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Impacts of urea-formaldehyde resin residue on recycling and

reconstitution of wood-based panels

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Abstract

Recycling and reuse of wood-based panels has been attracting great attention since it is an environmentally friendly means of managing wood waste. The recycled wood-based panels usually still contain adhesive residue. In the current research, the impact of urea-formaldehyde (UF) resin, one of the most widely used adhesives, on recycling and reconstitution of wood-based products has been evaluated by adding cured UF resin particles into a new adhesive system. The morphology, chemical structure, curing behavior, and bonding quality of an adhesive system containing 0-6% previously cured resin particles of varied sizes (< 600 μm) were investigated and compared to those properties of a regular adhesive without any impurity. Three-ply plywood glued with a UF adhesive containing 0-10 wt% previously cured resin of varied particle sizes (< 600 μm) were prepared and characterized. The results indicated that the resin residue could significantly affect the curing behavior and bonding quality of the new adhesive system. Higher content and greater size of the particles caused more severe deterioration in the bonding strength.

Keywords: Urea–formaldehyde resin, wood-based panel, recycling, bonding quality **1. Introduction**

Currently, China produces and consumes the highest amount of wood-based panels in the world. In 2014, the production of wood based-panels was 302 million cubic meters in China and this number will probably increase in the future. Every year, roughly 10% of the

panels from old wooden construction, furniture, and packaging[1] after completion of their service life need to be discarded or recycled [2]. The above wastes are mainly manufactured from adhesive glued particleboards, fiberboards and plywood containing roughly 92% wood and 8% resin.

Three waste management techniques are applied for used wood-based panels including land filling, energy production, recycling and reconstitution[3]. While land filling and direct combustion result in a negative impact on the environment, pyrolysis for energy production [4] and the reconstitution of used panels into new products are relatively environmentally benign. Recently, value added materials, such as activated carbon, wood plastic composites and cellulose nanocrystals, have been produced from recycled wood-based panels [5-7]. Several methods have been developed for reconstituting particleboard (PB) and medium density fiberboard (MDF) into new panels using hydrothermal[3, 8, 9], chemical[10], mechanical[10, 11] methods or their combination[12]. The recycled materials can be bonded with urea-formaldehyde (UF) resin or phenol-formaldehyde resin (PF) using conventional techniques for manufacturing wood-based panels.

Recycling and reconstitution of used wood-based panels into new ones is a complicated process requiring an understanding of the interaction of recycled wood materials containing cured resin residues and the new resin system. Compared to other adhesives, UF resins are most widely used for wood-based panels such as plywood, PB and MDF in China and worldwide due to its low price, good bonding performance and colorless [13]. It is a polymeric condensation product of the chemical reaction of formaldehyde with urea. Current research work on UF resins has focused mainly on its curing behavior [14, 15], formaldehyde emission[16, 17], crystallinity and domain size [18]. Different U/F ratios, curing agents, and other additives all have impact on the bonding strength of UF resins [13, 19-21]. UF resin residues, although probably in a disfunctional form, still unavoidably remain in most of the recycled wood-based materials even after hydrothermal, chemical or mechanical treatment due to their broad applications and relative stability[19]. To reconstitute the used wood-based panels into new products, new adhesive is expected to be applied to the recycled wood-based materials. Up until now, no systematic research has

been reported to explore the impact of resin residue from the recycled panels on the behavior of the new adhesive system.

In this work, cured UF resin was ground to fine particles and added to a new UF resin system to mimic the recycling and reconstitution process of wood-based panels. The impact of UF resin residue on the new adhesive system was studied with emphasis on the cured resin particle size and content. The morphology, chemistry, curing behavior, and bonding strength of the adhesive system containing previously cured resin particles were investigated and compared to the regular adhesive without any cured resin impurity.

2. Materials and methods

2.1 Materials

All chemicals were analytical grade and used as received. Aqueous solutions of sodium hydroxide (NaOH, 20%) and ammonium chloride (NH4Cl, 20%) were used to adjust the pH level during the UF resin preparation process. Plywood was made of eucalyptus veneers of 1.7 mm thickness and 9% moisture content.

