Physico-Mechanical Properties and Formaldehyde Emission of Rubberwood Particleboard Made With UF Resin Admixed With Ammonium and Aluminium-Based Hardeners

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ABSTRACT

In this study, the effects of addition of ammonium and aluminium-based hardeners into urea formaldehyde resin (UF) on the physico-mechanical properties and formaldehyde emission of the rubberwood particleboard were investigated. Four types of hardeners, namely ammonium chloride (AC), ammonium sulphate (AS), aluminium chloride (AlC) and aluminium sulphate (AlS), were added into UF resin. The acidity, gelation time, viscosity and free formaldehyde content of the UF/hardener mixtures were determined. Particleboard made with the UF/hardener mixtures were tested for physico-mechanical properties and formaldehyde emission. The pH values of the resin after addition of aluminium-based hardeners were higher and resulted in higher viscosity and shorter gelation time. Consequently, despite lower formaldehyde emission was recorded, the physico-mechanical properties of the resulted particleboard were inferior compared to that of ammonium-based hardeners. The best quality particleboard in terms of mechanical, physical and formaldehyde emission were obtained from the particleboard made with AS, followed by AC.

INTRODUCTION

A great variety of aminoplastic resins are in use in present-day wood-based panels industry. Among the aminoplastic resins,

urea formaldehyde (UF) resin is the most prevalently used binding agent in wood based panels industry, particularly in particleboard manufacturing, owing to its low cost and high reactivity (Moslemi, 1974). Nevertheless, aminomethylene linkages in UF resin are instable and susceptible to hydrolysis in high relative humidity state, which generate a permanently release of formaldehyde (Dunky, 1998). Formaldehyde is classified as carcinogen to human by The International Agency for Research on Cancer (IARC) and therefore the emittance of formaldehyde has caused concern among the users and manufacturers.

In order to tackle the aforementioned issue, efforts have been taken over the past decades in reducing the level of formaldehyde emission (Younesi-Kordkheili et al., 2016). The most economic and direct ways of reducing formaldehyde emission is through lowering the formaldehyde to urea (F/U) molar ratio in UF resin. Unfortunately, lowering F/U molar ratio inevitably gives adverse effect on the properties of the produced particleboard. In addition, Maminski et al. (2008) reported that the possibilities by lowering F/U ratio had been exhausted with the ratio reduced to 0.85. The results revealed that the formaldehyde emission did not reduce significantly but the strength of joints produced from the mentioned resin was around 20% lower compared to that of the resin with F/U ratio of 1.1.

On account to that, addition of formaldehyde scavenger, or formaldehyde catcher into the resin is the most convenient and effective method to reduce the emittance of formaldehyde from wood or wood-based products (Aizat et al., 2017; Lum et al., 2014; Zaidon et al., 2016). One of the methods to reduce formaldehyde emission is to modify the chemistry of urea formaldehyde resins by using other ammonium salts as cure catalysts or hardener instead of latent ammonium chloride. Curing agents, also called hardener or catalyst are chemical substances added to the UF resin to speed up polymerization. UF resins are acid catalyzed resin and therefore acidic environment is needed for it to cure. These chemicals are either acidic substance by themselves or can liberate acids when mixed with the adhesives. These hardeners are normally used for UF-resin curing, however, in excess, they can act as formaldehyde scavengers which react with free formaldehyde to form hexamine (Moslemi, 1974). The most widely used hardeners are ammonium salts of strong acids which usually include the salts of chloride, sulphate, phosphate, nitrate, fluoride and borate. Non-ammoniacal salts such as aluminium and magnesium salts was also reported as potential hardener as well as formaldehyde scavengers for UF resin (Atar et al., 2014). Dunky (1998) reported that direct addition of acids such as maleic acid, formic acid, and phosphoric acid or acid compounds which dissociated in water such as aluminium sulphate were also one common practice in facilitate the curing speed of the resin.

