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경화제 타입이 TMP 개질된 UF 수지 및 이를 대나무 파우더와 혼합한 복합재료의 경화 거동과 물리적 성질에 미치는 영향

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Effects of Hardener Type on Cure Behavior and Physical Properties of TMP Modified UF Resin and Its Composites with Bamboo Powder

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Abstract: In this study, effects of mixed hardeners (ammonium chloride mixed with ammonium persulfate, oxalate and phosphoric acid) on the properties of 2,4,6-trimethylolphenate modified UF resin (TMPUF) were studied, and the pot-life, gel time and curing temperature of TMPUF/hardeners were characterized. The results indicated that the hardeners used have critical influence on the pH value of TMPUF and some proton donor can be used to lower the pH value of TMPUF with low formaldehyde content. Besides, bamboo particleboards were made with TMPUF/hardeners, and tensile properties, water absorption ratio and the swelling ratio of bamboo particleboards were characterized. The study demonstrated that the tensile strength of bamboo particleboards increased when partial ammonium chloride was replaced by ammonium persulfate to cure the TMPUF adhesives in our experiment.

Keywords: mixed hardener, UF resin, bamboo particleboard, tensile strength.

Introduction

In the past several decades, urea formaldehyde resins (UF) have been widely used as adhesives for interior-grade wood-based panels because of their good properties including low cost, fast curing, colorless and excellent adhesion to wood.[1-6] Nevertheless, their main drawbacks are low water resistance and emission of formaldehyde from the wood panels.[7-10] Nowadays, companies are compelled to manufacture low formaldehyde release panels due to the increasingly stringent environmental requirements in the world.[11,12]

Until now, many strategies have been attempted to reduce the formaldehyde content of UF resins. For example, reducing the F/U mole ratio is one of effective and practical methods to decrease formaldehyde emission at the expense of deteriorated mechanical properties of particleboard panels.[13,14] Addition of formaldehyde scavenging agents such as melamine,[15-17] tannin[18] ethyl cellulose microcapsules[19] and phenol[20-22] is another way that commonly used to reduce formaldehyde emission or improve water resistance of UF resins.

UF resin with low formaldehyde is helpful to produce particleboards with low formaldehyde emission. However, UF resins with low formaldehyde content may impair the action of a conventional hardener like ammonium chloride and be unable to generate sufficient acid for resins to cure.[23] Decreasing pH prior to application is not feasible. For one thing, it reduces the pot life of the UF resin. For another thing, the hot pressure applied would destroy the early formed cross-linked structures.[23] Until now, scarce information can be found in literature about alternative hardeners for UF resins. Costa *et al.*[23] reported that the phosphoric acid can be taken as hardener for UF resin to avoid producing hexamine. Nevertheless, the pot

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life of UF resin is greatly shortened. Recently, we have prepared 2,4,6-trimethylolphenate modified UF resins (TMPUF) with low formaldehyde content, and we have tried to use some protonic acids instead of partial ammonium chloride to improve curing properties of the TMPUF with low formaldehyde content and studied the effects of mixed hardeners on the adhesion properties of TMPUF in the production of bamboo particleboards.

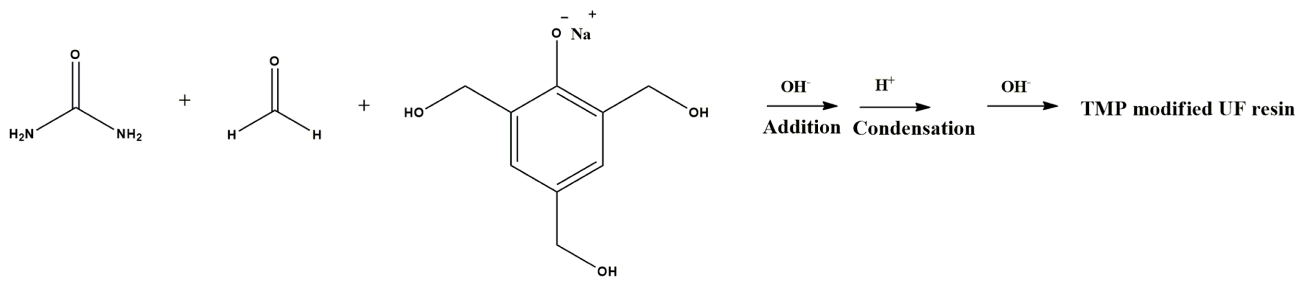
In this work, effects of four mixed curing hardeners on the properties of TMPUF resin are compared with each other. Besides, tensile properties, fracture morphology and water absorption ratio of bamboo particleboards bonded with TMPUF/hardeners were investigated by tensile tests, SEM and water absorption analysis, respectively.

