Thermo-hydrolytic Recycling of Urea-Formaldehyde Resin-Bonded Laminated Particleboards

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The large global production of particleboards creates an equal quantity of particleboard waste after completion of their service life. Given increasing demand for green products and government environmental policies, it is urgent to develop technologies to recycle these used composite panels into valuable raw materials. This study was conducted to recover particles from waste laminated particleboards using various thermo-hydrolytic treatments. The recovered particles were used as raw materials with different substitutions of fresh particles to manufacture particleboard panels. The performance of the resulting particleboards was evaluated in terms of their mechanical properties and formaldehyde emissions. The nitrogen content of the control and resulting particles were measured to determine the resin removal in recycled particles. The results suggested that different thermo-hydrolytic treatments did not have significant influence on particles size distribution. Approximately 65% of urea-formaldehyde resin was removed from the particleboards treated at 140 °C/20 min. Particles recycled at 140 °C/20 min were comparable to fresh particles in terms of mechanical properties and formaldehyde emissions, and 100% of the recycled particles were used in the manufacture of particleboard without an adverse impact on the board performance.

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INTRODUCTION

Every year, a large amount of wood panel wastes is generated, consisting of used furniture and other constructed wood items, production panel residues, and rejects. These wood panel wastes are commonly disposed *via* landfilling or incineration. However, landfilling used panels is no longer considered an acceptable option because the leached resins and other chemicals (wax, scavenger, catalyst, and wood extractives) contaminate the groundwater, and biological degradation leads to the formation of methane, which causes the green-house effect about 80 times more than carbon dioxide (Kharazipour and Kües 2007). Current environmental regulations have limited the disposal of wood composite panels. Therefore, technologies to recycle this waste are important for protecting environment and for the sustainable development of the wood composite industry.

Three different principles are commonly applied for recycling wood composite panels: mechanical, thermo-hydrolytic, and chemical, or combinations thereof (Kharazipour and Kües 2007). The target of any recycling process is to recover high-quality particles or fibers for subsequent utilization as raw materials. Wan *et al*. (2014) investigated the impacts of three different panel disintegration methods—hammermilling, steam explosion, and thermal chemical impregnation—on the properties of recycled composite panels. Their research found that hammermilling was the easiest method to break down recycled panels, but it could not remove laminated papers from recycled panels. Thermal chemical impregnation using 0.5% butanetetracarboxylic acid solution disintegrates the urea-formaldehyde (UF) resin boned medium density fiberboard (MDF) and particleboard. The steam explosion method could be used in recycling all kinds of panels, but the recycled fibers could only be used for fiberboard. Particularly, the steam explosion treatment resulted in an increase of pH and buffer capacity of UF resin bonded panels, but a decrease in pH of phenol-formaldehyde (PF) resin bonded panels and an increase in the number of short fibers.

Particles from mechanical disintegration have a high degree of damage, which negatively impacts board properties. Moreover, particles from mechanical disintegration have a lower water retention value, and their wettability with UF, PF, and polymeric diphenyl-methandiisocyanates (pMDI) tends to be lower quality in comparison to fresh particles and particles obtained from thermohydrolytic degradation of composite panels (Roffael and Kraft 2004; Hameed *et al*. 2005a,b). For manufacturing of high-quality particleboards under conventional production conditions, no more than 20% of the raw material should come from such recycled wood. Due to the lower wettability of the recycled particles, higher amounts of adhesives are required for gluing, resulting in higher formaldehyde release from the products (Kharazipour and Kües 2007). One positive effect of mechanical disintegration is that no additional dry processes are needed for the chopped materials, which saves energy and avoids formaldehyde emission during drying (Kharazipour and Kües 2007).

In a thermo-hydrolytic treatment, steam and pressure are applied to cleave existing bonds in wood composite panels that were glued by hydrolysable adhesives. UF resin is the main adhesive used for particleboard manufacturing due to its low cost, short curing time, and high reactivity (Baharoğlu *et al*. 2012). Cured UF resins are susceptible to hydrolysis, so wood particles can be recovered from the used panels once the UF resin has been hydrolyzed. Cured UF resins are more susceptible to hydrolysis under moist acidic conditions than under neutral and alkaline conditions (Dutkiewicz 1983). Myers (1983) revealed that acid can catalyze the degradation of UF resin, acid lowers the activation energy required to disintegrate the structure of cross-linked UF resin. The UF resin residues in the recovered wood furnishes could significantly affect the curing behavior and bonding quality of the new resin system (Zhong *et al*. 2017). Lykidis and Grigoriou (2008) suggested that mild hydrothermal conditions should be used for particle recovery to avoid severe decrease of the mechanical properties of the resulting panels. Fu *et al*. (2020) reported that high temperature facilitated the decomposition of wood polymers and UF resin. Thermo-hydrolytic temperature and duration had significant effects on the chemical properties of wood particles. Wood particles recycled at a high temperature had a negative effect on the curing time of UF resin.

