Properties of low-formaldehyde-emission particleboard made from recycled wood-waste chips sprayed with PMDI/PF resin

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Abstract

The objective of this study was to manufacture low-formaldehyde-emission particleboard from recycled wood-waste chips using polymeric 4,4'-methylenediphenyl isocyanate (PMDI) and phenol-formaldehyde (PF) resins for use in indoor environments. The influence of PMDI/PF ratios in particles on the formaldehyde emission and its mechanical properties were investigated. The experimental results showed that the formaldehyde emission released decreased linearly with increasing PMDI/PF particle ratio. The relationship could be represented by a linear regression formula. Formaldehyde emission was below 0.3 mg/L when the weight percentage of PMDI/particles was up to 70%. The formaldehyde emission from melamine-formaldehyde (MF) resin-impregnated paper-overlaid particleboard was 17% lower than that for particleboard. Moreover, the bending strength, internal bonding strength and screw holding strength increased with increasing PMDI/PF particle ratio. However, the percentage thickness swelling of the particleboard decreased with increasing PMDI/PF particle ratio. In addition, there were significant positive relationships between the ultrasonic velocity and the bending strength, internal bonding strength and screw holding strength of the particleboard, which allowed evaluation of the properties of the particleboard using ultrasonic velocity.

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1. Introduction

In recent years, the recycling systems and reuse techniques of wood wastes in Taiwan have become an important part of sustainable utilization of resources. The Taiwanese government has already issued regulations regarding to recycling and reusing wood-based wastes such as flooring, ceiling, and decorations from construction and demolition sites [1]. According to the end uses, wood wastes could be sorted, processed and provided as raw materials for manufacture of pulp, fiberboard and particleboard. In these reuse products, particleboard has found

typical applications as flooring, wall and ceiling panels, office dividers, bulletin boards, furniture, cabinets, counter tops, and desk tops [2], and it seems that the manufacture of particleboard from recycled wood-based wastes is the most common way to reuse such waste materials.

During the process of using recycled wood wastes for further reutilization, some problems should be taken into consideration, such as formaldehyde can be released from particleboard used as an interior building product due to incompletely reacted urea-formaldehyde (UF), urea-melamine-formaldehyde (UMF), melamine-formaldehyde (MF) or phenol-formaldehyde (PF) resins used in particleboard. Baumann et al. [3] also indicated that emissions from building and furnishing materials, which are frequently constructed from particleboard and medium density fiberboard, are a potentially important contributor of indoor volatile organic compounds (VOCs). As a result, the indoor air quality can deteriorate and a serious health risk

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may arise, especially in modern homes and offices, which are frequently more airtight than older structures. This increases the significance of the role of formaldehyde in inducing illness via immune mechanisms, infectious processes, and direct toxicity. This phenomenon has become known as sick building syndrome (SBS), whereby the occupants of certain affected buildings repeatedly describe a complex range of vague and often subjective health complaints. To overcome these problems related to air quality and health in the indoor environment, the effects of press conditions (press temperature and time), mat moisture content (MC), lower molecular weight UF resins, and additive of formaldehyde scavenger on formaldehyde emissions or using low-formaldehyde and non-formaldehyde resins for the manufacture of various wood-based panels have been extensively studied in the past [4–11].

Moreover, a series of studies has been conducted to investigate the use of non-destructive evaluation (NDE) technologies to aid in the quality assessment of wood-based products [12–14]. These studies yielded similar results and established relationships between the mechanical strength and dynamic properties.

The purpose of this study was to investigate the quantity of formaldehyde released, density, bending properties, internal bonding strength, and thickness swelling of particleboard made from recycled wood chips of different species, which were sprayed with polymeric 4,4'-methylenediphenyl isocyanate (PMDI) for the core layer and with PF resin for the face and back layers. Furthermore, the ultrasonic properties of the manufactured particleboard were also evaluated.

2. Materials and methods

2.1. Materials

Chips from mixed hardwood species including oak (*Quercus* spp.) and lauan (*Shorea* spp.) were prepared from recycled construction and demolition wastes, which were collected and then classified by Jen-Chun Enterprises in Feng-Yuan, Taiwan.