Experimental

Materials. Analytical grade urea (≥ 99%) and formalin (37%) were used for the synthesis of UF resins. Formalin was titrated by sodium sulfite method[1] before use. 2,4,6-trimethylolphenate (TMP) was prepared according to the reference.[24,25] Aqueous solutions of both formic acid (20% mass fraction) and sodium hydroxide (20% mass fraction) were used to adjust the pH level during the UF resin synthesis. Compositions of four multi-component curing agents were list in Table 2. Paraffin wax (melt point 60-62 °C) was used to improve the water resistance of all bamboo particleboards.

Preparation of 2,4,6-Trimethylolphenate Modified Urea Formaldehyde Resin (TMPUF). TMPUF was prepared according to the reference as shown in the Scheme 1.[26] The formalin was placed in the 250 mL three neck flask with a stirring rate (200±10 rpm) and then adjusted pH to 8.0 with aqueous NaOH and then heated up to 70 °C. Subsequently, a certain amount urea (U1) and TMP were introduced into the flask (the mole ratio of F/U1 is 2.00 and the mole ratio of TMP/U is 1/20, U is represents the total mole of urea). Then the pH was regulating to 8.0 with formic acid and the mixture was heated to up to 90 °C under reflux for 1 h to allow for methylolation reactions. The second stage of UF resin synthesis consisted of the condensation of the methylolureas. The acidic reaction was brought by adding formic acid to obtain a pH value of about 4.5, and the condensation reactions were carried out until it reached cloud point, indicating that high molecular weight UF was formed. Then the second batch of urea (U2) was placed in the flask (the mole ratio of F/(U1+U2) is 1.45) and the pH was simultaneously adjusted to 6.5. The reaction was maintained for 30 min at 90 °C. Finally, aqueous NaOH was added to adjust the pH value of the final UF resin to 8.0 and the third urea (U3) was added (the mole ratio of F/(U1+U2+U3) is 1.20) and the temperature was cooled to 75 °C and held for another 30 min. The obtained resin was cooled to room temperature. The formaldehyde content of the prepared TMPUF was less than 0.01% (wt%), which is determined by sodium sulfite method at low temperature.[1]

Manufacturing of Bamboo Particleboard. The dimensions and density of bamboo particleboard panels were selected as 10 cm×10 cm×3 mm and 900±10 kg/m^3, respectively. Bamboo powder was blended with TMPUF, paraffin and ammonium chloride. In order to make the very fine-grained powder bind well, more adhesives were used in the experiment compared with the common bamboo particleboards preparation. The mass ratio of TMPUF solid resin to bamboo powder (over dried) was 25/75. The amount of paraffin and multi-component hardeners were 0.5% and 1 wt% (20 wt% water solution) for the solid resin, respectively. Each batch of sample was pressed under a temperature of 165 °C and the pressure of 8.0 MPa for 10 min. Four panels were manufactured for each TMPUF resins. The tensile strength, elongation at break, fracture morphology, water absorption ratio and swelling ratio of bamboo particleboards were char-



(TMP)

Scheme 1. Preparation of TMP modified UF resins.

acterized.

Properties of TMPUF Mixed with Hardeners. The pH variation of TMPUF mixed with hardeners was measured at room temperature by using pH meter (pHS-25, Shanghai Leici Instrument Factory, China); The viscosity of TMPUF resins mixed with hardeners at room temperature was measured using rheometer (DV-III Brookfield, US); The gel time was measured at 100 ºC by adding hardeners (1.2% (wt%) of the solid hardeners considered for the mass of the TMPUF resin).

DSC Analysis. Differential scanning calorimetry was made with a Perkin-Elmer Diamond DSC type calorimeter at a heating rate of 7 ºC/min over a temperature range from 50 to 150 ºC under N_2 atmosphere, and the curing temperature was recorded. The TMPUF resin used for test was mixed with hardeners (1.2 wt% of the solid hardeners considered for the mass of the resin). The mass of samples was about 5-10 mg.