The function of hardener is to react with the free formaldehyde or any uncombined free formaldehyde that presents in the UF resins where the reaction releases acid, hexamine as a by-product and water. The amount and type of hardener used in the resin formulation were found have a significant influence on the formaldehyde release from the resin and the UF-

bonded particleboard (Atar et al., 2014). Saffari (2011) identified that particleboard made with magnesium chloride released higher formaldehyde emission than those particleboards made with ammonium chloride and ammonium sulphate, respectively. On the other hand, Aras, Kalaycioglu et al. (2015) compared the effects of ammonium chloride and ammonium nitrate and found that the latter improved the physical properties and reduced the formaldehyde emission of the particleboard produced.

Therefore, it is important to study the effect of hardener used in UF-bonded particleboard production in order to obtain the lowest formaldehyde emission from the particleboard produced with most optimum quality of particleboard. The objective of this study was to evaluate the effects of ammonium- and aluminium-based hardeners addition to the UF resin properties and its effects on the particleboard fabricated from the resin. Properties such as thickness swelling, bending strength, internal bonding strength and formaldehyde emission of the particleboard were determined.

MATERIALS AND METHOD

Materials Preparation

Urea formaldehyde (UF) resin typed E1 was provided by Aica Chemical Sdn. Bhd and the resin specifications are listed in Table 1. Four types of hardeners, namely ammonium chloride (AC), ammonium sulphate (AS), aluminium chloride (AlC) and aluminium sulphate (AlS) were purchased from Evergreen Engineering & Resources. Rubberwood particles with 3% moisture content were obtained from a local particleboard manufacturing plant, Heveaboard Berhad which located in Gemas, Negeri Sembilan.

Table 1

Specifications of the urea formaldehyde resin used in this study provided by the supplier

Properties	Results	Specifications
Viscosity at 30°C (cps)	215	200-270
% N.V.C 3hrs at 105°C	66.9	66.0-68.0
pH at 30°C	8.62	8.5-10.0
Density at 30°C	1.282	1.280-1.290
Gel time at 100°C (Sec)	75	55-75

Evaluation of Properties of Admixture of Urea Formaldehyde (UF) Resin and Hardeners

Approximately 50g of UF resin was weighed to serve as a control. 1% of each type of hardener (based on the solid content of the UF resin) was weighed and added to UF resin,

respectively. The acidity, viscosity, gelation time and free formaldehyde content of the resin/hardener mixtures were determined.

Acidity (pH). Mi105 pH/temperature professional portable meter was calibrated with buffer 4.0 and 10.0 and the resin/hardener mixtures were cooled to 30°C. Next, the pH meter electrode was immersed into the mixtures and the pH reading was recorded.

Viscosity. Viscosity of the UF/hardener mixtures were measured with an AMETEK Brookfield rotational viscometer & rheometer at 20 °C with a spinning rate of 1 rpm according to the procedures stated in Ghani et al. (2018). 75ml of UF resin was poured into a 100ml beaker. Then, the spindle was lowered into the resin until the notch was just touched the liquid surface. Next, the reading from the viscometer was recorded.

Gelation time. Gelation time of the UF/hardener mixtures were determined according to the Ghani et al. (2018). Mixtures of UF resin and hardeners were poured into a beaker and stirred well. After that, 6.5g of the mixture was poured into a test tube which was then immersed (below water line) in 100 °C water bath. Immediately, the content was continuously stirred and the time (in seconds) needed for the resin mixtures to cure was recorded.

Free formaldehyde content. UF resin (10g) and 50ml of dimethyl sulphoxide solution were weighed and poured into a 250ml Eerlenmyer flask. HCl (0.1M, 30ml) and Na_2SO_3 (01.M, 30 ml) was added immediately to the mixture and stirred well. The mixture solution was cooled in an ice bath for 3 minutes to ensure complete reaction of the formaldehyde with sulphite. After 3 minutes, 1 ml 0.1% tylmolphtalein solution was added and the excessive acid was titrated with 0.1M NaOH solution to blue color. Volume of the 0.1M NaOH used was recorded as V_1. The blank test without addition of UF resin was carried out under the same condition and the volume of 0.1M NaOH used was recorded as V_2. The experiment was repeated using 1% of each hardener added to the UF resin. The free formaldehyde content was then calculated using the equation 1 as below:

