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Effects of Particles Moisture, Conditioning Environment, and Heat Treatment on the Formaldehyde Emission of Urea Formaldehyde-Bonded Particleboard

Lum Wei Chen[1,2*], Lee Seng Hua[3], Petar Antov[4], Muhammad Adly Rahandi Lubis[5], Lubos Kristak[6], Dominik Hrusovsky[6]

[1]*Faculty of Forestry and Environment, Universiti Putra Malaysia, 43400,UPM Serdang, Selangor, Malaysia*
[2]*Faculty of Bioengineering and Technology, Universiti Malaysia Kelantan, Kampus Jeli, 17600 Jeli, Kelantan, Malaysia*
[3]*Department of Wood Industry, Faculty of Applied Sciences, Universiti Teknologi MARA Pahang, Kampus Jengka, 26400, Pahang, Malaysia*
[4]*Faculty of Forest Industry, University of Forestry, 1797 Sofia, Bulgaria*
[5]*Research Collaboration Center for Biomass and Biorefinery Between BRIN and Universitas Padjadjaran, National Research and Innovation Agency, Jatinangor 45363, West Java, Indonesia*
[6]*Faculty of Wood Sciences and Technology, Technical University in Zvolen, 96001 Zvolen, Slovakia*

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ABSTRACT. The initial moisture content (MC) and conditioning environment are among the most important factors that affect the properties as well as formaldehyde emission of particleboard. In this study, a three-layer rubberwood particleboard bonded with urea formaldehyde (UF) resin was prepared. Fine and coarse rubberwood particles with initial MC (of 3, 5, 7, 9 and 11%), respectively, were used to fabricate the three-layer particleboard. The produced particleboard was then evaluated for physical and mechanical properties. It was found that particleboard made with particles with 9% MC had the best properties. In the next phase of the study, this particleboard was chosen and exposed to different conditioning environments. The selected conditioning temperatures were 20 and 30 °C while the relative humidity was 30%, 65% and 100%. The findings suggested that the formaldehyde emission of particleboard increased with increasing conditioning temperature and relative humidity. However, the effect of the previous conditioning was nullified after the samples were re-equilibrated in a setting that served as the control. In order to reduce the formaldehyde emission of the particleboard, the samples were subjected to thermal treatment at 130, 150, 170 and 190 °C. It was found that the formaldehyde emission varied with treatment temperatures. Particleboard with the Super E0 level (≤ 0.3 mg/L) was successfully achieved by samples that were treated at 170 °C.

Keywords: Formaldehyde emission, Particleboard, Relative humidity, Thermal treatment

INTRODUCTION

Dimensional instability is one of the main drawbacks of particleboard in use (Lee et al., 2015). A higher dosage of resin may be effective in overcoming the dimensional instability; however, this led to a proportional increase in both the cost of manufacturing and the emission of formaldehyde (Antov et al., 2020; Kristak et al., 2022). For this reason, it was necessary to consider methods other than increasing the amount of resin dosage application in order to bring down the level of dimensional instability and emission issue.

When it comes to producing boards with desirable properties, one of the most important variables to consider is the

*Corresponding author: Tel.: +6012-5611256
E-mail address: *weichen.l@umk.edu.my*

moisture content of the particles. The moisture content of the particles has a significant impact on the properties of particleboard (Dukarska et al., 2022). The final board's moisture content will reflect levels of moisture in the particles that are either excessively high or inadequate. Low moisture content can result in poor surface wetting characteristics for the particles, which can then inhibit the flow of resin and transfer of the resin. Apart from that, the moisture content of the particles could affect the formaldehyde emission of the resultant particleboard. A study by Petinarakis et al. (2006) has listed a number of factors that could affect the formaldehyde emission level from particleboards. One of the important factors is the moisture content of the particles after mixing with resin. The study revealed that the formaldehyde emission increased along with increasing moisture content of the particles after mixing with resin.