Thermo-hydrolytic degradation of UF resins may be catalyzed by acids originating from wood (such as formic and acetic acid) as well as other acids obtained from the usually used hardeners such as ammonium sulphate and ammonium nitrate. These acids may lower the activation energy needed to disrupt the structure of cross-linked UF resins, leading to the formation of UF pre-polymers. Lubis *et al*. (2018) used five aqueous solutions (including water, two acids, and two alkalis) to hydrolyze medium density fiberboard

(MDF) panels for removing the UF resin. They reported that an optimum condition for the removal of cured resins from MDF was 80 °C for 2 h using 1% oxalic acid solution. Lykidis and Grigoriou (2008) reported that most of the mechanical properties, except for the modulus of elasticity (MOE) of the particleboards manufactured with recycled particles, decreased in comparison to those of the particleboards manufactured with virgin wood particles. The degree of degradation in the quality of recycled particleboards can be significantly reduced by using milder conditions during hydrothermal treatments for particle recovery. One of the main issues of UF-bonded panels is formaldehyde emission, which is mostly due to the presence of unreacted free formaldehyde and slow but continuous hydrolysis of cured UF resin in the panels. Formaldehyde emission is an important factor in evaluating the environmental and health safety effects of wood-based panels. Formaldehyde concentration in indoor environments is restricted as it is considered a carcinogenic substance (IARC 2004). After thermal hydrolytic treatment, the cured UF resin in the recycled panels decomposes to some extent, but UF resin residues remain in the recycled particles. These resin residues may affect the curing behavior and bond quality of new particleboard made with recycled particles. Moreover, these resin residues probably affect the formaldehyde emission of the new particleboard.

The objectives of this study were to investigate the effectiveness of the thermo-hydrolytic treatment to disintegrate the cured UF resin in the waste particleboards; to evaluate the quality of the particleboard manufactured with recycled particles; to investigate whether the recovered particles were appropriate and comparable to fresh particles; and to determine the potential substitution level of the recycled particles for the fresh particles.

EXPERIMENTAL

Materials

UF resin and CASCOWAX® EW-58A wax emulsion designed for particleboard manufacturing were obtained from a local supplier (Hexion Canada Inc., Lévis, Québec, Canada) with the following typical characteristics: the viscosity of this UF resin was 0.355 Pa·s, its solid content was 67.2%, pH at 25 °C was 8.2, the buffer capacity was 11.49 mL, and the refractive index was 1.4772. The viscosity of wax emulsion was 0.030 to 0.250 Pa·s, its solid content was 58±1%, pH at 25 °C was 8.75±0.5, and its specific gravity was 0.935±0.005. Virgin particles were obtained from a local particleboard manufacturer (Uniboard Canada inc. Val d'or, Québec, Canada).

Thermo-hydrolytic Treatment

Used UF resin bonded paper-laminated particleboards collected from the waste furniture available at FPInnovations's Laboratory in Québec City (Canada) were cut into small pieces (2 in × 2 in). Subsequently these small pieces were soaked in hot water at 100 °C for 30 min, to remove their overlaid paper. Approximately 429 g (moisture content of 9.5%) of the particles after removing the overlaid paper and 2.5 L of tap water were transferred into a 5-L volumetric pressure reactor (Parr Series 4580 Bench Top Reactor, Moline, Illinois, USA). Three different thermo-hydrolytic treatments were performed in various temperature-duration conditions: 100 °C/100 min; 120 °C/60 min; 140 °C/20 min, and the resulting particles were identified as $P_{100\text{-}100}$, $P_{120\text{-}60}$ and $P_{140\text{-}20}$, respectively. For all the treatments, the treatment time started when the target temperature was reached. After the treatment, gas was released under fume hood. The virgin particle was identified as P_0. All resulting particles were oven-dried and stored in Ziploc bags.

Evaluation of Particle Size Distribution

The size distribution of the resulting particles was evaluated using the Tyler Standard Sieve Series (W.S. Tyler Company, Mentor, OH, USA). Approximately 60 g of oven-dried particles per sample was classified using 12-, 20-, 32- and 48-mesh screens and a pan. The screens and pan were used in stacks and equipped with a Tyler portable sieve shaker (Model RX-24, Gilson company, Inc. Lewis Center, OH, USA) by shaking 15 min for each sample to divide particles into five size fractions (size >1.4 mm, 0.84 to 1.4 mm, 0.5 to 0.84 mm, 0.3 to 0.5 mm, and <0.3 mm). The particle size distribution was based on the weight percentage of each size fraction.