% free formaldehyde = $[(V_2 - V_1) \times M \times 3.002] / W$ [1]
where;
V_1 = Volume in ml of 0.1M NaOH solution for resin
V_2 = volume in ml of 0.1M NaOH solution for blank test
M = molarity of NaOH solution
W = weight in grams for resin

Fourier Transform Infra-Red (FTIR) Spectroscopy

Perkin-Elmer FT-IR Spectroscopy (model spectrum 100 series, USA) was used to determine any differences occurring to the functional group on UF resin sample and after the UF resin

was mixed with different hardeners. FT-IR spectra tests were run at ambient temperature using cured samples within the wave number range of 4000 to 500 cm^{-1} and at a resolution of 4 cm^{-1}.

Particleboard Production

Particleboards with dimensions of 340 mm width x 340 mm length x 12 mm thickness were fabricated from the rubberwood particles resinated with the UF resin admixed with four respective type of hardeners. The target density for the boards produced was 650 kg/m³. 8% of UF resin (based on the oven-dried particles weight) was mixed with 0.5% wax (based on oven-dried particles weight) and 1% proposed hardener (based on resin solid content) and sprayed onto the rubberwood particles during blending process. After the particles were blended with resin, the resinated rubberwood particles were manually distributed into a wooden mold having dimensional of 340 mm length x 340 mm width to form a mat. The formed mats were then hot-pressed in a hot press at 180 °C for 270 s under pressure of 4 MPa. The produced particleboards were kept in a conditioning room at relative humidity of 65 ± 5% and temperature of 23 ± 3 °C until constant weight was reached. After conditioned, the samples were cut according to the relevant standard for properties evaluation.

Properties Evaluation

Samples for properties evaluation were prepared in accordance with JIS A 5908: 2003. Properties such as density, moisture content, thickness swelling, water absorption, modulus of rupture, modulus of elasticity and internal bonding strength were conducted according to the procedure specified in JIS A 5908: 2003. On the other hand, formaldehyde emission from the particleboards was determined in accordance with JIS A 1406: 2001.

Statistical Analysis

The data were analyzed statistically to verify the significance of the variable studied. The data were analyzed using Statistical Package for the Social Science (SPSS) procedure for the analysis of variance (ANOVA) at 95% confidence level (P≤0.05). Tukey's HSD (Honestly significance difference) test was performed to further determine the significance level of the test properties.

## RESULTS AND DISCUSSION

Properties of the UF resin and UF/hardener Mixtures

Acidity, gelation time, viscosity and free formaldehyde content of the UF resin before and after addition of different hardeners, namely ammonium chloride (AC), ammonium sulphate

(AS), aluminium chloride (AlC) and aluminium sulphate (AlS) are listed in Table 2. The initial pH value and gelation time of the UF resin without addition of hardener was 7.4 and 75 s, respectively, with the viscosity of 186 cp and free formaldehyde content of 0.25%. After the addition of hardeners, the selected properties of the UF resin varied accordingly to the type of hardener used.

Generally, after the addition of hardener, the mixtures of UF and hardener displayed lower pH values, shorter gelation time, higher viscosity and lower free formaldehyde content. The lowest pH value was recorded when UF were mixed with AlC and AlS, which was 3.2 and 3.0, respectively. The findings were in line with Bektha et al. (2016) who reported that the pH values of the UF resin admixed with aluminium sulphate for birch plywood production were ranged from 2.7-2.8. Correspondingly, these mixtures had the shortest gelation time of 28 and 31 s, respectively, and the highest viscosity. In comparison, higher pH value of 5.7 and 5.5 were observed when UF were mixed with ammonium-based hardener, AC and AS, as well as longer gelation time and lower viscosity.