2.2 Preparation of UF resin samples

An aqueous solution of formalin (37%) and urea were mixed with an F/U mole ratio of 2.1 in a reactor and the pH level was adjusted to 7.5 by adding several drops of 20% NaOH solution. The mixture was heated to 90 °C and left to stand for 40 minutes using an oil bath. Subsequently, the pH of the solution was brought down to 6.2 using 20% NH4Cl and maintained at this level until a white cloudy gel-like substance was observed in the mixture. The pH of the mixture was brought up to 7.2 and the F/U mole ratio was adjusted to 1.8 by adding the appropriate amount of urea. After reaction for another 40 minutes, the mixture was pH adjusted to 8.0 and a third batch of urea was added to obtain a UF resin prepolymer F/U ratio of 1.4. The resin suspension was cooled to room temperature and stored for further use.

The properties of the UF resin prepolymer suspension was tested according to ASTM D1081, resulting in a viscosity of 33 s, curing time of 75 s, pH of 7.0, solid content of 53%, and free formaldehyde emission of 0.21%.

A predetermined amount of the UF resin prepolymer mixture was poured into a Petri dish and NH₄Cl aqueous solution of final concentration of 0.2% was added as a hardener.

The UF resin was kept in an oven (DHG-9123A, Shanghai, China) at 60 °C for 24h to make sure it was fully cured. The final thickness of the cured resins was from 2 mm to 4 mm.

The cured UF resin was ground to fine particles with a grinder (HC-1600Y2, Huangcheng, China, 20, 000 rpm, 3 min) and sieved to three classes by passing through 40#, 100#, and 200# meshes sequentially (Class I: passing through 200#, Class II: 100-200#, Class III: 40-100#). Particles larger than 40# were discarded.

The three classes of UF resin particles obtained from the grinding process were added to the freshly prepared UF resin prepolymer system at 2 wt%, 4 wt% and 6 wt%, respectively, and mixed thoroughly. Curing of UF resins containing previously cured resin particles was performed as mentioned above using 0.2% NH₄Cl as the hardener. The second-time cured UF resin was sectioned for morphology observation or ground to a fine powder and sieved as mentioned above. Particles passing through 100# but not 200# mesh (Class II) were again added to the UF resin prepolymer system at 6 wt% to study their impact on the system. The new system was cured again as above and will be mentioned thereafter as the third-time cured system.

2.3 Analytical methods

2.3.1 Morphology of UF resins

The morphology of the ground UF resin particles was studied by an optical microscope (Leica DMIL, Germany) and an environmental scanning electron microscope (XL-30-ESEM, Holland FEI, resolution 2.5 nm). The surface morphology of the second-time cured UF adhesive system containing 6% resin particles was also examined by SEM. The resin samples were sectioned to 5 mm × 5 mm × 2 mm and adhered onto SEM substrates. All SEM samples were sputter coated with gold and imaged by the environmental scanning electron microscope operating at 2.5 kV. To obtain the size distribution of the resin particles, about 150 – 200 particles were randomly selected for each class to measure their longest diameters.

2.3.2 FTIR analysis

The chemical structure of the UF resins was characterized by Fourier transform infrared spectroscopy (FTIR) (PerkinElmer, USA). Each sample was pressed into pellet with anhydrous

KBr powder at 1:99, w/w resin: KBr ratio, before measurement. The IR spectrometer was operated over 4000-400 cm⁻¹ at a resolution of 4 cm⁻¹ under transmission mode.

2.3.3 DSC analysis

To understand the curing behavior of the UF resins, their thermal properties were analyzed by differential scanning calorimeter (DSC 200F3 NETZSCH, Germany). Approximately 10 mg of prepolymer samples with 6% Class III particles and without any particles were added to 40 μL aluminum crucibles, respectively. High-pressure sealed capsules, which can withstand 3 MPa vapor pressure, were used to prevent the sample components from evaporating at high temperature. The samples were heated from 20°C to 220 °C at the heating rate of 10 °C/min. Nitrogen flow was 50 mL/min.

2.4 Plywood preparation and testing

Three-ply laboratory plywood panels were made of eucalyptus veneers. Freshly prepared UF resins with and without previously cured UF resin particles were applied on one side of the veneer at 300 $\rm g/m^2$. NH₄Cl of 1 wt% was added as a hardener and the plywood was assembled and hot pressed under 1.5 MPa pressure at 120 °C for 15 min.

The bonding quality of the plywood was determined by shear test according to British Standard BS EN 314-1:2004. The plywood was cut into test pieces of 100 mm in length and 25 mm in width, with shear area length of 25 mm. The shear strength was calculated according to the following equation:

$$
\sigma_{\mathbf{b}} = \frac{F_{\mathbf{m}\ \mathbf{a}}}{l \times b} \tag{1}
$$

where F_{max} is the failing force of the sample; *l* is the length of the shear area, 25 mm; *b* is the width of the shear area, 25 mm. The dip peeling property of the plywood was measured by immersing samples of 75 mm × 75 mm dimensions into hot water at 63 °C for 3 hours following by drying in an oven at 63 °C for 3 hours. The peeling ratio was obtained by calculating the total length of the peeling place divided by the perimeter of the plywood. Four specimens were used for each test.