Table 1. Experimental Design for Particleboard Manufacturing

Run No.	Particle Source	Panel ID (2 repetitions)
PB1	100% P_0	PB1-a & PB1-b
PB2	100% $P_{100\text{-}100}$	PB2-a & PB2-b
PB3	100% $P_{120\text{-}60}$	PB3-a & PB3-b
PB4	100% $P_{140\text{-}20}$	PB4-a & PB4-b
PB5	30% $P_{100\text{-}100}$ + 70% P_0	PB5-a & PB5-b
PB6	60% $P_{100\text{-}100}$ + 40% P_0	PB6-a & PB6-b
PB7	30% $P_{120\text{-}60}$ + 70% P_0	PB7-a & PB7-b
PB8	60% $P_{120\text{-}60}$ + 40% P_0	PB8-a & PB8-b
PB9	30% $P_{140\text{-}20}$ + 70% P_0	PB9-a & PB9-b
PB10	60% $P_{140\text{-}20}$ + 40% P_0	PB10-a & PB10-b

P_0: virgin particle; $P_{100\text{-}100}$: particle recycled at 100 °C/100 min; $P_{120\text{-}60}$: particle recycled at 120 °C/ 60 min; $P_{140\text{-}20}$: particle recycled at 140 °C/20 min.

Table 2. Particleboard Panel Manufacturing Parameters

Panel Dimension	6 mm × 610 mm × 610 mm (thickness × length × width)
Panel Construction	Homogeneous
Fresh And Recycled Particles Moisture Content	1 - 2% (before blending)
Target Panel Moisture Content	6 - 7% (after blending)
Wax Content	1% CASCOWAX®EW-58A (solids on a dry wood basis)
Resin Content	12% UF (solids on a dry wood basis)
NH_4Cl Content (Catalyst)	2% (solids on a liquid resin basis)
Estimated Resin Curing Time at 100 °C	55-62 sec.
Air Pressure for Resin Spray	40 psi
Resin Flow Rate	144.8 mL/min
Blender Dimension	914.4 mm (diameter) × 1219.2 mm (depth)
Blender Rotation Speed	30 rpm
Blending Sequence	Resin-catalyst-wax-wood (hand-forming)
Blending Time	9 min
Target Density	667 kg/m^3
Press Temperature	190 °C (platen surface)
Total Press Time	120 sec.
Press Closing Time	20 sec.
Degas Time	20 sec.
Replicate	2

Particleboard Manufacturing

A series of homogeneous particleboards were manufactured with different substitution levels of the recycled particles for virgin wood particles. The experimental design for particleboard manufacturing is presented in Table 1. Table 2 presents the particleboard panel manufacturing parameters.

Evaluation of Particleboard Performance

All the particleboards were cut into the specified dimension (Fig. 1) and identified. The resulting specimens were stored in a conditioning room at 65% RH/20 °C for two weeks until their equilibrium moisture content was reached prior to their property determination. All the resulting boards were evaluated for internal bond (IB) strength, modulus of rupture (MOR), modulus of elasticity (MOE), 24-h thickness swelling (TS), and 24-h water absorption (WA) according to the ANSI A208.1-2016. For the IB test, 24 specimens (50 mm x 50 mm) were prepared for each panel group, and only 12 specimens which density was closed to the target density were selected for IB measurement. The subsequence formaldehyde emission from the resulting particleboard was measured by the desiccator method according to the ASTM D 5582-14 (2014).