UF resin is well known as an acid catalyzed curing resin. Reducing pH value values from alkaline to acidic condition indicating that the addition of hardener has increased the acidity of the UF resin and such acidity is necessary as it acted as an acid catalyst to facilitate the curing of the UF resin (Atar et al., 2014). The ammonium and aluminium based hardeners, also called as curing agents, are chemical substances added to the UF resin to speed up the polymerization, where these substances can liberate acids when mixed with the UF adhesive by reacting with any uncombined formaldehyde shown in the following equations. The reaction of ammonium chloride hardener with formaldehyde will form hexamethylenetetramine, hydrochloric acid and water as shown in equation 2.

$$4\ NH_4Cl + 6\ HCHO \rightarrow (CH_2)_6N_4 + 4\ HCl + 6\ H_2O \qquad [2]$$
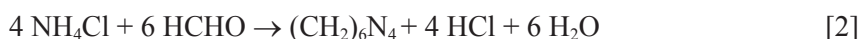
On the other hand, reaction of ammonium sulphate with formaldehyde will form hexamethylenetramine, sulphuric acid and water as shown in equation 3.

$$2\ (NH_4)_2SO_4 + 6\ HCHO \rightarrow (CH_2)_6N_4 + 2\ H_2SO_4 + 6\ H_2O \qquad [3]$$

As for aluminium chloride, the hardener will react with free formaldehyde in the UF adhesive, then it will liberate aluminium formate and hydrochloric acid.

$$2\ AlCl_3 + 6\ HCHO \rightarrow 2\ (HCOO)_3Al + 6\ HCl \qquad [4]$$

By using the aluminium sulphate as the hardener, the reaction will produce aluminium formate, sulphuric acid and hydrogen ion.

$$Al_2SO_4 + 6\ HCHO \rightarrow 2\ (HCOO)_3Al + H_2SO_4 + H^+ \qquad [5]$$

According to Atar et al. (2014), the free formaldehyde in the UF adhesives resin reacts with the hardener to generate acid. The liberation of acid in this reaction results in immediate decreases of pH value. The hardener acts as an acid catalyst for the curing

reaction of the UF resin and subsequently shortens the gel time and leads to increasing of the viscosity of the resin.

The addition of hardener also affects the free formaldehyde content in the UF resin. When 1% hardener were incorporated into the UF resin, a slight reduction in free formaldehyde content was observed in comparison to UF resin alone (0.25%). Based on the result, the free formaldehyde content after the addition of aluminium-based hardener into the UF resin was reduced to 0.20%-0.21% compared to 0.22%-0.24% after the addition of ammonium-based hardener. Aluminium-based hardener portrayed a relatively lower free formaldehyde content as the hardener increased the depth of hardening of urea formaldehyde oligomers. Additionally, in the case of using aluminium-based hardener, a quite low pH obtained due to the formation of acetal linkage and resulted in additional formaldehyde binding (Bekhta et al., 2016).

Table 2

Acidity, gelation time and viscosity of the UF resin admixed with different hardeners

Type	pH of resin	Gelation time (s)	Viscosity (cp)	Free formaldehyde content (%)
UF	7.4	75	186	0.25
UF + AC	5.7	66	201	0.24
UF + AS	5.5	65	217	0.22
UF + AlC	3.2	28	277	0.20
UF + AlS	3	31	283	0.21

Characterization of UF resin and UF/hardener Mixtures Using FT-IR Spectroscopy

The effects addition of different type of hardeners on the chemical structure of the urea formaldehyde resins were analyzed by FT-IR spectroscopy and the results are illustrated in Figure 1. The assignments of the characteristic IR absorption peaks are summarised in Table 3.