Conditioning environment, i.e., temperature and relative humidity (RH), and condition period are important factors that could affect the properties of the particleboard. Atar et al. (2014) reported in their study that the particleboard that has been conditioned for a longer period of time exhibited superior mechanical strength, better dimensional stability, and lower formaldehyde emission. Jiang et al. (2017) conditioned particleboard samples in a ventilated environmental chamber maintained at three different temperatures, namely, 23, 35 and 50 °C. The authors found that the formaldehyde, as well as total volatile compounds emitted from the particleboard, increased along with increasing conditioning temperature. It is anticipated that, by conditioning the particleboard in an appropriate RH and temperature, the formaldehyde emission level of the particleboard can be better controlled.

Meanwhile, thermal treatment is an effective method to reduce the hygroscopicity and dimensionally instability of wood and wood-based panels (He et al., 2019; Kocaefe et al., 2015). Studies have shown that thermal treatment also exerts some effects on the formaldehyde emission of wood-based products. For instance, Murata et al. (2013) found that plywood made from thermally treated poplar veneers had different formaldehyde emission levels depending on the treatment temperature used. Therefore, the objectives of this study were to determine the effect that the initial particles' moisture content had on the mechanical properties and dimensional stability of the three-layer particleboard. In addition to this, the effects of the conditioning environment on the emission of formaldehyde and the dimensional stability of the three-layer particleboard were also investigated. Thermal treatment was applied to the particleboard in order to reduce the emission of formaldehyde. The effects of the heat treatment on the formaldehyde emission and the dimensional stability of the three-layer particleboard were evaluated.

METHODOLOGY

A three-layer particleboard with dimensions of 340 mm x 340 mm x 12 mm was produced using rubberwood particles. The manufacturing procedure was conducted according to the study reported by Lee et al. (2015). As can be seen in Table 1, there were five different levels of particle moisture content (MC) used: 3%, 5%, 7%, 9%, and 11%, respectively. These particles with different MC were used to fabricate particleboard and were denoted as 11%, 9%, 7%, 5% and 3% boards, respectively.

Table 1. Production of three-layer particleboard with different particle moistures

Board	Particle moistures (%)	
	Core	Fine
11%	11	11
9%	9	9
7%	7	7
5%	5	5
3%	3	3

For each panel, 8% UF resin based on the oven-dried weight of the wood particles was applied to the core layer, while 12% UF resin was applied to the surface layers. Meanwhile, 1% hardener (ammonium chloride) based on the solid weight of UF resin was applied to the core layer and 3.8% to the surface layers. 1% wax emulsion based on the oven-dried weight of wood particles was added to both surface and core layers.

Resultant mat moisture content

Before the mat is subjected to the hot press, the two most important sources of moisture are the moisture that is contained within the wood particles and the water that is added during the blending process of the additives (resin, wax, and hardener). The moisture content of the mat can be calculated based on the following equation:

Resultant mat moisture content, M_R (%) = $[(W_w + W_b) / (W_o + W_a)] \times 100\%$

where, W_w = weight of water contained in wood particles
W_b = weight of water added during blending
W_o = oven-dry weight of the wood particles
W_a = weight of additive solid

If we let $M_w = W_w / W_o$, $k = W_a / W_o$, and $M_a = W_b / W_a$, then we can get:

$$M_R (\%) = \frac{[M_w + kM_a]}{(1 + k)} \times 100\%$$

Where, M_w = moisture content of wood particles
M_a = moisture content of the additive
k = constant

Hot-pressing of particleboard

The panels were hot-pressed for 4.5 minutes at the temperature of 180 °C with a press pressure of 100 bars. After being pressed, the boards were conditioned for seven days at temperatures ranging from 20 ± 2 °C and with relative humidity levels between 65 ± 5%. Prior to the testing, each board was trimmed to the required dimension as outlined in JIS A 5908:2003 after it had been subjected to the conditioning process.

Conditioning environment

In this part of the research, samples were prepared for formaldehyde emission testing at six different exposures, which were as follow:

(1) 20° C – 30% relative humidity
(2) 20° C – 65% relative humidity
(3) 20° C – 100% relative humidity
(4) 30° C – 30% relative humidity
(5) 30° C – 65% relative humidity
(6) 30° C – 100% relative humidity

Every sample was dried out for a week in a desiccator before being analysed. Inside the desiccator, the relative humidity (RH) was managed by utilising water (at an RH level of 100%), saturated solutions of magnesium chloride, $MgCl_2$ (at an RH level of 65%), and sodium chloride, NaCl (at an RH level of 35%), respectively. Formaldehyde emissions were evaluated in accordance with JIS A 1460:2001 once the sample had been extracted from the conditioning desiccator.