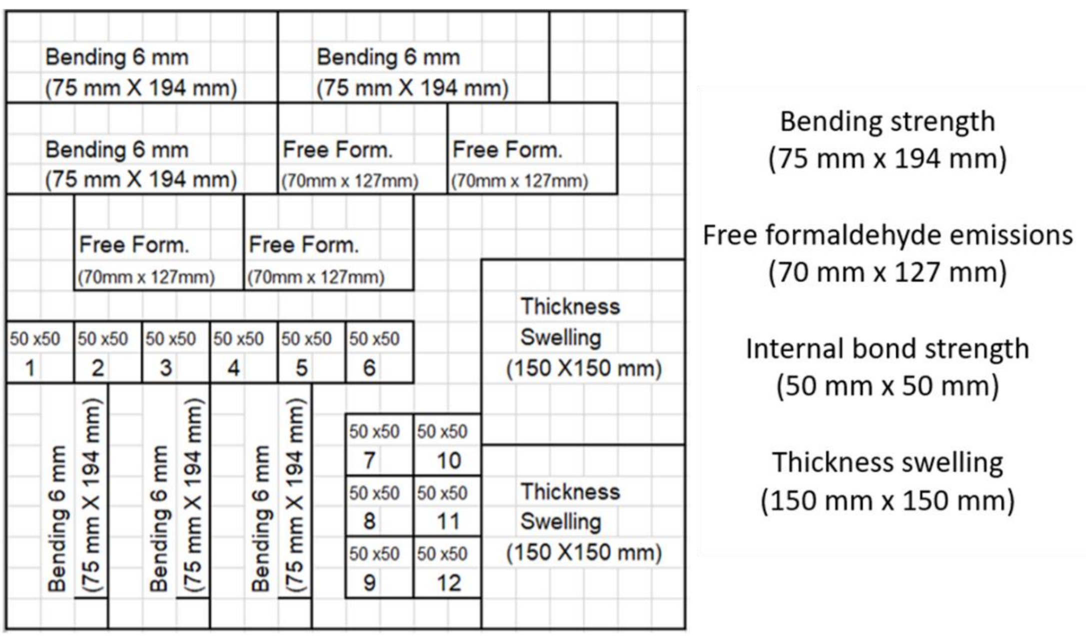

Fig. 1. Particleboard cutting plan used for physical and mechanical properties evaluation

Measurement of Nitrogen Content and Estimation of Resin Removal

PB1 containing 12% of UF resin content produced with 100% of virgin particles was cut into small pieces (2 in × 2 in) and ground into powder (< 1 mm) using a Thomas Wiley lab mill (Model 4, Thomas Scientific, Swedesboro, NJ, USA).

To investigate the efficiency of resin removal under different thermo-hydrolytic treatments, 400 g of oven-dried powder ground from PB1 with 2.5 L tap water were transferred into a 5-L volumetric pressure reactor (Parr Series 4580 Bench Top Reactor, Moline, Illinois, USA). Three different thermo-hydrolytic treatments were performed in various temperature-duration conditions: 100 °C/100 min; 120 °C/60 min; 140 °C/20 min. For all the treatments, the treatment time started when the target temperature was reached. After treatment, the recycled particles were oven-dried.

All of the resulting particles, powders obtained from PB1 and pure UF resin were sent to Université Laval (Laboratoire Daishowa, Centre de recherche en Horticulture, Québec, Canada) to perform the nitrogen content analysis using the digestion Kjeldahl method.

The resin content was calculated as follows,

$$\mathrm{RC} = \frac{N_p}{N_u} \times 100\% \tag{1}$$

where RC is the measured resin content (%), N_P is the nitrogen content (%) in the particle, and N_u is the nitrogen content (%) in the pure UF resin.

Resin removal (%) from the sample was calculated as follows,

$$\mathrm{RR} = \frac{(RC_b - RC_a)}{RC_b} \times 100\% \tag{2}$$

where RR is the resin removal, RC_b is the resin content (%) of particleboard particles before treatment, and RC_a is the resin content (%) of the particleboard particles after treatment.

Statistical Analysis

Duncan's test was conducted using SAS 9.4 (SAS Institute Inc., Cary, NC, USA) at the significance level α = 0.05, for multiple comparisons between the average values of several physical and mechanical properties of the resulting particleboards.

RESULTS AND DISCUSSION

Particle Size Distribution

Size distribution is an important physical property of wood particles. The smaller particle size results in higher IB strength of particleboard (Li *et al*. 2010). Particle size had a significant effect on MOR and MOE and a positive impact on dimensional stability of particleboard. Large particles achieve higher bending strength (Arabi *et al*. 2011; Rofii *et al*. 2013). The increase of particle size resulted in less water or moisture adsorption of particleboard. The particle size also had an important effect on the gel time of the UF resins. A shorter gel time was observed with the finer particles (Medved and Resnik 2004).

Table 3. Typical Size Distribution of Different Wood Particles

Size (mm)	P_0 (%)	$P_{100-100}$ (%)	P_{120-60} (%)	P_{140-20} (%)
>1.4	61.0	40.9	45.2	48.3
0.84-1.4	18.2	29.3	31.0	29.9
0.5-0.84	12.4	17.8	17.5	14.8
0.3-0.5	5.8	10.2	2.7	5.2
<0.3	2.7	1.8	3.7	1.8

P_0: virgin particle; $P_{100-100}$: particle recycled at 100 °C/100 min; P_{120-60}: particle recycled at 120 °C/60 min; P_{140-20}: particle recycled at 140 °C/20 min

Table 3 presents the size distribution of different wood particles used in this study. For all of the particles the highest percentage of particles (40.9 to 61.0%) was found for those being retained on a 12-mesh (size > 1.4 mm) screen. The lowest percentage of particles (1.8 to 3.7%) was found for those passing through a 48-mesh (size < 0.3 mm). Fresh particles (P_0) had more large particles (size > 1.4 mm) than those of recycled particles. Particles recovered from different thermo-hydrolytic treatments had similar size distribution. This result also indicated that the different thermo-hydrolytic treatments did not have significant influence on particles size distribution.