From the FTIR spectra shown in Figure 1, basic structures of UF resin ($CO(NH_2)_2$) are shown where strong absorptions were detected at 1637 cm^{-1} and 1554 cm^{-1} which is assigned to amide I (C=O stretching) and amide II (N-H bending and C-N stretching), respectively, as well as CH_2OH, CH_3 and CN at the regions around 1400–1360 cm^{-1} (Zorba et al., 2008). The broad bands at 3000 to 3700 cm^{-1} has been identified as hydroxyl (-OH) stretching of the methylol group (Jada 1988). From the figure, it can be seen that
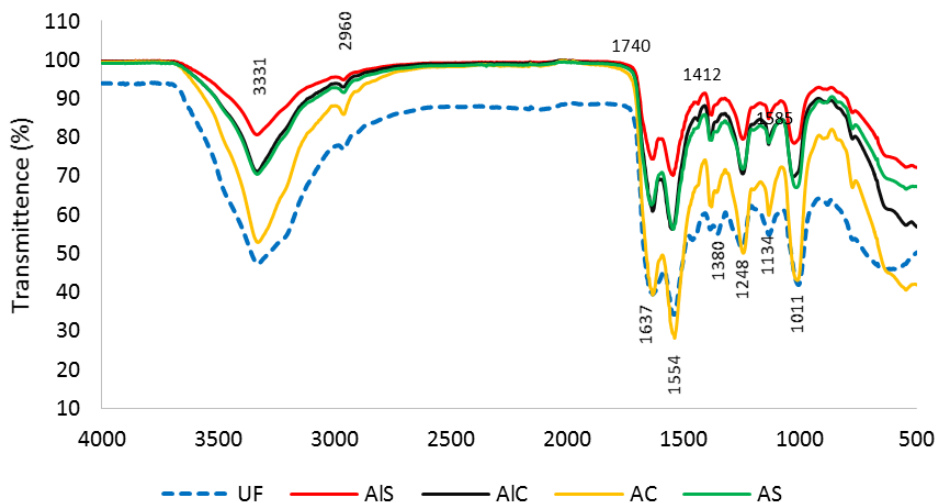
Figure 1. FT-IR spectra of urea formaldehyde (UF) and UF mixed with ammonium and aluminium based hardener

Table 3

Absorption band assignment of FT-IR spectra of UF resin (Jada 1988, Myers 1981)

Absorption (cm^{-1})	Observed Band (cm^{-1})	UF functional group
3500-3100	3331	N-H stretching mode
2960-2970	2960	CH mode of CH_2, CH_2OH and $N-CH_2$
1740-1720	1740	C=O stretch aliphatic aldehyde (formaldehyde)
1680-1630	1637	Amide I, mainly due to C=O stretching
1600-1550	1554	Amide II, mixture of C-N and N-H deformation
1400-1360	1380	C-H stretching of the CH_2OH group
1300-1260	1260	-OH, deformation of CH_2OH
1150-1130	1134	Asymmetric stretching of >N-CH_2<N
1060-970	1011	C-O stretch in methylol

the addition of hardeners had reduced the intensity of the absorption in comparison to the pure UF resin. The finding was in agreement with Puttasukkha et al. (2015) who observed a decrement in these OH groups when formaldehyde scavenger was added into the UF resin. Another weak yet distinct absorption band appears around 2960 cm^{-1} is assigned to the asymmetric -CH stretching of the –CH$_2$OH. As can be seen from the figure, frequency of this peak for ammonium- and aluminium-based hardener become less intense compared to the urea formaldehyde peak and it is probably due to the high conversion of the –CH$_2$OH in the polymer (Jada, 1988). According to Poljansek et al. (2006), the peaks at 1720 cm^{-1} to 1740 cm^{-1} are correspondent to the -CO stretching of the formaldehyde. Clearly from the band, the peak for formaldehyde was decreased after the addition of the ammonium and aluminium-hardeners. The absorption band at around 1585 cm^{-1} might be attributed to the formic acid salts (HCOO$^-$) while the band near 1248 and 1011 cm^{-1} might be attributed to hexamethylenetetramine (Vinogradoff et al., 2011).