IR Spectroscopy. A FTIR spectrophotometer (Bruker, TENSOR 27, Germany) was used. KBr tabletting method was taken to characterize the cured TMPUF resin mixed with different hardeners. TMPUF/hardeners were cured at 100 ºC for 10 min.

Tensile Properties. Tensile tests were conducted using a universal testing machine (SANS-CMT4104, Meitesi Industry System (china) Co., Ltd.). The specimens were prepared with 10 mm width, 70 mm length and 3 mm thickness.

SEM Analysis. The fracture surface morphology of the bamboo particleboards were studied by SEM (Hitachi-S3400N, Japan). The fracture surfaces of bamboo particleboards were sputtered with gold and then photographed.

Water Absorption. Water absorption was measured by immersing bamboo particleboards (40 mm×40 mm×3 mm) in de-ionized water at room temperature, calculating the relative weight change for 24 h. The bamboo particleboards were withdrawn from water and weighed after shaking off any free water at different time. Swelling ratio of bamboo particleboards was measured by the volume increase ratio of the bamboo particleboards immersed in water for 24 h compared with the original sample. The sample size was measured by vernier caliper with the accuracy of 0.02 mm. Each value is the average of three replications.

Results and Discussion

pH Variation of TMPUF Resin Mixed with Different Hardeners. Four hardeners were used to cure the prepared TMPUF resin and the compositions of these four hardeners were list in Table 1. Formally, we have studied the effects of TMPUF resins on the tensile properties of TMPUF/ammonium chloride impregnated filter papers.[26] The results showed that when the formaldehyde content of the prepared TMPUF resin (TMP/U=1/40 and 1/20) is very low (<0.01%), ammonium chloride is not able to make the resin cure effectively which caused the reduction of tensile strength of filter papers impregnated with TMPUF/ammonium chloride. Some hardeners that can donate proton are taken to solve the problem.[23] In the experiment, we have chosen ammonium persulfate, oxalate and phosphoric acid as proton donor which are compounded with ammonium chloride and urea, respectively. In order to illustrate the effect of these four multi-component hardeners on the cure behavior of TMPUF resin, pH variation of the TMPUF resins (mixed with hardeners) with time was tracked and the results are shown in Figure 1. It can be seen from Figure 1 that pH decreases as time goes on after the TMPUF resins mixed with CS-1. This was caused by the reaction between the free formaldehyde and ammonium chloride which makes the TMPUF resins acidic. However, the final pH value of the TMPUF mixed with CS-1 was around 7, while the pH value of common UF resin/NH_4Cl system is around 5,[10,27] indicating that the formaldehyde content of prepared TMPUF resin is low. We can also observe from Figure 1 that TMPUF mixed with hardeners that contain proton donor exhibit a low pH value, indicating that proper amount proton donor can make sufficient acid for TMPUF curing. Besides, under the condition of the same mass concentration, the pH value of oxalate is lower than that of phosphoric acid at room temperature. This is due to the higher first ionization equilibrium constant

Table 1. Compositions of the Curing System

Composition of the curing system	NH_4Cl (wt%)	$(NH_2)_2CO$ (wt%)	$(NH_4)_2S_2O_8$ (wt%)	$(COOH)_2$ (wt%)	H_3PO_4 (wt%)
CS-1	20	1	0	0	0
CS-2	15	1	5	0	0
CS-3	15	1	4	1	0
CS-4	15	1	4	0	1

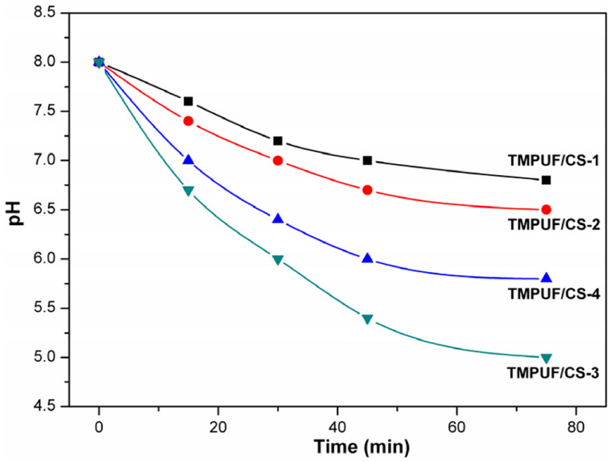


Figure 1. pH variation of TMPUF resin mixed with different hardeners.

of oxalate.