The adhesives used were a water-soluble PF resin (76–78% solid content and pH 8–9; Tai-Lin Resin, Taipei, Taiwan) and a water-soluble PMDI resin (100% solid content; Mu-Chau Resin, Tainan, Taiwan). MF resin-impregnated paper was overlaid on the face and back layers of the particleboard.

2.2. Chip production process

Chips were divided into coarse chips, which passed through a four-mesh and were retained by an eight-mesh screen, and fine chips, which passed through an eight-mesh and were retained by a 20-mesh screen. The ratio of coarse/fine chips used for particleboard was 2:1. The chips were oven-dried to 2–4% MC at a temperature of 338 ± 2 K. They were then sprayed with a water-soluble PMDI or PF

Table 1
Particleboards fabricated for this study

Sample group	Chip content (%)	
	PMDI	PF
A	100	0
B	80	20
C	70	30
D	50	50
E	30	70
F	20	80
G	0	100
MD	50	50
MC	70	30
MA	100	0

Sample groups A–G: particleboards. Sample groups MD, MC and MA: MF resin-impregnated paper-overlaid particleboards. PMDI was used for the core layer of the particleboard. PMDI resin absorption was 4% of the oven-dried weight of the chips. PF was used for the face and back layers of the particleboard. PF resin absorption was 6% of the oven-dried weight of the chips.

resin according to the experimental program shown in Table 1.

Chips were then placed into a frame of 500 mm × 500 mm. Three-layer particleboard was formed using coarse and fine chips at a ratio of 2:1. A conventional hot press was used for fabrication of the particleboard and the temperature, pressure and pressing time were 453 K, 2.9 MPa and 5 min, respectively. The nominal dimensions of particleboard were 500 mm × 500 mm × 12 mm (thick), with three layers. The thickness of the particleboard was controlled by stop bars and the target density was 0.8 g/cm³. Five samples of each PMDI/PF ratio (weight of chips sprayed with PMDI/weight of chips sprayed with PF) (Table 1) were fabricated in this study. Furthermore, MF resin-impregnated paper-overlaid particleboards with three different PMDI/PF ratios (Table 1) were also manufactured for our experiments.

2.3. Testing methods

The particleboards produced for this study were tested in accordance with the Chinese National Standard (CNS 2215), which involves various tests [15].

2.3.1. Test for the quantity of formaldehyde released

(1) Acquisition of formaldehyde: A 120-mm-diameter, 60-mm-high crystallizing dish containing 300 mL of distilled water was placed in the bottom of a 240-mm-diameter desiccator with a capacity of 10 ± 1 L. Nine sheets of test pieces (with dimensions of 12 mm thick × 50 mm wide × 150 mm long) were clamped in a metal support, with no contact between specimens and allowed to stand in the crystallizing dish at 293 K for 24 h. Formaldehyde released from the specimens was

absorbed by the distilled water, which was subsequently used as the sample solution.

- (2) Quantification of formaldehyde concentration: The formaldehyde concentration in the sample solution was determined using acetylacetone-ammonium acetate solution and the acetylacetone method, with colorimetric detection at 415 nm.

Prior to testing, the particleboards were conditioned at 293 K and 65% relative humidity, and full panels of each particleboard were prepared for ultrasonic wave tests. Test points on opposite sides of the particleboard were investigated at intervals of 50 mm. An ultrasonic wave instrument (Pundit, 54 kHz; CNS Electronics) was used to measure the time required for the transmission of an ultrasonic pulse through the specimen. The ultrasonic velocity was then calculated from the length of the specimen divided by the propagation time.

After the ultrasonic investigation, the particleboards tested were cut into specimens for static bending and internal bond strength (IB) tests according to CNS 2215 [15], and their positions relative to the full particleboard were recorded. These static bending specimens were first used to measure the ultrasonic velocity (VI) through the face direction using a Pundit ultrasonic apparatus at a frequency of 54 kHz. Then transit times for the ultrasonic pulse were measured at a frequency of 500 kHz through the thickness of each specimen. The velocity was then calculated using the following formula:

$$VI(V_t)(\text{m/s}) = \frac{L}{t}, \quad (1)$$

where VI is the ultrasonic velocity in the face direction of the particleboard, V_t is the ultrasonic velocity through the thickness of the particleboard, L is the length, and t is the transit time.

