



Research article

Hydrolysis of particleboard bonded with urea-formaldehyde resin for recycling

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ABSTRACT

In this study, the removal of urea–formaldehyde (UF) resin adhesives from waste wood particleboards (PBs) via hydrolysis was discussed, particularly the use of this application to combat environmental issues often encountered in recycling projects. Herein, the conditions required for producing PBs with poor binding properties were examined. Additionally, we determined the appropriate formaldehyde: urea (F/U) mole ratios, namely, 0.95, 1.05, and 1.15, required for generating UF resins that can be characterized and used as PB binders. The resulting values were compared with those obtained for a high mole ratio of UF resin (F/U = 2.0) as well as a commercially available PB sample for binding. Aqueous hydrochloric acid (HCl) solutions of various concentrations and water were used to leach the adhesive from the wood residues, and the effectiveness of these leaching agents was determined using a combined scanning electron microscopy–energy dispersive spectroscopy (SEM–EDS) technique in addition to the Kjeldahl method. Swelling tests were performed on the UF resins to measure the sol fraction (ω_{sol}) and evaluate the network behavior of the resulting resins. Our results showed that factors, such as solid content, density, viscosity, and gel time, were necessary for generating an effective adhesive; herein, we determined that a solid content between 37.17 and 56.57%, density between 1.45 and 1.54 g/cm³, viscosity ranging from 115–444 MPa.s, and gel time between 8.50 and 13.13 min were feasible. Whereas the physical properties of the resulting PB (i.e., the density and moisture content) fulfilled the criteria established by the Japanese Industrial Standard, as laid out in the document entitled JIS A 5908: Particleboards (2003), the mechanical properties failed to pass the aforementioned standard as low bending strength and weak internal bonding were noted for the PBs produced. The use of hydrolyzing agents successfully decomposed the UF resin adhesives by altering their nitrogen (N) content; confirmation of this was obtained through SEM–EDS analysis along with the Kjeldahl method. Swelling tests showed that despite containing a reasonable amount of nitrogen owing to its dissolution in either HCl or water, the ω_{sol} parameter was heavily influenced by the concentration of the hardener and type of F/U mole ratio adhesive used for the PB under investigation. These results indicate that wood residues can be used as raw materials for recycling PBs.

1. Introduction

Urea–formaldehyde (UF) resins are the most commonly used thermosetting adhesive worldwide. Their presence is integral in a variety of wood-based composite products, including plywood, particleboards (PBs), and fiberboards. Excellent bonding strength is achieved in UF resins containing formaldehyde: urea (F/U) mole ratios between 1.6 and 1.8. However, recent studies have shown that F/U molar ratios as low as

1.0–1.2 are ideal for the production of UF adhesives [1, 2] as they have a lower formaldehyde content and, therefore, lower formaldehyde emissions (FE) are obtained from the bonded products. This finding is a significant step forward in combating the issue of “sick building syndrome” as it ensures compliance with international emissions standards. On the downside, however, is the problem of weakened mechanical properties, i.e., lowered internal bond (IB) strength, and noticeably reduced dimensional stability related to the thickness swelling (TS) of the panel

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[3, 4]. As such, wastage of panel products is rampant in the industry, as noted by the overabundance of broken furniture made from wood composite products that litter garage depots worldwide.

Experts recommend the use of various recycling techniques as a means of combating this growing issue and mitigating the associated environmental impact [5]. Unfortunately, recycling fiberboard waste for use in the manufacture of new panels is an arduous task as the copious amounts of adhesives that had been used to ensure adhesion between the wood fibers must first be removed from the fiberboard waste product [6]. In other words, waste wood panels can only be rendered useful once the excess amounts of UF resins are efficiently and completely extracted. Cured UF resins are susceptible to hydrolysis; however, using this technique results in reduced water resistance and unwanted FE from the final product. Therefore, it is crucial to find an appropriate hydrolyzing agent capable of efficiently extracting the UF resin content in waste wood for wood recycling.

