulus of rupture

Figure 7 shows how the pressing method, treatment by ozonization, and mat moisture content are correlated to the MOR of the produced boards. The surface layer density generally increases with the increase of the mat moisture content, and the MOR increases accordingly.[3] This trend of DP was also confirmed in this study. Investigation of the change of surface layer density by measuring the DP showed that the surface layer density of boards with a mat moisture content of 12% and 16% was higher than that of boards with a mat moisture content of 8%, notwithstanding the treatment by ozonization and pressing methods (Fig. 8). Nevertheless, the MOR remained almost at the same level (about 40 MPa) irrespective of the increase in the mat moisture content (high surface layer density). The high MOR values are assumed to be the result of the length of strands made from wood raw materials, which is long in the fiber direction; however, the MOR values of the nonoriented boards increased to around 40 MPa at maximum generally,[6] then showed no further increase. The MOR did not increase in line with the increase in the mat moisture content resulting from the effect of the shape of strands, which was long in the fiber direction. The length of strands is 25.4 mm in this study, which is enough for attaining high MOR. Even if the length would be long, the MOR will not increase. In spite of the fact that the surface layer density was low when the mat moisture content was low, the effects of the shape of strands (long in the fiber direction) surpassed the influence of the increase in the mat moisture content (high surface layer density), which usually increases the MOR.

Internal bond strength

Figure 9 shows the correlation between the mat moisture content and IB. The IB of the open U-boards was lower than that of other boards irrespective of the mat moisture content. A comparison between the open U-boards and the open Oz-boards showed that the IB of the open Oz-boards was higher in general, thereby proving the effect of ozonization. However, for the boards produced by sealed pressing, no effect of ozonization was confirmed. On the other hand, the IB of the sealed U-boards and the sealed Oz-boards decreased when the mat moisture content was 16%, but that of the open U-boards and the open Oz-boards did not vary greatly regardless of the increase in mat moisture content. In general, the adhesion between wood materials is enhanced in line with the increase in moisture content because the plasticization of wood materials progresses.[7] As this increases the effective adhesion area, the IB was also expected to increase; however, no increase of the IB values was observed. This result is considered to be due to the high mat moisture content, which hindered the boards from drying sufficiently in the sealed press and disturbed the increase of bonding strength, because sufficient water evaporation is required for the SP adhesive to attain high bonding strength.[1] When the mat moisture content was 8%, the IB of the sealed U-board and the sealed Oz-board was higher than that of the open U-board and the open Oz-board, thereby proving the effectiveness of sealed pressing when the mat moisture content was low. We had expected that the IB of the boards produced by sealed pressing, which fixes the compressive deformation of wood materials, would be high, and indeed the test results showed mainly that the IB of the sealed boards was higher than that of the open boards at mat moisture content of 8% and 12%. It is considered that the IB of the boards produced by sealed pressing with high mat moisture content of 16% would have become higher if sufficient water could have evaporated.

Thickness swelling

A comparison between the open boards and sealed boards showed that the TS of the latter was lower than that of the former (Fig. 10). In particular, sealed pressing effectively lowered the TS when the mat moisture content was as low as 8%. Meanwhile, a comparison between the U-boards and the Oz-boards showed that ozonization took effect when the moisture content was high, resulting in the low TS of the Oz-boards. Sealed pressing, ozonization, and high moisture content effectively served to lower the TS. The TS of the open U-board with a mat moisture content of 8% was

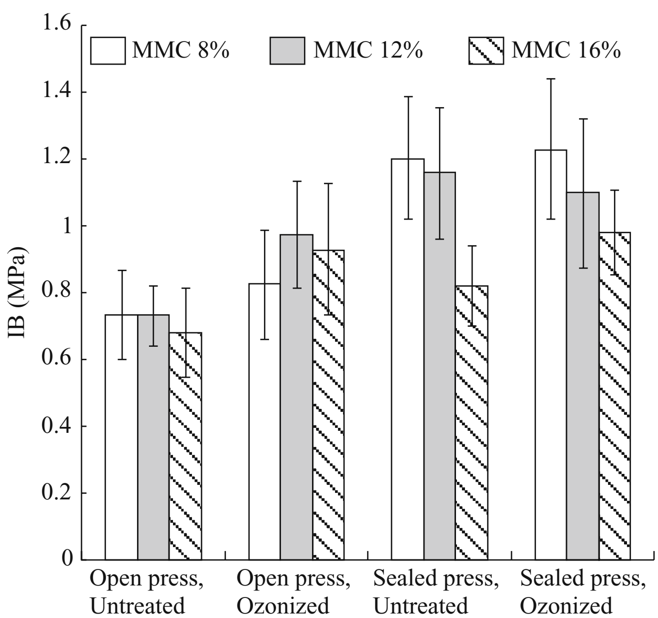

Fig. 9. Effects of pressing method, treatment by ozonization, and mat moisture content (*MMC*) on internal bond strength (*IB*). *Vertical bars* denote standard deviations