2.3.2. Density and moisture content

Fabricated particleboards were conditioned at ambient humidity for 1 week and then tested for density and MC according to the following formulae:

$$\text{Air-dried density (g/cm}^3\text{)} = \frac{W_a}{V_a}, \quad (2)$$

$$\text{Moisture content (\%)} = \frac{W_a - W_0}{W_0} \times 100, \quad (3)$$

where W_a is the air-dried weight, V_a is the air-dried volume, and W_0 is the oven-dried weight of the particleboard.

2.3.3. Static bending test

After the ultrasonic wave tests, bending specimens of 50 mm wide $\times$ 230 mm long were cut from each particleboard. A concentrated bending load was applied at the center with a span of 15 times the thickness of the specimen. The bending modulus of elasticity (MOE) and modulus of rupture (MOR) were calculated from load–

deflection curves according to the following formulae:

$$\text{MOR} = \frac{3P_b L}{2bh^2}, \quad (4)$$

$$\text{MOE} = \frac{P_{bp} L^3}{4bh^3 Y_p}, \quad (5)$$

where MOR is the static bending strength (modulus of rupture) (MPa), MOE is the static bending modulus of elasticity (MPa), P_b is the maximum load (N), P_{bp} is the load at the proportional limit (N), Y_p is the deflection corresponding to P_{bp} (mm), b is the width of the specimen (mm), h is the thickness of the specimen (mm), and L is the span (mm).

2.3.4. Internal bond strength

The tensile strength perpendicular to the surface was determined using three conditioned specimens of 50 mm $\times$ 50 mm from each particleboard according to CNS 2215 [15]. The rupture load (P_s) was determined and internal bond strength was calculated using the following formula:

$$\text{IB} = \frac{P_s}{bl}, \quad (6)$$

where IB is the internal bond strength (MPa), P_s is the rupture load, and l is the length of the specimen. It should be noted that the ultrasonic velocity of IB specimens was measured as described above before the IB tests.

2.3.5. Thickness swelling

Specimens with dimensions of 50 mm $\times$ 50 mm were prepared for evaluation of the thickness swelling according to CNS 2215 [15]. The thickness at the middle of the test specimen was measured with a micrometer. Then the test specimens were placed into water in parallel for 30 mm and soaked for 2 and 24 h before further measurement of the thickness. The thickness-swelling rate (TS) was determined from the following formula:

$$\text{TS}_2 = \frac{(t_2 - t_0)}{t_0} \times 100, \quad (7)$$

$$\text{TS}_{24} = \frac{(t_{24} - t_0)}{t_0} \times 100, \quad (8)$$

where TS is the thickness swelling rate (%), t_0 , t_2 and t_{24} are the thickness at the middle of the test specimen before soaking, and soaking in water for 2 and 24 h, respectively.

3. Results and discussion

3.1. Quantity of formaldehyde released

In this study, chips of mixed wood species were sprayed with a water-soluble PMDI or PF resin according to the experimental program shown in Table 1. It was found that the quantity of formaldehyde released decreased linearly

Table 2

Quantity of formaldehyde released from the particleboards tested and reduction for MF resin-impregnated paper-overlaid particleboard

Sample	Formaldehyde released (mg/L)						
	A	B	C	D	E	F	G
Particleboard	0.08±0.02	0.21±0.03	0.29±0.02	0.49±0.06	0.62±0.04	0.73±0.06	0.86±0.03
MF resin-impregnated paper overlaid particleboard	0.06±0.03	0.17±0.05	0.24±0.03	0.41±0.04	0.53±0.06	0.61±0.03	0.71±0.05
Reduction rate (%)	16.81	17.10	16.38	17.78	14.52	16.60	17.44

Table 3

Correlation among the quantity of released formaldehyde (FR), ultrasonic wave velocity (V), modulus of rupture (MOR), modulus of elasticity (MOE), internal bonding strength (IB) and screw holding strength (SH) analyzed by linear regression ($Y = AX + B$) formulae for tested particleboard