Following the formaldehyde emission measurement, the same samples were reconditioned for another seven days at 65% relative humidity and 20 °C. This was done in order to investigate the effect of the conditioning environment on the subsequent formaldehyde emission from the board.

Heat treatment on particleboard

In accordance with the JIS A 1460:2001 standard, samples measuring 50 mm x 150 mm were prepared. The heat treatments were carried out at temperatures of 130 °C, 150 °C, 170 °C, and 200 °C, respectively. The samples were subjected to 30 minutes of pressing in a hot press at the temperatures as described earlier. The application of pressure has the purpose of densifying the samples while they are being heated in a hot press in order to compensate for the strength loss that was caused by the severe heat treatment. Nine samples measuring 50 mm by 150 mm were prepared for each temperature level. After the treatment, the samples were analysed with JIS A 1460:2001 to determine the amount of formaldehyde emission level.

Statistical Analysis

The results of the test were entered into the analysis of variance (ANOVA) in SPSS 16.0, and a comparison of the means of the treatments was analysed using Tukey's HSD test with a significance level of 5%.

RESULTS AND DISCUSSION

Resultant mat moisture content

The theoretical moisture content of the mat before hot pressing is summarised in Table 2. A moisture metre was used to calculate the actual moisture after blending. Based on the results shown in Table 2, the values obtained from the two approaches are quite comparable to one another. It is obvious that the mat moisture content is higher when particles with higher initial moisture content are used during the blending process.

Table 2. The theoretical and actual comparison of mat moisture after blending

Initial particle moisture (%)		**Moisture after blending (%) (Theoretical)**		**Moisture after blending (%) (Actual)**	
Core	Fine	Core	Fine	Core	Fine
11	11	16.33	17.13	16.02	17.16
9	9	14.04	14.92	13.64	14.96
7	7	11.85	12.79	11.51	12.15
5	5	9.75	10.74	8.91	10.32
3	3	7.73	8.78	6.98	8.56

Physical and mechanical properties of particleboards

The density and moisture content of the produced particleboard are shown in Table 3. 11% board was delaminated after the hot pressing process. Meanwhile, the density of the particleboard made with particles with an initial moisture content of 3% to 9% had density and MC ranging from 689.83 to 695.63 kg/m^3 and 6.56 to 9.85%, respectively. It was observed that the density and MC of the resultant boards increased along with increasing particle moisture.

Table 3. The density and moisture content of particleboard made with different particles of moisture

Board	**Density (kg/m^3)**	**MC (%)**
11%	delaminated	delaminated
9%	695.63	9.85
7%	694.71	8.63
5%	690.76	7.89
3%	689.83	6.56

After the blending process, particles of core and surface layers had different moisture content as a result of the different amounts of resin, hardener and wax received during the blending process. Particles of surface layers possess higher moisture than the core layer. The differentiation of moisture levels has the potential to produce two significant beneficial effects. Firstly, a more durable and smoother surface can be produced by first rapidly consolidating the more malleable particles that are present on the surface. The steam shock effect occurred due to the difference in moisture levels between the mats, and the moisture on the surfaces of the mat is quickly turned into hot steam and then rushed toward the cooler and drier regions of the mat, which are the core layer. This phenomenon helps in rapidly raising the temperature of the core, thereby reducing the time needed where the boiling point to be reached and providing a more complete resin cure in the core layer.

According to Table 4, however, 11% board that had a mat moisture content of more than 16% after blending experienced delamination in the core layer. When high levels of mat moisture are applied, problems with blow and delamination can occur (Cai et al., 2006). Blows, also referred to as blisters, are partial ruptures that occur close to the surface of the board. The term "delamination" refers to a separation that occurs between the board's two roughly equal halves (occurring in the core). Because moisture has a slowing effect on the curing of the resin, excessive mat moisture almost always results in an extension of the total amount of time required for the pressing process. During the hot pressing process, it is the cause of delamination in the board's core and, occasionally, delamination closer to the board surfaces (Hubbe et al., 2018). Therefore, a longer pressing time should be adopted. In this study, the delamination issue was resolved by increasing the pressing time from 4.5 minutes to 6 minutes. By increasing the hot pressing time, the moisture vapour that is being trapped in the core layer has sufficient time to escape and reduces the chance of sudden blows when the pressure is being released after the hot pressing process.