The hydrolysis of cured UF resins is often thought of as being the reverse of its synthesis [7, 8]. Cured UF resins are prone to such reactions mainly due to bond alterations contained within the adhesive and/or the interface caused by decomposition, swelling, or shrinking. This results in particle movement that facilitates moisture penetration and degrades the chemical composition of the UF resin, thereby decomposing the methylene linkages within the cured resins [9]. Some studies focused on the mechanism by which the hydrolysis of UF resins was achieved via the loss of bond strength and the release of formaldehyde (FE) in neat cured UF resin [10, 11, 12, 13, 14, 15, 16] and UF resin-bonded wood panels [17, 18, 19, 20]. Recently, the hydrolysis of cured UF resins using acid was undertaken to determine the micro-morphology of the sample with the aid of scanning electron microscopy (SEM) [21], transmission electron microscopy (TEM) [22], and Confocal laser scanning microscopy (CLSM) [23]. The results of this investigation indicated that the cured UF resins were composed of amorphous regions which were removed under acid etching and crystalline parts that underwent decomposition after extended reaction periods.

Although many studies on the hydrolysis of UF resin focused on FE as a marker for reaction completion, this is inherently associated with the release of urea or urea-derived species. Relatively high amounts of nitrogen-containing UF resin components were found to be extractable when fiberboard panels were exposed to water, though this was not extensively investigated for UF hydrolysis and FE [24, 25]. UF resin adhesives contained several types of chemical clusters, including macromolecules, polymer networks, and sol fractions. These sol fractions, which are unlinked polymer chains, are free within the network and, therefore, are extractable [21]. Recently, Lubis et al. [6] reported that water could also be used as the hydrolyzing agent during the fiberboard recycling process. Here, solid residues and extract solutions were shown to contain nitrogen derived from UF resin adhesives, thereby making it feasible to recycle UF resin bonded PBs using an extended water-soaking process.

Based on the aforementioned findings, this study was focused on the removal of UF resin adhesives from PB via immersion treatments in water, a strong acid (1N HCl), or a weak acid (0.1N HCl). The extent of UF resin adhesive removal was measured using an SEM-EDS system in combination with the Kjeldahl method. Here, the nitrogen content of the resin was noted as the parameter of choice for this process because UF resins have several nitrogen-containing moieties such as the mono-, di-, and tri-hydroxymethyl urea originated from addition reaction as well as methylene linkage, methylene ether linkage, and uron species derived from condensation reaction [14, 26] during production of UF resins as shown in Figure 1.

Swelling tests were also conducted to determine the molecular integrity of the UF resin adhesives. Here, the sol fraction (ω_{sol}) was measured since this parameter was free of solvent entanglement issues [27, 28].

Overall, the objective of this study was to evaluate the effectiveness of acid-and water-based hydrolysis for the removal of UF resin adhesives

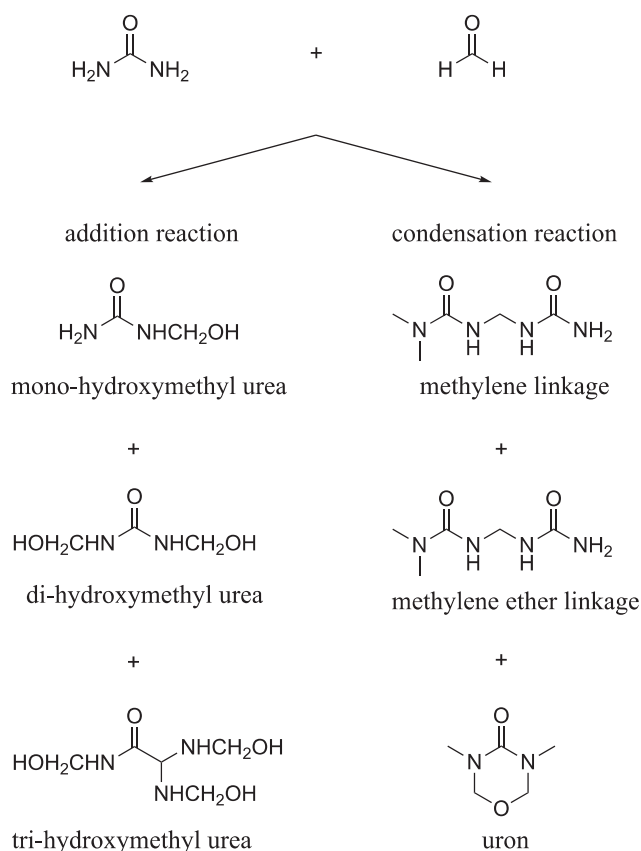


Figure 1. The formation of mono-, di-, and tri-hydroxymethyl urea species from addition reaction as well as methylene linkage, methylene ether linkage, and uron species from condensation reaction resulted in nitrogen-containing UF resins.

from PBs with the assistance of SEM-EDS analysis in combination with the Kjeldahl method.