Mechanical Properties of Laboratory Particleboards

Table 4 presents mechanical properties of different homogeneous particleboards. The IB value of PB1 made with 100% of fresh particles was 0.69 MPa. The IB values of particleboard for PB2 to PB10 were higher than that of the PB1. From PB5 to PB10, the IB value of particleboard made with 70% P_0 (fresh particle) was lower than that of the particleboard made with 40% P_0, which might be due to the fresh particle had larger particle size than that of the recycled particles. These observations indicated that particle size had an important effect on the IB strength of particleboard. The smaller the particle size, the higher the IB strength of particleboard. This result is in accordance with the finding of Li *et al*. (2010). Another factor that may impact the IB is resin residues in the recycled particles. Zhong *et al*. (2017) reported that the resin residue could significantly affect the curing behavior and bonding quality of the new adhesive system. The author also found that a higher content of resin residue caused more decrease in the bonding strength. However, in this study, the results revealed that the particle size had a more important effect on the IB of particleboard than the content of resin residue.

The MOR and MOE of the control panel PB1 was 11.21 MPa and 2.06 GPa, respectively. Comparing the static bending properties of PB1 with other particleboards, the MOR and MOE of the PB4 and PB10 were close to those values of PB1.The MOR of PB4 was even higher than that of PB1. PB4 was made with 100% of recycled particles (P_{140-20}). This indicated that particles recycled at 140 °C/20 min were comparable to fresh particles (P_0) in terms of static bending properties of the resulting particleboard. PB9, PB10, and PB4, were made with 30%, 60%, and 100% substitution levels of the recycled particles (P_{140-20}) for fresh particles, respectively; it was observed that both the MOR and MOE increased with an increase in the substitution level.

Based on the above analysis, the particles recycled at 140 °C/20 min were appropriate and comparable to the fresh particles in terms of resulting board mechanical properties. Thus, 100% of these recycled particles could be used to manufacture the new particleboard without a significant deterioration in the term of the mechanical properties.

Hydroscopic Properties of Laboratory Particleboards

Thickness swelling and water absorption are very important issues for wood composite panels because they influence the dimensional stability and mechanical properties of panels. The 24-hour water soaking resulted in thickness swelling ranging from 14.9% to 19.2% as shown in Table 5. The PB5 (particleboard made with 30% of $P_{100-100}$ and 70% P_0) had the best dimensional stability in terms of the lowest thickness swelling and the least water absorption after 24-h water soaking at ambient conditions.

Formaldehyde emission values of the experimental panels ranged from 0.29 to 0.48 µg/mL. The lowest formaldehyde emission value was observed for panels made with 60% $P_{100-100}$ + 40% P_0 while the highest value was observed for particleboard made with 30% P_{120-60} + 70% P_0. From PB1 to PB4, the free formaldehyde emission of particleboard had no statistical difference. This indicated that the recycled particles used in the particleboard should not raise an environmental issue in terms of formaldehyde emission. The small difference in free formaldehyde emissions between particleboards might be due to the difference in UF resin residues from the recycled particles.

Table 4. Mechanical Properties of Different Laboratory Particleboards

Board ID	Particle Source	Moisture Content (%) n=4	Density (kg/m^3) n=4	Internal Bond (MPa) n=12	Modulus of Rupture (MPa) n=12	Modulus of Elasticity (GPa) n=12
PB1	100% P_0	8.35 (0.31)	633.75 (52.64)	0.69^{c} (0.17)	11.21^{abc} (2.14)	2.06^{a} (0.32)
PB2	100% $P_{100-100}$	7.87 (0.23)	637.00 (33.77)	0.90^{a} (0.18)	10.24^{bcd} (1.62)	1.55^{cd} (0.23)
PB3	100% P_{120-60}	8.43 (0.22)	627.75 (17.27)	0.72^{bc} (0.23)	9.69^{cd} (1.16)	1.45^{d} (0.21)
PB4	100% P_{140-20}	7.51 (0.23)	617.5 (45.04)	0.81^{abc} (0.19)	12.35^{a} (1.86)	1.91^{ab} (0.29)
PB5	30% $P_{100-100}$ + 70% P_0	8.71 (0.30)	615.25 (23.47)	0.80^{abc} (0.24)	9.49^{cd} (1.41)	1.67^{bcd} (0.25)
PB6	60% $P_{100-100}$ + 40% P_0	8.41 (0.23)	619.75 (33.81)	0.93^{a} (0.08)	9.39^{d} (1.79)	1.61^{cd} (0.26)
PB7	30% P_{120-60} + 70% P_0	9.01 (0.32)	627.00 (44.89)	0.73^{bc} (0.14)	10.59^{bcd} (1.84)	1.88^{ab} (0.31)
PB8	60% P_{120-60} + 40% P_0	8.20 (0.13)	615.50 (11.79)	0.81^{abc} (0.23)	10.51^{bcd} (2.98)	1.75^{bc} (0.47)
PB9	30% P_{140-20} + 70% P_0	7.60 (0.23)	642.25 (37.18)	0.70^{bc} (0.18)	10.00^{cd} (1.85)	1.75^{bc} (0.27)
PB10	60% P_{140-20} + 40% P_0	8.32 (0.34)	611.25 (32.72)	0.88^{ab} (0.20)	11.79^{ab} (1.62)	1.89^{ab} (0.24)