Table 4. Average values of mechanical and physical properties of particleboard made with different particles of moisture

Board	MOR (N/mm^2)	IB (N/mm^2)	TS (%)	WA (%)
11%	delaminated	delaminated	delaminated	delaminated
9%	17.04 ± 1.34^{a}	1.85 ± 0.09^{a}	11.67 ± 1.03^{a}	40.41 ± 3.98^{a}
7%	16.97 ± 1.09^{a}	1.62 ± 0.14^{a}	12.78 ± 1.11^{ab}	43.77 ± 4.67^{ab}
5%	15.51 ± 1.02^{ab}	1.41 ± 0.11^{ab}	12.99 ± 1.15^{ab}	46.52 ± 2.78^{b}
3%	14.44 ± 0.98^{b}	1.25 ± 0.09^{b}	13.79 ± 1.07^{b}	47.58 ± 4.22^{b}

Note: Means followed by the same letter in the same column are not significantly different at $P\leq0.05$.

Generally, an increase in the initial particles' moisture content could lead to an increase in the boards' mechanical properties. When there is a higher percentage of mat moisture, the wood particles become more pliable (Rosenfeld et al., 2022). 9% of boards had the highest modulus of rupture and internal bonding strength. The increased amount of moisture content contributes to the production of a mat that is more compressible, which in turn promotes the board's compaction ratio. Thickness swelling and water absorption of the particleboard decreased as the initial particle moisture content increased. 9% of boards exhibited the lowest thickness swelling and water absorption compared to the boards made with lower initial particles' moisture content. In addition, 9% of boards had a thickness swelling value of 11.67%, fulfilling the requirement of JIS A 5908, which only allows a maximum swelling value of 12%. It is possible to explain that wood particles with a higher initial moisture content were partially filled with water, and as a result, their capacity to absorb more water and swell is limited.

Low moisture content can result in poor surface wetting characteristics for the particles, which can inhibit the flow of resin and transfer of the resin (Baharoğlu et al., 2012). It is believed that the insufficient amount of binder in the particle-to-particle contact area is caused by the excessive amount of resin absorbed by the excessively dry particles (Lee et al., 2015). The lack of pliability that results from the dry mat's composition causes a reduction in the total contact area within the structure of the mat, which is perhaps the most significant disadvantage associated with a dry mat.

Effect of RH and temperature on formaldehyde emission

Figure 1 illustrates the effects of relative humidity and temperature environment on the emission of formaldehyde of the particleboard. At low temperatures and low RH (i.e., 20 °C and 30%), the particleboard had an emission class of SE0 (≤ 0.3 mg/L). At 30°C and 30% RH, E0 class was observed (≤ 0.5 mg/L). Particleboards that were being conditioned at other temperatures and RH had an emission class of E1 (≤ 1.5 mg/L). However, when at a high temperature and high RH (i.e., 30 °C and 100%), the formaldehyde emission from the particleboard increased drastically and achieved E2 emission class, which is > 1.5 mg/L and ≤ 5 mg/L.