2. Materials and methods

2.1. Synthesis and characterization of UF resin

The UF resin for this study was produced using technical grade formalin (37%) and urea granules (99%). Formic acid (20%wt) and sodium hydroxide (20%wt) were also prepared for adjusting the pH levels during and after synthesis. The synthesis of the relevant UF resin was conducted as previously reported [9]. Briefly, a traditional alkaline-acid two-step reaction was performed in which the pH of the addition and the condensation reactions was 7.8 and 4.6, respectively. The addition of the second round of urea resulted in low formaldehyde: urea (F/U) mole ratios of 0.95, 1.05, and 1.15 for the respective reactions.

Formalin was placed in the glass reactor, and the pH of the mixture was adjusted to between 7.8 and 8.0. Then, the first round of urea was added to the mixture to yield an F/U molar ratio of 2.0. Hydroxymethylation was encouraged by heating the mixture to 90 °C for 1 h. Next, the temperature was adjusted to 80 °C, and the condensation reaction was performed at pH 4.6 to promote viscosity in the mixture. Once the mixture had attained the desired level of viscosity, the second addition of urea was done to consume any excess formaldehyde and to establish the relevant F/U mole ratios of the final UF resins. After the dissolution of the added urea was completed, the UF resins were cooled to ambient temperature, and the pH was subsequently adjusted to 8.0 to terminate the reaction. For comparison studies, a UF resin with a higher

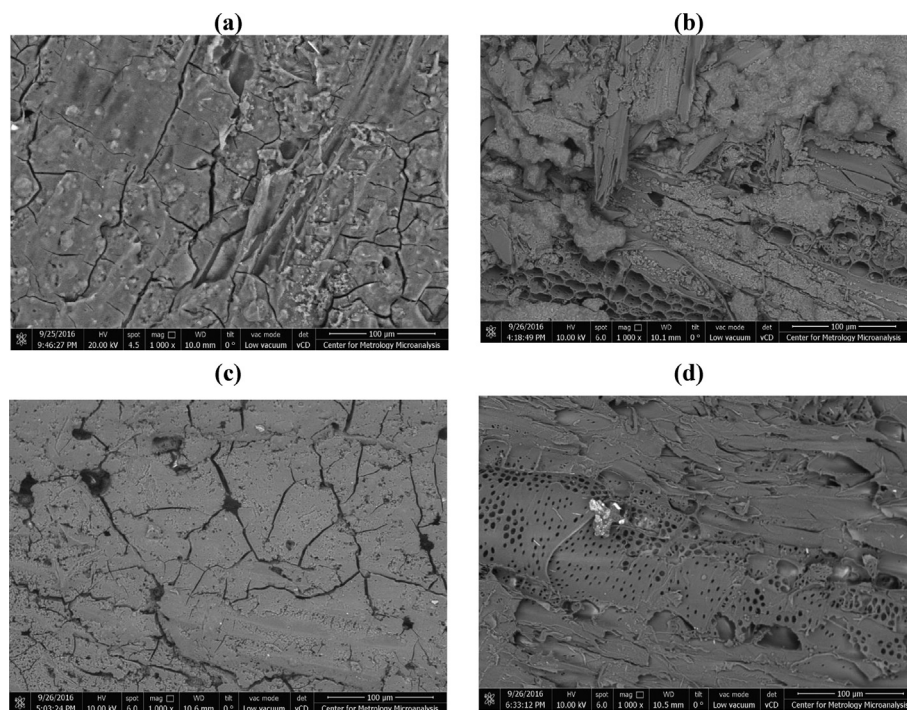


Figure 2. Typical SEM microphotographs: (a) The cured UF resins (F/U = 1.15) covered the wood particles before immersion (control). (b) The UF resin (F/U = 2.0) was leached during hydrolysis using 1 N HCl, resulting in the exposure of both the surface and the lumens of the wood. (c) The UF resin (F/U = 1.05) was hydrolyzed using 0.1 N HCl, resulting in extensive cracks on the surface of the cured UF resin. (d) The commercial UF resin was leached after a 24-h immersion period in water, resulting in exposed wood pores.

mole ratio (F/U = 2.0) was prepared, and a commercial UF resin (F/U mole ratio unknown) was purchased from PT. Pamolite Adhesive Indonesia, Probolinggo, Indonesia.