2.5 Statistic analysis

Single factor analysis of variance was used to determine the impact of the cured resin particle size or its addition, while other factor was fixed at certain level. Two-factor analysis

of variance was used to determine the impact of resin particle size, content and their interaction. In two-factor analysis of variance, the amount of particles included only three test levels (2%, 4% and 6%). The significant difference of the data was evaluated using the student test (with p<0.05 confident interval). All the statistical analyses were performed by Excel 2007. All the variations shown in this work are standard deviations.

3. Results and discussion

3.1 Morphology of UF resins

For the cured and powdered UF resins, the particles were all irregular polygons (Fig. 1-3). Particle sizes of all three classes showed nearly normal distribution (Fig 1c, Fig 2b, Fig 3b.). The Class I particles (passing through 200# mesh) were all <50 μm with an average at 35.5±17.6 μm. The Class II particles (100-200# mesh) were 50-150 μm wide with an average at 107±33 μm. The Class III particles (40-100# mesh) had widths of 150-600 μm averaged at 439±122 μm.

Fig. 1. Class I resin particles passing through 200# mesh: a) Light microscope image (200×), b) SEM image (800×), c) particle size distribution

Fig. 2. Class II resin particles passing through 100# but not 200# mesh: a) Light microscope image (200×), b) particle size distribution

Fig. 3. Class III resin particles passing through 40# but not 100# mesh: a) Light microscope image (200×), b) particle size distribution

A cross section of the cured UF resins is shown in Fig. 4. Compared to the smooth surface of the regular UF adhesive system without previously cured UF particles (Fig. 4a), the second-time cured UF systems exhibited inhomogeneous surfaces containing tiny grains (Fig. 4b-d). For the second-time cured UF system with 6% Class I particles, the size of most grains (<50 um) was close to that of the Class I particles, indicating that the previously cured UF particles were not able to form a continuous network with the new resin during the second curing process. The fact that a small amount of grains with size greater than 50 μm were also found may indicate aggregation of the previously cured particles. The density of the grains was roughly 60-100 particles per square mm. For the second-time cured UF system with 6% Class II particles, the diameter of the grains was roughly in the range from 100 μ m to 150 μ m, consistent with the size of the Class II particles. The density of the grains decreased to 10-15 particles per square mm. In the system containing 6% Class III particles, the diameter of the grains increased to 200-500 μm, again consistent with the size of the Class III particles. The grains were found to be less than 1-2 particles/mm².

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Fig. 4. SEM images of the cured UF resin cross section: a) UF resin without previously cured particles. b) second-time cured UF resin with 6% Class I particles. c) second-time cured UF resin with 6% Class II particles. d) second-time cured UF resin with 6% Class III particles.

From the results above, it can be concluded that the previously cured UF particles were not able to fuse with the new UF resin. UF resins are thermosets and the previously formed chemical bonds in the particles were hard to break and reform during the second curing process. Compared with Class II and III particles in the resin, Class I particles had the sharpest boundary with the resin, indicating the weakest bonding. Also, aggregation of Class I particles observed by SEM imaging may probably be due to the non-thorough mixing of the fine powder into the resin prepolymer suspension.

3.2 Chemical structure analysis of UF resins

The chemical structure of UF resin is very complicated depending on the urea concentration, pH value, and F/U molar ratio. Samples prepared under neutral or basic

conditions are mainly composed of methylolurea and/or dimethylene ether bridges between urea units[22]. The FTIR spectra of the first-time cured UF resins exhibited characteristic functional groups of typical conventional cured UF resins [15](Fig. 5a): a strong and broad NH stretching at around 3100-3500 cm $^{-1}$, OH and CH stretching peaks at 3120 and 2970 cm $^{-1}$, respectively, as well as amide I and II at 1650–1550 cm⁻¹ and CH₂OH, CH₃ and CN at 1400– 1360 cm^{-1} , etc. The assignment of all the peaks is listed in Table 1 according to previous research work. The second-time cured UF resins containing 6% Class I, II, III resin particles, respectively, also showed common cured UF resin peaks (Fig. 5b, c, d). The IR spectrum of third-time cured UF resin with 6% second-time cured Class II UF resin particles was also almost identical to the first-time cured UF resin (Fig. 5e). Compared to the first-time cured UF resin, the C-O stretching at around 1000-1040 cm^{-1} of the second-time and third-time resins seemed to shift to a higher wavenumber. Since this C-O stretching peak came from an ether (at 1060-1065 cm⁻¹) plus a methylol structure (at 1000-1005 cm⁻¹)[23], it is possible that the second-time and third-time cured resins contained more ether structure due to the repeating curing processes. All these results indicated that the addition of previously cured resins had little impact on the chemical structure of the entire adhesive system. Neither the particle size nor the recycling times had any obvious effect.