Sample	$Y = AX + B$					
	Y	X	A	B	r^2	F
Particleboard	FR	PMDI/PF	0.0081	0.06	0.99	142**
	MOR	VI	0.02	−31.88	0.73	132**
	MOE	VI	1.87	−1751.40	0.78	172**
	IB	V_t	0.001	−0.20	0.71	116**
	SH	V_t	0.274	−170.52	0.95	37**
MF resin-impregnated paper-overlaid particleboard	FR	PMDI/PF	0.0066	0.06	0.99	102**
	MOR	VI	0.04	−77.3	0.71	79**
	MOE	VI	0.03	−52.45	0.87	206**
	IB	V_t	0.007	0.033	0.87	194**

FR, quantity of formaldehyde released (mg/L); PMDI/PF ratio (%), ratio (%) of chip weight sprayed with PMDI to chip weight sprayed with PF in the particleboard; MOR, modulus of rupture; MOE, modulus of elasticity; IB, internal bonding strength; SH, screw holding strength; VI, ultrasonic velocity propagated through the face direction of the particleboard; V_t , ultrasonic velocity propagated through the thickness of the particleboard.

**Significant difference at the 1% level ($P < 0.01$).

with increasing PMDI/PF ratio (weight of chips sprayed with PMDI/weight of chips sprayed with PF) (Table 2).

Linear regression analysis showed a high degree of correlation ($r^2 = 0.99$) between the quantity of formaldehyde released and the PMDI ratio for the particleboards tested.

The quantity of formaldehyde released from various PMDI/PF particleboards is shown in Table 2. For samples in group D, with a PMDI/PF ratio of 50:50, the quantity of formaldehyde released was 0.49 mg/L, which meets the E_0 standard (< 0.50 mg/L) of CNS 2215 [15]. Moreover, the quantity of formaldehyde released was 0.29 mg/L (< 0.30 mg/L) from group C samples with a PMDI/PF ratio of 70:30.

One of the advantages of using PMDI was no formaldehyde emission, hence, the more PMDI/PF ratio sprayed on particle, the less formaldehyde released from particleboard, and one possible explanation for the reduction in formaldehyde from particleboard bonded with PMDI adhesive is that the PMDI molecules can react with water rapidly during hot-pressing to form amines, which can further react with other PMDI molecules to produce PMDI polymers [16].

The similar report also illustrated that the PMDI may have cured faster, and the PMDI adhesive film on the

particle surfaces may have prevented VOCs volatilizing from the particle surfaces [11].

The quantity of formaldehyde released from MF resin-impregnated paper-overlaid particleboard decreased with increasing PMDI/PF ratio, as shown in Table 2. This relationship could be represented by a linear regression formula (Table 3). The relationship between the quantity of formaldehyde released and the PMDI/PF ratio for the particleboards and MF resin-impregnated paper-overlaid particleboard samples is demonstrated in Fig. 1.

In comparison to the quantity of formaldehyde released from the particleboards tested and that released from MF resin-impregnated paper-overlaid particleboards was 10–21% lower, as shown in Table 2. In a study by Wiglusz et al. [10], the quantity of formaldehyde released from UF resin particleboard overlaid with a decorative paper foil in the early period after manufacture was 30% lower than that of UF resin-impregnated particleboard.

3.2. Bending properties and ultrasonic velocity

Results for the bending properties and ultrasonic velocity (VI) of specimens are listed in Table 4. It is evident that MOR and MOE values increased with increasing PMDI/PF ratio. In a comparison between PMDI- and

Fig. 1. Relationship between the quantity of formaldehyde released and PMDI/PF ratio. PMDI/PF ratio: weight of chips sprayed with PMDI/weight of chips sprayed with PF □: particleboard; ■: MF resin-impregnated paper-overlaid particleboard.

Table 4

Air-dried density, modulus of rupture (MOR), modulus of elasticity (MOE), ultrasonic wave velocity (VI), internal bonding strength (IB), screw holding strength (SH), thickness swelling (TS) and ultrasonic velocity (V_t) of tested particleboard sprayed with various PMDI/PF ratios