The solid content (SC), density, viscosity, and gel time were characterized for all UF resins. The non-volatile SC was determined using gravimetric analysis. Here, the UF resin adhesive (3 g) was weighed in an aluminum weighing boat and placed in a convection oven at 105 °C for 24 h. The non-volatile SC was calculated as the weight of the dry sample. The initial sample in the crucible (W1) was dried at 103 ± 2 °C for 24 h until constant weight (W2) was achieved. The SC was, thus, determined using Eq. (1):

$$SC = \frac{(W2)}{(W1)} \times 100\% \quad (1)$$

Density measurements were performed at room temperature in a pycnometer in which water was used as the non-swelling medium (Eq. (2)):

$$Density (g/cm^3) = \frac{(w_3 - w_1)}{(w_2 - w_1)} \quad (2)$$

where w_1 , w_2 , and w_3 are weights obtained from the dry pycnometer, the weight of water and the pycnometer combined, and the weight of the sample in the pycnometer, respectively. The sample's viscosity was measured using a viscometer with the appropriate spindle. Here, the UF resin adhesive sample was placed in a 200-ml beaker, and readings were taken at 25 °C using a spindle rotating at a given velocity (rotations per minute, rpm). The gel time is defined as a period required for the prepolymer liquid adhesive to solidify or cure. A resin's gel time is often reduced with the aid of a hardener or a catalyst such as ammonium chloride (NH_4Cl), particularly in the case of amino resin formaldehyde-based adhesives. In the current study, the gel time was determined by mixing the UF resin (10 g) with a 3% ammonium chloride solution (20% wt) immersed in a water bath at 100 °C.

2.2. Construction of PBs bonded by UF resins

With the relevant UF resins in hand, we then proceeded to construct the respective PBs. Here, the PB consisted of shavings obtained from

Melina wood (*Gmelina arborea* Roxb.), 3% NH_4Cl (20%wt) solution as the hardener, and 8% UF resins based on oven-dried weight wood shavings. During the manufacturing process, the temperature, compression, and time parameters of the hot-press were 130 °C, 30 kgf/cm², and 10 min, respectively. The target thickness was 10 mm, and the dimensions of the finished product were 25 cm × 25 cm. In total, five types of PBs were produced: one PB bonded using the commercial UF resin and four PBs bonded using UF resins with F/U mole ratios of 0.95, 1.05, 1.15, or 2.0. The Japanese Industrial Standard (JIS) A 5908 (2003) served as the official guideline against which the physical, mechanical, and class emission properties of the resulting PBs were evaluated [29].

Samples of the respective PBs were cut-to-pattern for the physical tests (i.e., density, moisture content, and TS) and the mechanical analysis (i.e., bending strength and IB). Dimensional stability was examined via the physical tests, whereas mechanical analysis was geared toward establishing the relevant loading parameters. For determining the bending strength, the modulus of elasticity (MOE) and the modulus of rupture (MOR) tests were conducted using a Tensilon universal testing machine with a 15-cm span; this was done via the primary method of one-point loading at a load speed of 10 mm/min at room temperature (25 °C). The FE parameters were measured for the formaldehyde-based adhesives using the desiccator method. Finally, statistical analysis was applied to each parameter using a completely random design with a single factor (i.e., the UF resin adhesives). Duncan's multiple range test (MRT) was applied to all quantitative measurements. A p value of 0.05 was considered to be statistically significant.

2.3. Hydrolysis of the PBs

For the PB testing samples, the UF resin adhesive was subjected to hydrolysis using aqueous hydrochloric acid (HCl) solutions (0.1 N and 1 N) or water. The disintegration of the bonds between the UF resin adhesives and the wood shavings was achieved by immersing the pattern-cut PB samples in the respective HCl solution or in water with mechanical stirring for 24 h. Random PB samples were withdrawn and subjected to SEM-EDS analysis after a drying period of 24 h at an ambient temperature to facilitate qualitative (Figure 2) and quantitative (i.e., the C, N, O, and Cl content per sample) analyses.