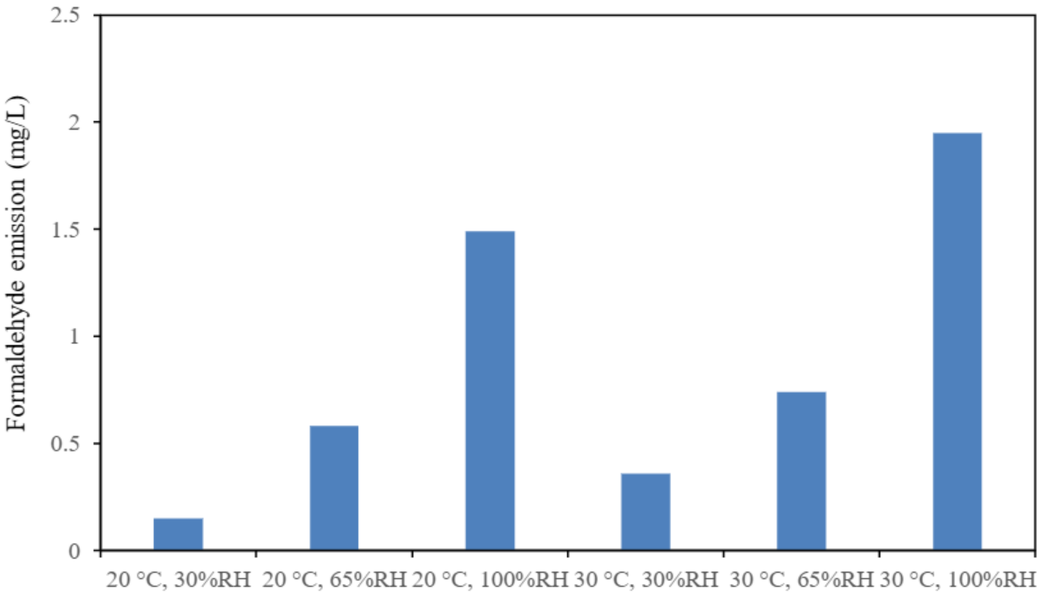

Figure 1. Formaldehyde emission of particleboard conditioned at different conditioning environments.

Based on Figure 1, at a constant temperature of 20°C, an increase in relative humidity from 30% to 65% or 100% results in a 3.87- and 9.93-fold increase in the amount of formaldehyde emitted. The emission of formaldehyde increased by a factor of 13 when the temperature was raised from 20°C to 30°C from a relative humidity of 30% to

100%. These findings are comparable to those found by Parthasarathy et al. (2011), who discovered a 1.9-3.5-fold increase in the amount of formaldehyde emission when the temperature was raised by 10°C and an increase of 1.8-2.6-fold when the relative humidity was increased by 35%.

Table 5. Formaldehyde emission of re-conditioned particleboard

Board	Formaldehyde emission (mg/L)
20°C, 30% RH	0.53
20°C, 65% RH	0.58
20°C, 100% RH	0.62
30°C, 30% RH	0.55
30°C, 65% RH	0.59
30°C, 100% RH	0.53

Nevertheless, although the samples were conditioned in a variety of environments, the results of the reconditioning at 65% RH and 20°C showed that the samples had the same emission (Table 5). According to the table, there was not a discernible difference found. It is possible to draw the conclusion that exposure to the various environments did not have an irreversible effect on the emission of formaldehyde from the particleboard. The effect of the previous conditioning was nullified after the samples were re-equilibrated in a setting that served as the control.

Effect of heat treatment on formaldehyde emission

Formaldehyde emission values of the UF-bonded particleboard that was being treated with different temperatures are presented in Figure 2. Heat treatment up to 170°C is effective in reducing the formaldehyde emission of the particleboard.

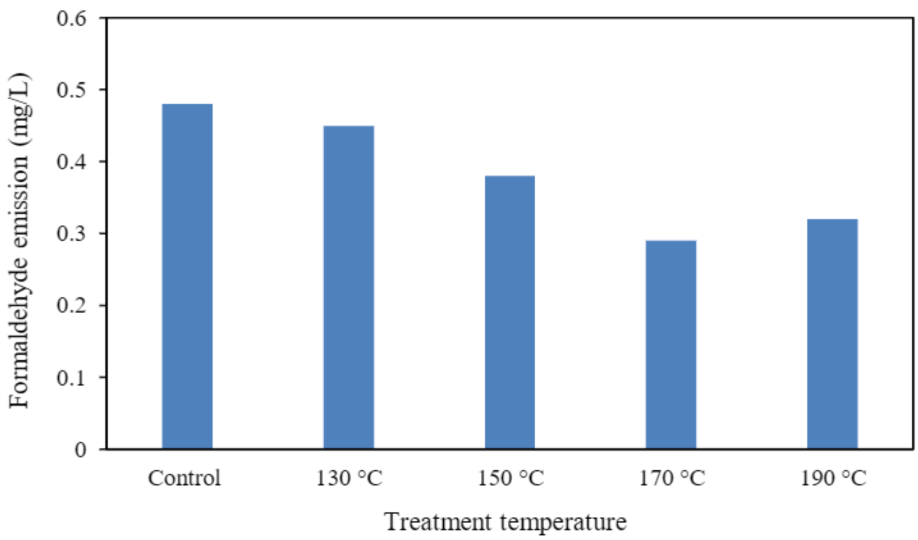

Figure 2. Formaldehyde emission of particleboard treated with different temperature