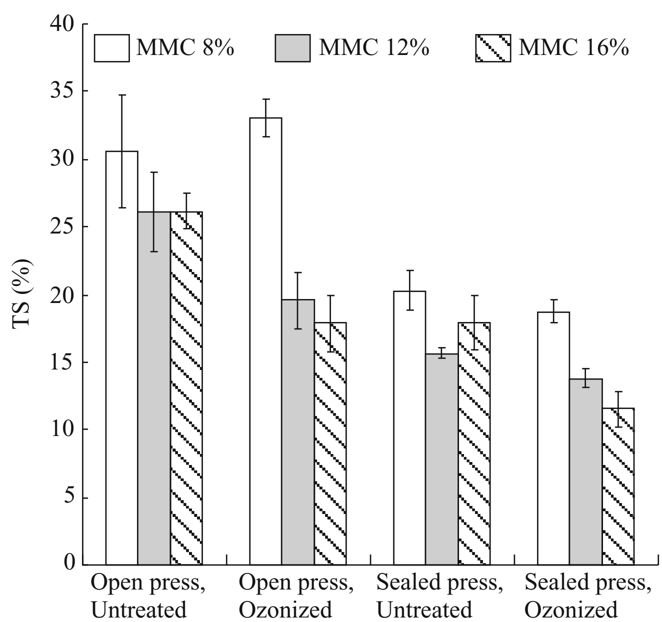

Fig. 10. Effects of pressing method, treatment by ozonization, and mat moisture content (*MMC*) on thickness swelling (*TS*). *Vertical bars* denote standard deviations

Table 2. Effects of ozonization and sealed pressing on properties of the boards by two-stage pressing

Production conditions		MOR	IB	TS
Open	Untreated	33.2 (2.88)	0.47 (0.09)	36.4 (2.55)
Sealed	Untreated	36.3 (4.09)	0.69 (0.06)	20.9 (1.52)
Open	Ozonized	32.7 (3.50)	0.63 (0.10)	30.9 (2.23)
Sealed	Ozonized	35.9 (5.29)	0.78 (0.07)	19.0 (0.78)

Parentheses indicate standard deviations

30.6%, whereas that of the sealed Oz-board with a mat moisture content of 16% was 11.5%. Actually a decrease of about 38%, this is considered to be caused by the effects of plasticization of wood materials by high mat moisture content, an increase of bonding strength by ozonization, and fixation of compressive deformation by sealed pressing.

Effects of ozonization and sealed pressing on the properties of the two-stage boards

The impaired properties of the two-stage boards were assumed to be improved by ozonization and sealed pressing. Table 2 shows properties of the two-stage boards from ozonized wood raw materials by sealed pressing. The MOR indicated almost the same value irrespective of the production conditions; IB and TS were improved. The IB was improved by the sealed pressing, and further improved by both sealed pressing and ozonization; TS was improved greatly by the sealed pressing. Thus, the properties impaired by the two-stage pressing were improved by sealed pressing and ozonization.

Conclusions

MOR, IB, and TS were successfully improved by adding polyethylene instead of clay, which is normally used for the SP adhesive. The properties of MOR, IB, and TS decreased when boards were produced by two-stage pressing; however, when polyethylene was added, the properties of the two-stage boards became competitive with those of the one-stage boards using clay. Although the TS could be improved by adding polyethylene, it was still as high as 30.6%. Nonetheless, the TS could be greatly decreased to 11.5% when boards were produced using ozonized wood materials with a mat moisture content of 16% by sealed pressing. In addition, ozonization and sealed pressing increased the IB. Thus, the properties impaired by two-stage pressing were improved by ozonization and sealed pressing.

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References

1. Korai H, Kobayashi M, Sumida A, Yasuda O, Osada T (2009) Effects of ozonization on properties of wood-based board bonded with styrene-butadiene rubber and polyethylene glycol adhesive (in Japanese). Mokuzai Gakkaishi 55:285–292
2. Ehara M, Suzuki S, Machiyashiki T (2000) Evaluation of wood-based board bonded with PEG adhesive (in Japanese). Nihon Mokuzai Gakkai Taikai Kenkyu Happyo Yoshishu 50:280
3. Korai H, Uemura K, Easashi T, Suzuki M (1999) Dimensional stability and strength properties of particleboard produced by a closed-press system. J Wood Sci 45:402–410
4. Inoue M, Kadokawa N, Nishio J, Norimoto M (1993) Permanent fixation of compressive deformation by hygro-thermal treatment using moisture in wood (in Japanese). Wood Res Tech Notes 29:54–61
5. Japanese Industrial Standard Particleboards. JIS A 5908:2003
6. Kajita H, Mukudai J (1981) Particleboard with oriented particles (in Japanese). J Soc Materials Sci Jpn 30:736–741
7. Kawai S, Suda H, Nakaji M, Sasaki H (1986) Production technology for low-density particleboard II. Effects of particle moisture content and resin content on board properties (in Japanese). Mokuzai Gakkaishi 32:876–882