Table 1. Characteristics of the UF resins synthesized in this study.

No.	F/U mole ratio	Solid content (%)	Density (g/cm ³)	Viscosity (mPa.s)	Gel time (minutes)
1	0.95	52.65	1.54	215	13.13
2	1.05	48.64	1.48	144	11.41
3	1.15	46.99	1.47	131	9.45
4	2.00	37.17	1.45	444	4.41
5	Commercial	56.57	1.48	115	8.50

Remarks: UF resins no.1–4 were the same resins used in a previously published work [41].

Table 2. The properties of PB bonded using UF resin with various F/U mole ratios.

Property	Unit	UF resin with the respective F/U mole ratio					
		0.95	1.05	1.15	2.0	Commercial	JIS A 5908 (2003)
Density	g/cm ³	0.51 (0.02)	0.49 (0.01)	0.50 (0.02)	0.55 (0.03)	0.53 (0.03)	0.40–0.90
Moisture Content	%	7.39 ^c (0.62)	8.11 ^{ab} (0.40)	8.79 ^b (0.85)	9.54 ^a (0.23)	8.50 ^b (0.63)	<14
Thickness swelling 2 h	%	54.87 ^c	51.14 ^c	50.94 ^c	12.19 ^a	28.08 ^b	-
Thickness swelling 24 h	%	66.15 ^c (8.54)	61.07 ^c (2.26)	63.24 ^c (3.16)	19.45 ^a (2.41)	28.38 ^b (5.72)	<12
Water absorption 2 h	%	106.95 ^c	106.82 ^c	107.03 ^c	55.58 ^a	66.26 ^b	-
Water absorption 24 h	%	128.90 ^c (5.66)	129.20 ^c (4.34)	131.19 ^c (7.62)	88.02 ^a (5.34)	96.79 ^b (7.15)	-
IB	kgf/cm ²	0.31 ^a (0.27)	0.25 ^a (0.16)	0.50 ^a (0.16)	1.80 ^b (0.53)	1.30 ^b (0.72)	>1.50
MOE	kgf/cm ²	5269 ^a (2433)	3995 ^a (1562)	4283 ^a (2734)	10659 ^c (4711)	7505 ^b (1935)	>20000
MOR	kgf/cm ²	24 ^a (13)	18 ^a (5)	22 ^a (13)	70 ^c (18)	49 ^b (19)	80
Formaldehyde emissions	mg/L	0.2 ^a	0.1 ^a	0.3 ^a	0.4 ^a	1.3 ^b	F**** to F*S

Remarks: The mean values are based on five replications from five different types of PBs, whereas the values in parentheses represent the standard deviation. Values with the same letters are not statistically significant at a *p* value of 0.05.

The results of the SEM–EDS analysis were compared by applying the Kjeldahl method. Here, the nitrogen content of the composites before and after hydrolysis under various reaction conditions was examined. Briefly, the samples (0.5 g) were ground into a fine powder using a Fomak grinder passed through a 60 mesh. Next, the various sample powders were digested in a Buchi Speed digester K-425 in the presence of the catalyst and sulfuric acid (EM.100731). Titration with 0.1 N HCl (EM.100317) was conducted using a K-350 Buchi distillation apparatus by adding 3%wt boric acid (EM.100165) and three drops of methyl red bromo cresol green indicator to the respective sample mixture. Eq. (3) was used to calculate the nitrogen content of the respective samples:

$$\text{Nitrogen content (\%)} = \frac{((V_1 - V_2) \times N \times 0.0014 \times f_p)}{W} \tag{3}$$

where *V*₁ and *V*₂ are the volumes of HCl required during the sample and the blank titrations, respectively, *N* is the HCl normality, *W* is the sample weight in mg obtained using a Mettler Toledo (MS 204 TS) analytical balance with a deviation of 0.0001, and *f*_{*p*} is the dilution factor.

2.4. Swelling test

Swelling tests were conducted according to a previously reported study [21]; in this case, all parameters remained unchanged except for

Table 3. SEM–EDS analysis of PB samples showing the cured UF resin components either on the surface of the wood residues or in the wood tissue.