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Fig. 5. IR spectra of cured UF resins: a) UF resin without previously cured particles. b) second-time cured UF resin with 6% Class I particles. c) second-time cured UF resin with 6% Class II particles. d) second-time cured UF resin with 6% Class III particles. e) third-time cured UF resin with 6% Class II second-time cured resin particles.

| Wavenumbers cm^{-1}) | Assignment | Reference |
|--------------------------------|---|-----------|
| 3359 | $v(NH)$ in NH ₂ | [23, 24] |
| 3128 | v(OH) of N-CH ₂ OH | $[24]$ |
| 2977 | $v(CH)$ of CH ₂ of ether, CH ₂ OH and N-CH ₂ | [22, 23] |
| 1640 | $v(C=O)$ in $-CONH2(amide I)$ | [14, 23] |
| 1552 | mixture of CN and NH (amide II) | [13, 22] |
| 1378 | v (CH) in CH ₂ | [14, 24] |
| 1246 | Amide $I\!I\!I$ | [14, 23] |
| 1108 | $v_{\text{as}}(N=CH_2-N)$ | [22, 23] |
| 1005 | $v(CO)$ of ether and $-CH2OH$ | $[23]$ |
| 832 | $v(NH)$ of NH ₂ | $[25]$ |
| 754 | $v(CN)$ of $-NH-CH_2-NH-$ or $v(NH)$ | [23, 25] |

Table 1. Assignment of IR peaks in cured UF resins.

3.3 Thermal analysis of UF resins

Fig. 6 presents heat flow as a function of temperature for UF resins with and without previously cured resin particles. For the original UF resin, a small endothermic peak was observed at 80 °C with a reaction enthalpy of 1.89 J/g probably due to the glass transition of the prepolymer. A large exothermic peak started at 130 °C indicated curing of the resin. The maximum change of heat flow occurred at 135 °C and then decreased. The reaction enthalpy was 225 J/g.

The heat flow curve of the UF resin containing previously cured resin particles did not show any endothermic peaks below 100 °C. An exothermic peak was observed starting from 83 °C and with a maximum at 109 °C and reaction enthalpy of 15.7 J/g. A large exothermic peak attributed to the curing of the resins started at 130 °C. The maximum of the peak was found at 144 °C and its reaction enthalpy was 398 J/g. While the temperature rose to more than 200 °C, an exothermic peak occurred probably due to the degradation of the resins from their methylene-ether bridges.

Fig. 6: DSC thermograms of UF prepolymers with or without previously cured resin particles.

By comparing the DSC thermograms of the UF resins with and without previously cured resin particles, it is concluded that the addition of previously cured resin resulted in more exothermic thermal incidents and an increase of reaction enthalpy. Condensation reactions may occur between the previously cured UF resin particles and the prepolymers leading to

more exothermic incidents as also indicated in the FTIR study. Also, thermal degradation of the UF resins may start at a lower temperature for the resins containing previously cured particles.

3.4 Properties of plywood glued with UF resins

Plywood glued with UF resin containing particles with different amounts and sizes were prepared and tested in order to further understand the impact of previously cured resin particles on the bonding quality of UF resins. The shear strength and dip peeling ratio of plywood containing 0-10% Class I UF resin particles are shown in Fig. 7. The shear strength of the plywood using original UF resin was 2.37 MPa and decreased slightly to 2.31 MPa with 2% resin particles and to 2.23 MPa with 4% particles. When the particle content increased to 6%, a statistically significant reduction in plywood shear strength (2.06 MPa) was observed. A further increase in resin particle content to 10% of the total glue resulted in a substantial decrease of the shear strength of the plywood to 1.33 MPa, which was 44% lower than that using the original resin. With every 2% increase in resin particle content, the drop of shear strength increment increased, indicating disruption of the intact resin network. Using UF resin without any impurity, less than 5% of the gel peeled after dipping and drying. With more resin particles, the dip peeling ratio of the plywood increased from 4.5% for the control sample to 47.5% for the sample containing 10% resin particles. A jump of peeling ratio from 17.8% to 47.5% was observed when the resin particle content changed from 8 % to 10 %. Single factor analysis of variance confirmed that the amount of the particles incorporating into the system had a significant negative impact on the shear strength and peeling ratio of the plywood.