P_0: virgin particle; $P_{100-100}$: particle recycled at 100 °C/100 min; P_{120-60}: particle recycled at 120 °C/60 min; P_{140-20}: particle recycled at 140 °C/20 min. Values in parenthesis are standard deviations; Average values with the same letter indicate no significant difference at α = 0.05 in Duncan's multiple-comparison test

Table 5. Hydroscopic Properties and Free Formaldehyde Emission of Laboratory Particleboards

Board ID	Particle Source	Moisture Content (%) n=4	Thickness (mm) n=8	Density (kg/m^3) n=4	24 h- Thickness Swelling (%) n=8	24 h- Water Absorption (%) n=8	Formaldehyde Emission (µg/mL)
PB1	100% P_0	8.35 (0.31)	6.11 (0.10)	633.75 (52.64)	19.03^{a} (1.39)	27.18^{cd} (1.66)	0.38
PB2	100% $P_{100-100}$	7.87 (0.23)	6.26 (0.06)	637.00 (33.77)	19.03^{a} (0.96)	31.3^{ab} (2.34)	0.37
PB3	100% P_{120-60}	8.43 (0.22)	6.39 (0.19)	627.75 (17.27)	19.15^{a} (0.94)	33.40^{a} (1.59)	0.34
PB4	100% P_{140-20}	7.51 (0.23)	6.02 (0.16)	617.50 (45.04)	17.3^{ab} (2.5)	31.18^{ab} (1.31)	0.36
PB5	30% $P_{100-100}$ + 70% P_0	8.71 (0.30)	6.54 (0.11)	615.25 (23.47)	14.85^{c} (1.66)	25.80^{d} (0.98)	0.37
PB6	60% $P_{100-100}$ + 40% P_0	8.41 (0.23)	6.40 (0.10)	619.75 (33.81)	15.83^{bc} (0.54)	28.68^{bcd} (2.51)	0.29
PB7	30% P_{120-60} + 70% P_0	9.01 (0.32)	6.42 (0.15)	627.00 (44.89)	17.73^{ab} (0.99)	30.43^{abc} (2.80)	0.48
PB8	60% P_{120-60} + 40% P_0	8.20 (0.13)	6.44 (0.15)	615.50 (11.79)	16.38^{bc} (1.62)	27.55^{cd} (2.83)	0.33
PB9	30% P_{140-20} + 70% P_0	7.60 (0.23)	6.36 (0.07)	642.25 (37.18)	16.75^{bc} (1.62)	25.23^{d} (2.46)	0.46
PB10	60% P_{140-20} + 40% P_0	8.32 (0.34)	6.11 (0.11)	611.25 (32.72)	17.38^{ab} (0.48)	30.25^{abc} (3.05)	0.41

P_0: virgin particle; $P_{100-100}$: particle recycled at 100 °C/100 min; P_{120-60}: particle recycled at 120 °C/60 min; P_{140-20}: particle recycled at 140 °C/20 min. Values in parenthesis are standard deviations; Average values with the same letter indicate no significant difference at α = 0.05 in Duncan's multiple-comparison test.

Nitrogen Content and Resin Removal Estimation

The UF resin contains elemental nitrogen (N) from urea, so the nitrogen content in the UF-bonded panel can be used to estimate the quantity of UF resin in the panel. The nitrogen content was applied to evaluate the resin residue and resin removal ratio of wood fibers recycled from UF-bonded MDF undergoing different treatments (Lubis *et al*. 2018).

The nitrogen content and resin removal of the control and recovered particles are presented in Table 6. The nitrogen content of virgin wood particle (P_0) was 0.19%. This value is in good agreement with the finding of (Cowling and Merrill 1966), who found that most of wood species examined contained around 0.1% of nitrogen. The pure UF resin had 28% nitrogen content. The nitrogen content of particles from PB1 was 3.37%, much higher than that of P_0. The large amount of nitrogen likely comes from UF resins and catalyst NH_4Cl added during the manufacturing of particleboard. Therefore, any treatment for UF bonded particles without introducing nitrogen, the change of nitrogen content in wood particles might be caused by the change of the content of UF resins and NH_4Cl. After thermo-hydrolytic treatment, the nitrogen content of particles decreased to different extents. The lowest nitrogen content value was observed for particles recycled at 140 °C for 20 min. And 65.17% of UF resin can be removed at this treatment condition. Because the amount of nitrogen removed corresponds to the degree of UF resin decomposed, this study revealed that an elevated temperature can facilitate the decomposition of UF resins.