Property	Element	UF resin with F/U mole ratio				
		0.95	1.05	1.15	2.0	Commercial
Control (without immersion in the leaching agent)	C	39.48 (0.01)	44.01 (0.01)	43.89 (0.06)	43.32 (0.13)	49.47 (0.04)
	N	35.70 (0.02)	31.61 (0.02)	30.55 (0.08)	25.83 (0.17)	16.87 (0.10)
	O	24.18 (0.01)	23.60 (0.02)	24.56 (0.02)	25.07 (0.04)	33.18 (0.04)
	Cl	0.63 (0.01)	0.77 (0.01)	1.01 (0.01)	1.77 (0.01)	0.49 (0.01)
1 N HCl	C	44.92 (0.03)	45.97 (0.02)	55.37 (0.01)	42.57 (0.02)	47.85 (0.04)
	N	22.99 (0.02)	20.59 (0.02)	7.39 (0.01)	31.37 (0.03)	19.81 (0.04)
	O	29.38 (0.01)	31.02 (0.01)	35.37 (0.01)	21.78 (0.01)	30.00 (0.06)
	Cl	2.71 (0.01)	2.41 (0.01)	1.87 (0.01)	4.27 (0.01)	2.33 (0.01)
0.1 N HCl	C	47.18 (0.02)	37.12 (0.01)	43.58 (0.07)	47.83 (0.01)	51.43 (0.01)
	N	25.13 (0.02)	35.84 (0.02)	27.25 (0.09)	24.75 (0.09)	22.59 (0.01)
	O	27.22 (0.01)	25.88 (0.01)	28.61 (0.04)	25.09 (0.01)	25.02 (0.01)
	Cl	0.47 (0.01)	1.16 (0.01)	0.55 (0.02)	1.50 (0.01)	0.96 (0.01)
Water	C	50.31 (0.05)	54.66 (0.04)	48.55 (0.09)	62.30 (0.01)	67.62 (0.08)
	N	22.64 (0.05)	16.16 (0.07)	28.28 (0.10)	13.30 (0.01)	1.22 (0.21)
	O	26.90 (0.04)	29.18 (0.03)	23.12 (0.04)	24.37 (0.01)	30.69 (0.01)
	Cl	0.15 (0.03)	0 (0)	0.05 (0.02)	0.03 (0.02)	0.46 (0.01)

Remarks: The values represent the mean of triplicate EDS scans conducted at various locations on the SEM samples. The values in parentheses represent the standard deviation.

the temperature used in our experiment. Since the predominant UF resin in our study was a low molar resin, the ideal temperature for the swelling test was 60 °C.

3. Results and discussions

3.1. Characterization of the UF resins

The method employed for the synthesis of the UF resins, as described in previously reported studies [15, 21, 30, 31, 32, 33, 34, 35, 36], was based on a traditional two-step procedure comprised of a hydroxymethylation or addition reaction followed by condensation. Using this procedure, we realized that sample purity heavily influenced the characteristics of the UF resin along with other factors such as the amount used, the sequence in which the raw materials were added, and the type of catalysts employed (i.e., alkaline and acid catalysts). Precise control of the reaction temperatures, pH, and the concentration of the reactants was crucial. As such, the addition reaction was initiated by reaction with an alkaline catalyst, followed by acidification at the appropriate point in time to promote the condensation step [37, 38].

Table 1 shows the characteristics of the UF resins synthesized in this work. Here, we determined that a decrease in the F/U mole ratio was accompanied by an increase in the SC of the resin; this was triggered by the second addition of urea during synthesis. Moreover, we noted that lower F/U mole ratios resulted in a denser UF resin, as noted by the ω_{sol} value. In previous studies, the ω_{sol} value was relatively higher in resins that had a low F/U mole ratio than in resins with a high F/U mole ratio [21]. Additionally, the resins were noticeably more viscous when the F/U mole ratios were high, indicating increased reactivity. Resins with a low molecular weight were less viscous [39, 40] and, thus, were less reactive; the failed decomposition of urea into the respective hydroxymethyl groups exemplified this point. Also, longer gel times were noted for UF resins with a lower F/U mole ratio in contrast to those with higher F/U mole ratios.