The absorption bands at 1443 – 1487 cm^{-1} and 1097 – 1145 cm^{-1} has been identified as methylene (-CH$_2$-) bridge and methylene-ether (-CH$_2$OCH$_2$-) bridge, respectively, by Jada (1988). The intensity of the methylene bridge at around 1443 – 1487 cm^{-1} increased when the hardeners were added into the UF resin. Acid released by the hardeners accelerated the curing process of the UF resin and subsequently affected the change of methylene-ether bridges to the methylene bridges (Puttasukkha et al., 2015). On the contrary, the absorption intensity at 1101 cm^{-1}, which represents methylene ether bridges, decreased after the addition of hardeners (Wu et al., 2016). According to Dunky (1998), hydrolysis of these methylene ether bridges contributed to the increment in emittable formaldehyde from wood-based products. Garnier et al. (2002) stated that the methylene ether bridges are relatively instable and tend to rearrange themselves to methylene bridges by splitting off formaldehyde. Therefore, reduction in methylene ether bridges resulted in lesser emittance of formaldehyde. The lowest content of methylene ether bridges was recorded when aluminium sulphate was added into UF resin and had the lowest free formaldehyde correspondingly.

Physico-mechanical Properties of Particleboard

Physico-mechanical properties of particleboard fabricated from the rubberwood particles resinated with four different resin/hardener mixtures were evaluated. All of the particleboards produced in this study achieved the targeted density (650 kg/m^3) with average density ranged from 660 to 685 kg/m^3. However, a great variation of densities of 599 kg/m^3 – 778 kg/m^3 were recorded in the individual particleboard samples after cutting. Owing to this, all the properties tested in this study were adjusted by performing and analysis of covariance (ANOCOV) where the density and moisture content are selected as concomitant variables. Table 4 listed the mean values density and moisture content of particleboard.

The adjusted thickness swelling (TS) and water absorption (WA) values of the particleboard made with UF admixed with different types of hardener after 24-h soaking are tabulated in Table 5. TS values of 28.01% to 36.79% were recorded in all the produced panels while WA values ranged from 46.07% to 54.08% were obtained. Generally, the particleboard made with aluminium-based hardener had higher TS and WA compared to that of the particleboard made with ammonium-based hardener. The lowest thickness swelling value of 28.01% was recorded in the samples made with AS while the highest TS value of 36.79% was observed in the sample made with AlC. The pattern of the WA mirrored that of the TS as the highest WA was recorded in the particleboard made of AlC (54.08%) and the lowest WA was recorded in the AS samples (46.07%). It was found that all the panels produced did not meet the requirement of TS ($\leq$ 12%) as stated in standard JIS A 5908. However, the requirement is not easy to achieve as the water soaking method is more severe than the actual outdoor environment. This finding was in agreement with Ashori and Nourbaksh (2008) who reported that the thickness swelling of particleboard made with different species of wood exceeded 12% even only immersed for 2 hours.

As can be seen from the Table 5, the aluminium-based hardener shows a relatively higher value for both TS and WA value compare to particleboard made from ammonium-based hardener. A possible explanation for these results might be due to the acidity of the UF/hardener mixtures. For good adhesion between the particles, the pH should be between 4 to 5 to results better adhesive performance among the particles (Akyuz et al. 2010). The acidity test showed that aluminium-based hardener (3.0-3.2 pH) have a lower pH than ammonium-based hardener (5.5-5.7 pH) and therefore the particleboard made from it displayed inferior TS and WA. Table 6 provides the adjusted mean value for internal bonding (IB), modulus of rupture (MOR) and modulus of elasticity (MOE) for UF-bonded particleboard made with different type of hardeners. The modulus of rupture (MOR) and modulus of elasticity (MOE) of all types of particleboard produced was ranging from 11.26 to 16.05 N/mm^2 and 1498 to 1982 N/mm^2, respectively. The hardener type significantly affected the MOR and MOE. From the results obtained, only particleboards made with ammonium-based hardener met the minimum requirement (13 N/mm^2) for MOR as specified in JIS 5908:2003. The highest MOE value was recorded in the particleboard panels made with AC, followed by AS, AlS and the lowest MOE were from particleboard made with AlC.