According to Hasegawa's (2008) findings, heat treatment facilities the release of formaldehyde from solid wood into the atmosphere. Heat treatment increases the rate of formaldehyde emission, but this rate decreases after the material is subjected to conditioning. In this study, formaldehyde emission was found to be significantly lowered after heat treatment when compared to the control samples. According to Figure 2, the Super E0 level (≤ 0.3 mg/L) was successfully achieved by samples that were treated at 170°C. Normally, when being heated at high temperatures, the mechanical properties of the particleboard decrease as a result of the thermal decomposition of wood chemical components as well as the degradation of UF resin (Lee et al. 2015). However, in this study, the particleboards were heat treated using a hot press with compression applied. The particleboard became more compact and therefore eliminated the negative effects induced by high temperature, as reported in the study by Lee et al. (2015). Therefore, a very minor reduction in strength was observed in the heat-treated particleboard in this study.

A similar observation was made by Hassan et al. (2020), where the formaldehyde emission of *Ficus retusa* branch wood particleboard decreased along with increasing heat treatment temperature. The internal structure of the cell wall was unaffected by heat treatment carried out at temperatures of 150°C and 170°C; however, the adsorption properties of the monomolecular layer were modified. The adsorption coefficients had a general tendency to decrease as the heat temperature increased, but they reached a plateau at 170°C (Murata et al., 2013). When a glue that contained formaldehyde was utilised, the formation of hemiacetal occurred as a result of the reaction between the hydroxyl groups of cellulose and hemicellulose and the formaldehyde that was released from an uncured adhesive. It is possible that the adsorption property of a monomolecular layer can have an effect on the hemiacetal reaction of the hydroxyl groups. This is because the adsorption property of a monomolecular layer is related to the accessibility of the hydroxyl groups. The decrease in adsorption coefficients led to a decrease in the accessibility of hydroxyl groups, which in turn led to a decrease in the reaction between the adhesive and the hydroxyl groups, which resulted in less formaldehyde being released as a byproduct after the adhesive had cured (Murata et al., 2013).

CONCLUSION

In this study, particleboards were fabricated from rubberwood particles with initial moisture contents of 3 to 11%. Thermal treatment was also performed to investigate its effects on reducing the formaldehyde emission of the particleboard. Generally, it can be concluded that rubberwood particles with an initial moisture content of 9% produced the most dimensionally stable particleboard with the best mechanical properties. Particles with too high of moisture content (11%) caused blows and delamination in the particleboard, while too low moisture content tends to retard water transfer to the core layer. When being conditioned in high temperature and relative humidity, the particleboard emitted higher formaldehyde compared to those being conditioned in lower temperature and relative humidity. However, the change in the environment did not have an irreversible effect on the amount of formaldehyde that was emitted from the particleboard. The effect of the previous conditioning was nullified after the samples were re-equilibrated in a setting that served as the control. In terms of formaldehyde emission reduction, heat treatment at temperatures ranging from 150 to 170°C proved to be very effective. This is because such treatment can control the emission of formaldehyde from particleboard while having very little impact on the mechanical properties of the material.

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AUTHOR CONTRIBUTIONS

Lee Seng Hua and **Lum Wei Chen** are responsible for conducting the research and drafting this manuscript. **Petar Antov, Muhammad Adly Rahandi Lubis, Lubos Kristak** and **Dominik Hrusovsky** are responsible for reviewing and editing the manuscript.

COMPETING INTEREST

The authors declare that there are no competing interests.

COMPLIANCE WITH ETHICAL STANDARDS

Not applicable.

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