3.2. Properties of PB bonded by UF resins

Table 2 shows the properties of UF resin-bonded PBs. Generally, the physical performance among the various PB samples was similar, as the density of the samples was below the target value of 0.75 kg/cm³. Fortunately, the average values for the density and moisture content fulfilled the requirements of the JIS standard. Thus, both UF resins with low F/U mole ratios and high F/U mole ratios (i.e., 2.0) were still within the defined density target of the JIS standard. We noted, however, that the thickness swelling and water absorption values were relatively high, which was a clear indication of dimensional instability. Therefore, using these types of PBs for manufacturing household items and furniture would be exceedingly disadvantageous since the lifetime of these products would be unnaturally short, thereby wasting the energy invested in the recycling efforts.

The mechanical properties of PBs bonded using UF resins with a low F/U mole ratio, i.e., the associated IB, MOE, and MOR characteristics, were generally lacking. By contrast, UF resins with higher F/U mole ratios (i.e., 2.0) and the commercially available resin displayed IB and MOR traits that were within the acceptable limits as defined by the JIS

standard; unfortunately, this was not the case for the MOE values. It was theorized that this poor showing was due to the molecular mobility of the cured UF resin network [42].

Except for the commercial sample, the quantity of formaldehyde emitted from all PB products in our experiment was between 0.1032 and 0.3741 mg/L, and this was within limits defined by the JIS standard. According to the guidelines, PBs with these F**** emission values were categorized as “very low.”

3.3. SEM–EDS results

Figure 2 shows images of the PB residue samples before and after immersion in aqueous HCl or water. Prior to immersion, the cured UF resins coated the wood residues (Figure 2a), indicating that extensive integration of the UF resin into the wood had occurred. However, the SEM microphotographs of the residues after acid immersion clearly show that the UF resins had been successfully leached by strong acid (Figure 2b), whereas the use of a more dilute HCl solution resulted in an extensive network of cracks with no exposed wood sections (Figure 2c). Surprisingly, the UF resin was leached by immersion in water, as shown in Figure 2d.

Table 3 shows the results of the EDS scan on SEM microphotographs. Although the values obtained seemed to vary, the general trend noted in the data was promising. SEM–EDS analysis confirmed the existence of the UF resin component in the wood residues despite the use of only superficial surface scans, and achieving the optimal sample position during analysis was heavily dependent on the experience of the operator. The scan procedure itself was uncomplicated; however, conducting such an experiment on uncoated material was risky due to possible degradation. Fortunately, differences between the wood residues and the UF resin adhesives were evident in this case since we could easily discern various structures and features such as the cracks of the cured UF adhesive, distinct wood pores, and the interface between the wood and the adhesive.

As expected, the nitrogen content of the cured UF resins in the wood residues generally decreased after immersion in the respective hydrolyzing agents. There were some fluctuations in the measurements obtained, as indicated by the standard deviations. Here, it was clear that water was a promising hydrolysis agent for future recycling purposes, a finding which was consistent with that reported by Grigsby et al. [25]. The nitrogen content values obtained using the Kjeldahl method are presented in Table 4.

Even though the values obtained for the percentage nitrogen content were smaller than those seen in the SEM–EDS analysis, one general trend noted in Table 4 was the decrease in the N content, which was indicative of successful hydrolysis of the UF resin (regardless of the F/U mole ratio) using acid or water.

The ω_{sol} values for various concentrations of hardeners/catalysts were measured for each F/U mole ratio as experimental confirmation of the hydrolytic degradation of the respective UF resins. Figure 3 presents the ω_{sol} and the number average molecular weight between cross-links (M_c) of the UF resin films with various concentrations of hardeners.

Here, the UF resin films with lower F/U mole ratios exhibited low or negative ω_{sol} values, which was indicative of incomplete cross-linking in the UF resins during the curing process [31, 43]. These UF resins were

Table 4. The nitrogen content (%) as measured using the Kjeldahl method.