Sample group	Air-dried density (g/cm ³)	MOR (MPa)	MOE ($\times 10^2$ MPa)	VI (m/s)	IB (MPa)	SH (N)	TS (%)	V_t (m/s)
G	0.81 ± 0.03	11.4 ± 0.8	20.8 ± 1.0	2112 ± 52.6	0.56 ± 0.02	507.6 ± 64.0	18.1 ± 2.3	823 ± 37.6
D	0.82 ± 0.02	16.1 ± 1.2	25.1 ± 1.8	2271 ± 177.4	0.62 ± 0.02	638.0 ± 45.7	14.2 ± 2.5	865 ± 50.1
C	0.82 ± 0.02	17.0 ± 0.9	26.5 ± 1.2	2302 ± 30.6	0.65 ± 0.01	740.9 ± 46.8	11.8 ± 1.9	873 ± 51.1
B	0.79 ± 0.03	21.2 ± 1.1	27.9 ± 1.1	2446 ± 26.3	0.68 ± 0.03	799.7 ± 37.5	10.6 ± 1.4	891 ± 52.2
A	0.81 ± 0.04	23.2 ± 1.8	30.9 ± 0.7	2520 ± 122.8	0.73 ± 0.02	848.7 ± 55.1	7.1 ± 1.1	938 ± 67.1
MD	0.83 ± 0.02	18.9 ± 1.6	28.9 ± 1.8	2367 ± 36.0	0.62 ± 0.03	752.6 ± 42.1	13.8 ± 2.3	836 ± 47.6
MC	0.83 ± 0.02	19.6 ± 1.9	31.1 ± 2.9	2451 ± 83.2	0.65 ± 0.03	847.7 ± 46.3	10.1 ± 1.5	851 ± 53.0
MA	0.82 ± 0.04	27.9 ± 1.2	34.5 ± 1.6	2533 ± 77.9	0.73 ± 0.03	936.9 ± 50.4	7.0 ± 1.6	939 ± 63.3

Sample groups A, B, C, D and G: particleboards. Sample groups MD, MC and MA: MF resin-impregnated paper-overlaid particleboards.

PF-impregnated particleboards of the same density [17], higher MOE and MOR values were found for the PMDI-sprayed particleboards, similar to the results of our study. Except for group G, the MOR values for samples in groups A–F meet standard 13 (MOR > 13 MPa) of CNS 2215. Furthermore, the MOR values for groups A and B meet standard 18 (MOR > 18 MPa) of CNS 2215. With regard to MOE values, group A samples meet standard 18 (2998.8 MPa) and groups B, C, and D meet standard 13 (2499 MPa), whereas group G could only meet standard 8 (1999.2 MPa) of CNS 2215 [15].

The bending properties and ultrasonic velocity of MF resin-impregnated paper-overlaid particleboards with a PMDI/PF ratio of 50:50 (group MD), 70:30 (group MC) and 100:0 (group MA) are also shown in Table 4. In comparison to the bending properties of sample groups D, C and A, the MOR values for groups MD, MC and MA were 17.69%, 15.81% and 20.11%, whereas the MOE values were 14.86%, 17.64% and 11.92% higher, respectively. After retrofitting particleboard with MF resin-impregnated paper overlaid on the face and back layers, the bending strength of the MC group met standard 18 of CNS 2215 [15].

Values for the ultrasonic velocity (VI) are shown in Table 4. It is evident that the ultrasonic velocity propagated in the mechanical direction (VI) increased with increasing PMDI/PF ratio. In addition, there were significant positive relationships between the ultrasonic velocity and the bending strength, as shown in Table 3.

3.3. Internal bond strength

High internal bond strength was observed for the particleboards with different PMDI/PF ratios in our study because of the even distribution of the PMDI/PF resin on the chips. The IB values of the PMDI/PF-resin particleboards in our study meet standard 18 of CNS 2215 and are superior to those for PF resin-sprayed particleboards (0.12–0.42 MPa) [18]. Compared to particleboard manufactured from mixed species [19], including pine, aspen, oak and other hardwoods (IB = 0.56 MPa), the IB values in our study are also higher.

Bao et al. [20] established a PMDI bonding model by 2D NMR and illustrated the relationship between wood and PMDI. The major components formed from the curing of PMDI in wood are urea structures arising from the

reaction with water. PMDI can penetrate into the wood cells and into the middle lamellae between the wood cells, where it reacts with the available moisture stored in the wood to form either linear urea structures and/or cross-linking biuret/dimimer/trimer structures. In addition, the PMDI may react with the chemical components (the hydroxyl groups in polysaccharide or the lignin phenolic groups) of the wood to form urethane structures, and can further contribute to adhesion. The crosslinking can also serve to increase the rigidity of the plywood, especially at high levels of crosslinking.