As for internal bonding strength, the panels made with ammonium-based hardeners displayed higher internal bonding strength compared to that of panels made with aluminium-based hardeners. However, all of the particleboard produced have fulfilled the minimum requirement of IB values of 0.2 N/mm^2 according to JIS A 5908:2003. Properties of the UF resin after the addition of the hardener played an important role in influence the physical and mechanical properties of the particleboards produced. Since the UF resin admixed

with aluminium-based hardener had a shorter gelation time, it hardened very fast during pressing and consequently inhibited an even spread of resin that help promoting good particle-particle bonding. Consequently, particleboard produced from UF resin admixed with aluminium-based hardeners exhibited inferior properties. Akyuz et al. (2010) stated that the low pH (< 4) of the admixture of UF resin and hardener caused precuring of the resin before hot pressing and consequently weakened the adhesive bond. In addition, acidic condition might degrade the cured UF resin and subsequently led to reduction in strength (Uner & Olgun, 2010).

Table 4

Mean density and moisture content (MC) values of the particleboard made with different type of hardener

Type of hardener	Density (kg/m^3)	Moisture content (%)
AC	684 (60.97)	6.01 (0.31)
AS	685 (7.97)	6.08 (0.18)
AlC	660 (48.05)	5.50 (0.08)
AlS	666 (40.62)	5.96 (0.14)

Note. AC: ammonium chloride; AS: ammonium sulphate; AlC: aluminium chloride; AlS: aluminium sulphate. Numbers in the parenthesis are standard deviation.

Table 5

Adjusted thickness swelling and water absorption of the particleboard made with different type of hardener

Type of hardener	Thickness swelling (%)	Water absorption (%)
AC	28.49[a] (2.6)	46.74[a] (2.1)
AS	28.01[a] (2.9)	46.07[a] (1.5)
AlC	36.79[c] (2.5)	54.08[bc] (1.1)
AlS	29.75[b] (0.9)	50.29[b] (2.6)

Note. AC: ammonium chloride; AS: ammonium sulphate; AlC: aluminium chloride; AlS: aluminium sulphate. Numbers in the parenthesis are standard deviation. Means in a column followed by the same letter is not significantly different at p≤0.05

Formaldehyde Emission

The average formaldehyde emission (FE) from the particleboard made with different hardeners are shown in Table 7. As shown in the table, the formaldehyde emission amount released from the particleboards produced with different types of hardener ranged from 0.45 mg/L to 1.14 mg/L. Since the particleboards produced with UF type E1, all the panels were emitting formaldehyde below 1.5 mg/L and met the requirement of F** class according to JIS A 1460: 2001. Based on the findings obtained, it can be said that the FE of the particleboard were significantly affected by hardener type. The lowest formaldehyde emission was measured from the particleboard manufactured with AlS (0.45 mg/L) while the highest formaldehyde emission was observed in particleboard made with AC (1.14 mg/L). Generally, particleboard made with aluminium-based hardener emitted a relatively lower FE (0.45 mg/L to 0.55 mg/L) than ammonium-based hardener (0.7 mg/L to 1.14 mg/L). The results correlate well with the FTIR spectra as exhibited in Figure 1. This may be explained by aluminium-based hardener holds more free formaldehyde than ammonium-based hardener.

Figure 2 displays the correlation between the formaldehyde emission from the particleboard and the free formaldehyde content of the UF resin used in the fabrication of those particleboard. As shown in the figure, a strong positive correlation (R= 0.92) between formaldehyde emission and free formaldehyde content was observed, suggested that the formaldehyde emission increased along with increasing free formaldehyde content. Although aluminium-based hardener particleboard emitted a relatively lower formaldehyde

Table 6

Adjusted bending strength and internal bonding values of the particleboard made with different type of hardener

Type of hardener	Modulus of rupture (N/mm^2)	Modulus of elasticity (N/mm^2)	Internal bonding (N/mm^2)
AC	16.05[a] (1.1)	1982[a] (431)	1.28[a] (0.5)
AS	15.13[ab] (1.9)	1772[ab] (231)	1.40[a] (0.31)
AlC	11.26[c] (2.2)	1498[d] (311)	0.95[a] (0.23)
AlS	12.74[cd] (3.7)	1644[c] (204)	0.87[a] (0.26)