Table 6. Typical Resin Content and Resin Removal of Particles Treated under Different Thermo-hydrolytic Conditions

Treatment NO.	Nitrogen Content (%)	Resin Content (%)	Resin Removal (%)
P_0: virgin particle	0.19	-	-
Pure UF resin	28	-	-
Particles from PB1	3.37	12	-
$P_{100\text{-}100}$ recycled from PB1	1.62	5.79	51.75
$P_{140\text{-}20}$ recycled from PB1	1.17	4.18	65.17

Comparison Results of Particleboard Performance

Figure 2 presents the performance comparison results of particleboards made with 100% fresh particles and with 100% recycled particles ($P_{140\text{-}20}$), respectively. The requirements of mechanical properties such as internal bonding strength (IB), modulus of rupture (MOR) and modulus of elasticity (MOE) for M-S and M-2 grades of particleboards (ANSI A208.1-2016) are also shown in Fig. 2 for comparison. The IB, MOR, MOE, 24-h thickness swelling, 24-h water absorption, and free formaldehyde emissions of the fresh particleboard and particleboard with 100% recycled particles ($P_{140\text{-}20}$) were 0.69 MPa *versus* 0.81 MPa, 11.21 MPa *versus* 12.35 MPa, 2.06 GPa *versus* 1.91 GPa, 19.03% *versus* 17.3%, 27.18% *versus* 31.18%, and 0.38 µg/mL *versus* 0.36 µg/mL, respectively. The IB of particleboard made with 100% recycled particles ($P_{140\text{-}20}$) was higher than that of the M-S and M-2 grades of particleboards. The value of MOR and MOE of particleboard made with 100% recycled particles ($P_{140\text{-}20}$) was higher than those of M-S grade of particleboard but lower than those of the M-2 grade of particleboard. Those results indicated that the particles recycled at 140 °C/20 min are comparable to the fresh particles and can be used at 100% in the particleboard manufacturing without an adverse impact on the board performance.

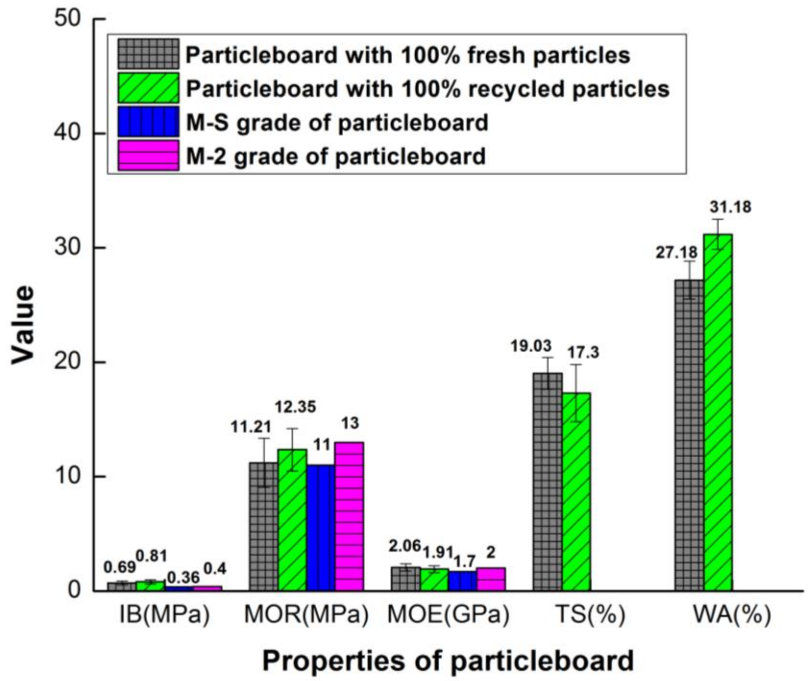

Fig. 2. Performance comparison between particleboards made with 100% fresh particles and 100% recycled particles (P_{140-20}).

CONCLUSIONS

1. Cured urea-formaldehyde (UF) resin is hydrolysable under steam and pressure, so wood particles can be recovered from the used panels once the UF resin has been hydrolyzed. Thermo-hydrolytic treatment is an effective method to recycle UF resin bonded particleboard.
2. Different thermo-hydrolytic treatments did not have significant influence on particles size distribution. Particle size has important effects on the internal bond (IB) strength and bending strength of particleboard.
3. Particles recycled at 140 °C/20 min had the lowest nitrogen content value. About 65.17% of UF resin was removed from the particleboards treated at 140 °C/20 min.
4. Particles recycled at 140 °C/20 min were comparable to the fresh particles and 100% recycled particles can be used in the particleboard manufacturing without an adverse impact on the board performance.

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REFERENCES CITED

ANSI A208.1 (2016). "American National Standard Particleboard," Composite Panel Association, Leesburg, VA, USA.