Johns et al. [17] compared the same densities of particleboards, which were manufactured by PMDI, EMDI, UF and PF resin. The results indicated that the IB values of particleboard manufactured by PMDI and EMDI resin were higher than those of particleboard manufactured by UF and PF resin.

However, IB values for experimental particleboard and MF resin-impregnated paper-overlaid particleboard were not significantly different from each other (Fig. 2). This is because the IB depends on the bonding effect of chips in the core layer of particleboard, rather than on any retrofitting on the face and back layers.

Moreover, IB values increased linearly with increasing ultrasonic wave velocity through the thickness of the particleboard (Table 3). This result is in agreement with a previous study of particleboard in which a high correlation was reported between ultrasonic velocity measured along the length or thickness and the IB, with r^2 values of 0.66 and 0.92, respectively [13]. Previous studies also indicated a high degree of correlation between ultrasonic wave velocity propagated through the thickness and the IB of PF resin-impregnated OSB [21–23]. This suggests that the ultrasonic velocity could be satisfactorily used to assess the IB of boards.

3.4. Thickness swelling

Results for the thickness swelling of particleboards manufactured in our study for groups A, B, C, MA and MC were lower than 12% (Fig. 3 and Table 4) and meet standard 8 of CNS 2215 [15]. Table 4 also shows that the thickness swelling decreased with increasing PMDI/PF ratio. The reason for this relationship is possibly a swelling effect induced by PMDI. In comparison to the thickness swelling of the particleboards tested, no significant effect was observed for retrofitting with MF resin-impregnated paper overlaid on the particleboard, as shown in Table 4.

3.5. Screw holding strength

Table 4 shows that the screw holding strength (SH) of particleboards manufactured in our study was 508.0–849.3 N, which is higher than the values measured for urea adhesive particleboards (413.8–668.9 N) fabricated by Wang and Chen [18]. Moreover, compared to the quality of particleboard type 18 according to the CNS 2215

Fig. 2. (a) Modulus of rupture (MOR), modulus of elasticity (MOE), (b) internal bonding strength (IB) and (c) screw holding strength (SH) for various particleboards. Sample groups A, C and D: particleboards. Sample groups MD, MC and MA: MF resin-impregnated paper-overlaid particleboards.

standard (500.1 N), higher SH values were observed for the different particleboard groups in our study. Moreover, the SH values increased with increasing PMDI/PF ratio. The results could be represented by linear formulae, as shown in Table 3. Table 3 also shows that there was a high degree of correlation between SH and the ultrasonic velocity.

4. Conclusions

This study investigated the quantity of formaldehyde released and the properties of particleboard produced using recycled wood-waste particles with different PMDI/PF ratios. Results showed that 0.49 mg/L of formaldehyde was released for group D samples (with a PMDI/PF ratio of 50:50), which meets the E_0 standard of CNS 2215, whereas

Fig. 3. Thickness swelling (%) for various PMDI/PF ratios in the particleboards tested in 24 h. Sample groups A, B, C, D and G: particleboards. Sample groups MD, MC and MA: MF resin-impregnated paper-overlaid particleboards.

for samples with a PMDI/PF ratio up to 70:30, <0.3 mg/L formaldehyde was released. The quantity of formaldehyde released from MF resin-impregnated paper-overlaid particleboard was 17% lower than that for particleboard.

The MOR, IB, TS and SH results for PMDI/PF particleboard samples of groups A and MC were superior to those for conventionally manufactured particleboard and could meet standard 18 of CNS 2215. However, the TS for group D was higher than 12% and could not meet the CNS 2215 standard. In addition, MF resin-impregnated paper overlaid on the face and back layers of particleboard significantly decreased the quantity of formaldehyde released, and increased the MOE and MOR, but showed no significant effect on IB and TS.

The negative relationships between the quantity of formaldehyde released and the PMDI/PF ratio could be represented by linear regression formulae. The ultrasonic wave velocity through the face direction (V_f) increased linearly with increasing MOR and MOE. However, the ultrasonic wave velocity through the thickness of the particleboard (V_t) increased linearly with increasing internal bond strength and with decreasing thickness swelling. This suggests that ultrasonic velocity can be used to assess the properties of PMDI/PF particleboard.

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