Fig. 7. a) Shear strength and b) dip peeling ratio of plywood containing 0-10% Class I previously cured UF resin.

The shear strength of plywood containing up to 6% resin particles of all three classes are shown in Fig. 8a. No intact plywood board could be obtained when greater than 6% Class III particles were incorporated. Incorporation of more resin particles from the same size class resulted in a decrease of plywood shear strength as observed in plywood containing Class I particles. However, with the same weight percentage of resin particles, plywood containing Class III particles showed the lowest bonding strength. For the extreme example, plywood glued with UF resin containing 6% Class III particles exhibited the lowest bonding strength of 1.24 MPa, which was 48% lower than that of sample using the original resin, followed by plywood glued with UF resin containing 6% Class II particles of 1.89 MPa, 20% lower than the control, and plywood glued with UF resin containing Class I particles of 2.06 MPa, 13% lower than the control. Single factor analysis of variance confirmed that the particle size had a significant impact on the shear strength of the plywood. The reason why larger particles caused a more severe decrease in shear strength was thought to be their larger individual surface area resulting in a discontinuous resin network.

The dip peeling property of plywood containing 0-6% resin particles of all three classes was also tested and compared (Fig. 8b). The peeling ratio of the plywood was affected by the amount of particles incorporated. For plywood containing 0-6% Class III particles, the peeling ratio increased from 4.5% to 41.0%. While incorporation of 2% Class III particles resulted in a peeling ratio of 11.8%, incorporation of 4% particles had a significant effect on the dip peeling ratio of 17.0%, which was almost three times higher than the control. A similar trend was observed for plywood containing Class II resin particles. The peeling ratios of plywood with 2%, 4%, 6% Class II particles were 7.5%, 6.8%, and 18.5%, respectively. By comparing plywood containing different sizes but same particle content, single factor analysis of variance again confirmed that the particle size had a significant impact on the peeling ratio of the plywood and larger resin particles had a more severe effect.

From the results above, we conclude that both particle size and resin residue content had an impact on the bonding quality of the UF adhesive. A higher addition of resin residue and a larger particle size resulted in more severe deterioration of bonding quality. In addition, it seemed likely that for plywood containing a certain class of resin particle, the bonding quality decreased substantially when the particle content exceeded a critical point. UF resin may not be able to form an intact 3D network above this point. However, a relatively lower amount and smaller size of resin particles did not have a significant negative impact on the bonding quality of the new adhesive system. A two-factor analysis of variance was applied to understand the interaction between particle size and content. As shown in

Tables 2 and 3, p-values of 0.056 and 0.055 for shear strength and peeling ratio respectively, were close to the critical level of 0.05, which indicated that there may be a weak interaction between particle size and content i.e. adding larger resin particles to the system resulted in a faster drop of bonding quality.

Table 2. ANOVO of shear strength

** Highly significant difference (p<0.01)

Table 3. ANOVA of peeling ratio

Total 5.148 35

** Highly significant difference (p<0.01)

4. Conclusions

The cured urea-formaldehyde (UF) resins were ground to a fine powder and added to a new resin system in order to simulate the recycling procedure of wood-based panels containing an adhesive residue. The impact of size and content of the cured resin particles on the properties of the new resin system was investigated in this study. SEM imaging showed that the previously cured resin particles could not form integrated networks with the new resin system during the second curing process. The presence of the cured resin particles altered the thermal properties of the system and resulted in more endothermic thermal incidents during the curing process. Repeating curing of the UF resin did not lead to obvious chemical structure change, but more ether bond formation according to the FTIR analysis. The shear strength and dip peeling properties of the plywood were weakened when glued with UF resins containing previously cured resin particles. The content and size of the particles had a significant negative impact on the properties of the plywood. The

higher content and greater size of the particles resulted in more severe damage to the bonding strength and peeling property of the plywood. In the worst case tested, 3-ply plywood glued with UF adhesive containing 6 wt% previously cured resin of varied particle sizes (< 600 μm) exhibited up to a 48% decrease of shear strength and 811% increase of dip peeling ratio compared to that with the regular adhesive system. This work, for the first time, demonstrates that the size and content of resin residue could have a significant negative impact on the properties of reconstructed wood-based products.

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