Note. AC: ammonium chloride; AS: ammonium sulphate; AlC: aluminium chloride; AlS: aluminium sulphate. Numbers in the parenthesis are standard deviation. Means in a column followed by the same letter is not significantly different at p≤0.05

Table 7

Formaldehyde emission of the particleboard made with different type of hardener

Hardener	Formaldehyde emission (mg/L)
Ammonium chloride	1.14[a] (0.02)
Ammonium sulphate	0.70[b] (0.03)
Aluminium chloride	0.55[c] (0.07)
Aluminium sulphate	0.45[d] (0.13)

Note. Numbers in the parenthesis are standard deviation. Means in a column followed by the same letter is not significantly different at $p \leq 0.05$

compared to ammonium-based hardener, AS is commonly used as the hardener for particleboard manufacturing (Stefka & Dunky, 2006; Xing et al., 2007). According to Markessini (1994), in most central and northern Europe countries, AS has replaced AC as hardener as AC imposed some environmental issues. As mentioned by Roffael (1993), the burning residue of UF-bonded particleboard containing AC may form dioxins compounds which is a group of polyhalogenated organic compounds that are significant environmental pollutants. On the other hand, there is no dioxins on burning residues found on UF-bonded particleboard hardened with AS. In addition, the physico-mechanical properties of the particleboard made with aluminium-based hardeners exhibited inferior properties in comparison to the ammonium-based hardeners. Therefore, AS is the most suitable hardener for the production of particleboard in this study.
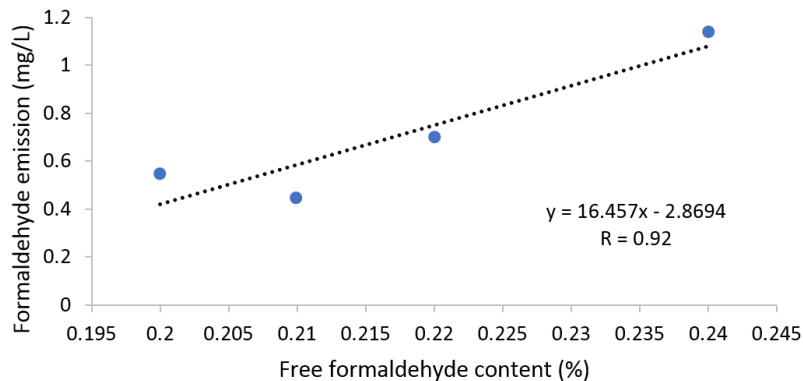

Figure 2. Correlation between formaldehyde emission and free formaldehyde content

CONCLUSION

In this study, the effects of addition of various hardeners on the viscosity, gelation time, acidity and free formaldehyde content of UF resin were evaluated. Physico-mechanical properties and formaldehyde emission level of the particleboard made from the UF/hardener mixtures were also assessed. The results revealed that both properties of UF resin and particleboard were significantly affected by the type of hardener used. Generally, aluminium-based hardeners (AlS and AlC) displayed higher efficiency in the reduction of formaldehyde emission from the particleboard. Nevertheless, physico-mechanical of the particleboard made with these hardeners were adversely affected to a greater extent in comparison to the ammonium-based hardeners. The particleboard made with aluminium-based hardeners failed to meet the minimum requirement of bending strength as stated in JIS A 5908. The reason for such phenomenon was closely related to the relatively higher viscosity, shorter gelation time and higher acidity of the UF resin after mixing that inhibit the formation of stronger bond between particles and adhesives. Based on the results obtained from the present study, it can be concluded that, among the hardeners used in this study, AS is the most suitable hardener for particleboard production. Among the two ammonium-based hardeners, AS exhibited better physical properties, internal bonding strength and lower formaldehyde emission compared to that of AC.

ACKNOWLEDGEMENT

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