Arabi, M., Faezipour, M., Layeghi, M., and Enayati, A. A. (2011). "Interaction analysis between slenderness ratio and resin content on mechanical properties of particleboard," *Journal of Forestry Research* 22(3), 461-464. DOI: 10.1007/s11676-011-0188-2

ASTM D 5582-14 (2014). "Standard test method for determining formaldehyde levels from wood products using a desiccator," ASTM International, West Conshohocken, PA, USA.

Baharoğlu, M., Nemli, G., Sarı, B., Bardak, S., and Ayrılmış, N. (2012). "The influence of moisture content of raw material on the physical and mechanical properties, surface roughness, wettability, and formaldehyde emission of particleboard composite," *Composites Part B: Engineering* 43(5), 2448-2451. DOI: 10.1016/j.compositesb.2011.10.020

Cowling, E. B., and Merrill, W. (1966). "Nitrogen in wood and its role in wood deterioration," *Canadian Journal of Botany* 44(11), 1539-1554. DOI: 10.1139/b66-167

Dutkiewicz, J. (1983). "Hydrolytic degradation of cured urea–formaldehyde resin," *Journal of Applied Polymer Science* 28(11), 3313-3320. DOI: 10.1002/app.1983.070281101

Fu, Q., Wang, X. M., Cloutier, A., and Roussière, F. (2020). "Chemical characteristics of thermo-hydrolytically recycled particles," *BioResources* 15(2), 3774-3786. DOI: 10.15376/biores.15.2.3774-3786

Hameed, M., Behn, C., Roffael, E., and Dix, B. (2005a). "Benetzbarkeit von Recyclingspänen und, frischen "Holzspänen mit verschiedenen Bindemitteln," *European Journal of Wood and Wood Products* 63(5), 394-395. DOI: 10.1007/s00107-005-0003-x

Hameed, M., Behn, C., Roffael, E., and Dix, B. (2005b). "Wasserrückhaltevermögen von Recyclingspänen und von direkt aus Holz gewonnenen Spänen," *European Journal of Wood and Wood Products* 63(5), 390-391. DOI: 10.1007/s00107-005-0012-9

International Agency for Research on Cancer (2004). "IARC Classifies Formaldehyde as Carcinogenic to Humans," Lyon, France.

Kharazipour, A., and Kües, U. (2007). "Recycling of wood composites and solid wood products," in: *Wood Production, Wood Technology, and Biotechnological Impacts*, U. Kües (eds.), Universität Sverlag Göttingen, Germany, 509-533.

Li, X., Cai, Z., Winandy, J. E., and Basta, A. H. (2010). "Selected properties of particleboard panels manufactured from rice straws of different geometries," *Bioresource Technology* 101(12), 4662-4666. DOI: 10.1016/j.biortech.2010.01.053

Lubis, M. A. R., Hong, M. K., and Park, B. D. (2018). "Hydrolytic removal of cured urea–formaldehyde resins in medium-density fiberboard for recycling," *Journal of Wood Chemistry and Technology* 38(1), 1-14. DOI: 10.1080/02773813.2017.1316741

Lykidis, C., and Grigoriou, A. (2008). "Hydrothermal recycling of waste and performance of the recycled wooden particleboards," *Waste Management* 28(1), 57-63. DOI: 10.1016/j.wasman.2006.11.016

Medved, S., and Resnik, J. (2004). "Influence of the acidity and size of beech particles on the hardening of the urea-formaldehyde adhesive," *Acta Chimica Slovenica* 51(2), 353-360.

Myers, G. E. (1983). "Use of acid scavengers to improve durability of acid-catalyzed adhesive wood bonds," *Forest Products Journal* 33(4), 49-57.

Roffael, E., and Kraft, R. (2004). "Zum thermohydrolytischen Abbau von UF-Harzen in Holzspanplatten," *Holz als Roh-und Werkstoff* 62(2), 155-156. DOI: 10.1007/s00107-003-0455-9

Rofii, M. N., Yumigeta, S., Kojima, Y., and Suzuki, S. (2013). "Effect of furnish type and high-density raw material from mill residues on properties of particleboard panels," *Journal of Wood Science* 59(5), 402-409. DOI: 10.1007/s10086-013-1353-3

Wan, H., Wang, X. M., and Shen, J. (2014). "Recycling wood composite panels: Characterizing recycled materials," *BioResources* 9(4), 7554-7565. DOI: 10.15376/biores.9.4.7554-7565

Zhong, R., Gu, J., Gao, Z., Tu, D., and Hu, C. (2017). "Impacts of urea-formaldehyde resin residue on recycling and reconstitution of wood-based panels," *International Journal of Adhesion and Adhesives* 78, 60-66. DOI: 10.1016/j.ijadhadh.2017.06.019

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