Property	UF resin with F/U mole ratio				
	0.95	1.05	1.15	2.0	Commercial
Control (without immersion in the leaching agent)	1.38	1.14	1.42	1.26	1.50
1 N HCl	0.97	1.02	0.84	0.84	1.48
0.1 N HCl	0.70	1.19	1.26	1.09	1.38
Water	1.15	1.01	1.59	1.21	1.32

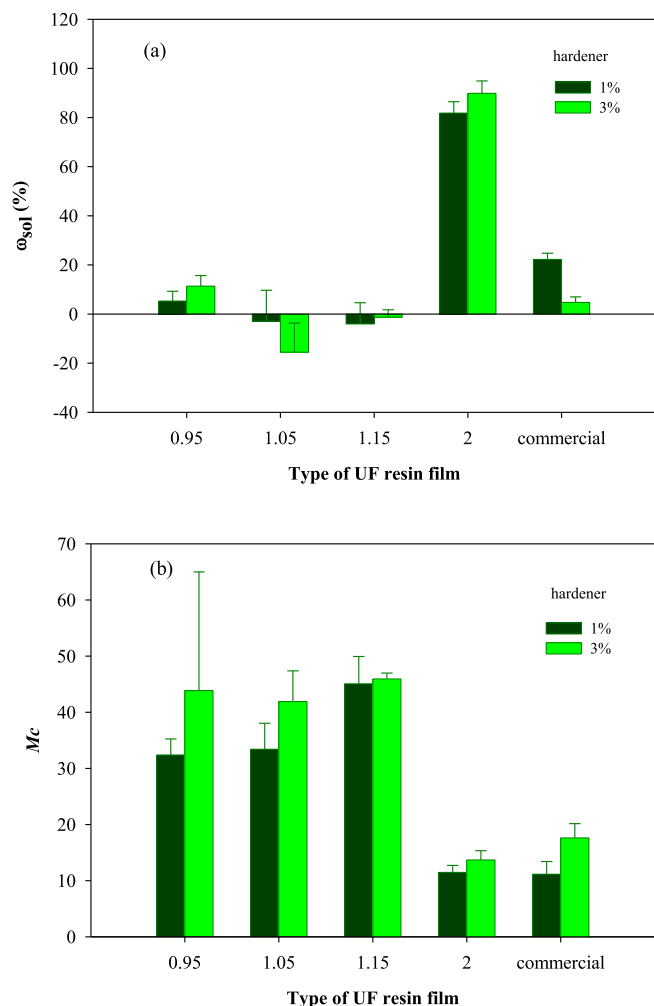


Figure 3. The ω_{sol} (a) and the M_c (b) of the cured UF resin films with various concentrations of hardeners.

partially dissolved upon immersion, resulting in only a small amount of resin available for surface coatings and, thus, a small ω_{sol} value. On the other hand, both the high F/U mole ratio resins and the commercially available product exhibited high sol fraction values due to entanglement in the UF resin network. The M_c value obtained for low F/U mole ratio resins was comparatively higher than their high F/U mole ratio counterparts, as reported in previously published studies [21]. Moreover, the concentration of the hardener was also a deciding factor as the addition of more concentrated hardener solutions led to higher M_c values.

In summary, we were able to establish the factors that exerted influence on the wood residues of recycled PBs using SEM–EDS analysis. Here, factors such as the nitrogen content of the sample within a given timeframe, the ω_{sol} values, and the M_c values all contributed to our understanding of the structure of the cured UF resin network as an adhesive system in wood residues.

4. Conclusions

The factors influencing the hydrolytic degradation of UF resin adhesives in PBs were quantitatively established using SEM–EDS analysis of uncoated samples. Here, we found that both low and high F/U mole ratio resins trapped in waste wood PBs degraded when subjected to water immersion or acid etching processes. However, the extent of degradation was dependent on multiple internal and external factors. The existing sample conditions, including the ω_{sol} content and the molecular integrity of the cured UF resins, which was characterized by the M_c value, were all

classified as internal factors, whereas external influences were primarily due to the positioning of the sample during analysis and were heavily dependent on the experience of the operator. Herein, it was shown that SEM–EDS analysis was useful for identifying the component chemistry of wood adhesives and determining the feasibility of using wood residues as raw materials for the manufacture of PBs.

Declarations

Author contribution statement

Arif Nuryawan: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Rahmawaty: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data.

Kartini D. S. Tambun: Performed the experiments; Contributed reagents, materials, analysis tools or data.

Iwan Risnasari: Conceived and designed the experiments; Performed the experiments.

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The authors declare no conflict of interest.

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