Evaluation of Pot-life. The pot-life of the UF resin/hardeners is one of the most important indicators in the application of UF resin. Figure 2 shows the evolution of the TMPUF resin viscosity containing hardeners at room temperature. Variation of the viscosity of TMPUF/hardeners system during storage is a discontinuous process and we used dash line in the figure. As it can be seen, the viscosity of TMPUF resin mixed with CS-1 increased slowly compared with other three hardeners, indicating that the pot-life of the TMPUF resin/CS-1 is long. What's more, the lower the pH values of the TMPUF resin/hardeners, the shorter the pot-life of the TMPUF resin/hardeners. It means that small amount of proton donor can immediately acidify the UF resin inducing the cure. Nevertheless, the short pot-life of the TMPUF resin containing hardeners

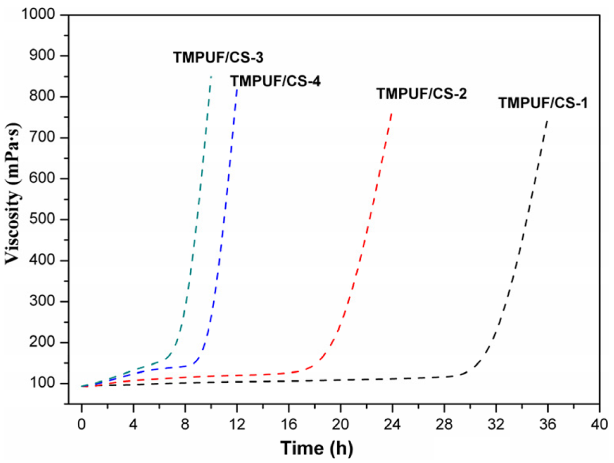


Figure 2. Pot-life of TMPUF resin with different hardeners.

with proton donor is a limitation to all the acid hardeners as mentioned in the reference, and Costa *et al.* suggested that encapsulation of the acid hardeners may overcome the problem.[23] By contrast, TMPUF/CS-2 has a suitable pot-life (about 20 h) shown in Figure 2.

Evaluation of Gel Time. Gel time was often used as an indicator of UF resin reactivity. In our experiment, the gel time of TMPUF/hardeners was test under the condition of boiling water bath (round 100 °C). The results of gel time measurement of the TMPUF resin mixed with the studied four hardeners were shown in Figure 3. As well known, the gel time of TMPUF resin decreased with the decreasing of pH value.[28] However, the gel time of TMPUF/CS-4 is shorter than that of TMPUF/CS-3, while the pH value of CS-3 is higher than that of CS-4 at room temperature as shown in Table 2. In order to explain the phenomenon, we have tested the pH value of CS-3 and CS-4 at different temperatures when they were diluted 100 times, respectively. An interesting variation of the pH value was seen in Table 2. At higher temperature, the pH value of CS-4 diluted is lower than that of CS-3 diluted. This might be explained by the fact that the first ionization of phosphoric acid is stronger than that of oxalate at higher temperature.

Curing Temperature of TMPUF Resin Mixed with Different Hardeners. In order to study the effect of hardener

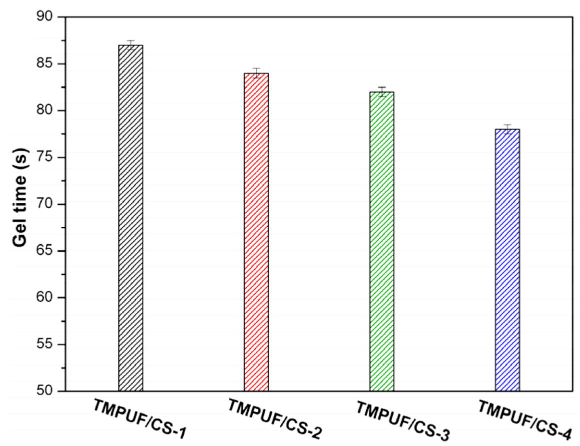


Figure 3. Gel time of TMPUF resin with different hardener.

Table 2. pH Value of the CS-3 and CS-4 at Different Temperature with Dilution of 100 Times

Acid \ Temperature (°C)	28	50	80
CS-3 diluted	4.11	3.50	2.61
CS-4 diluted	4.41	3.47	2.55

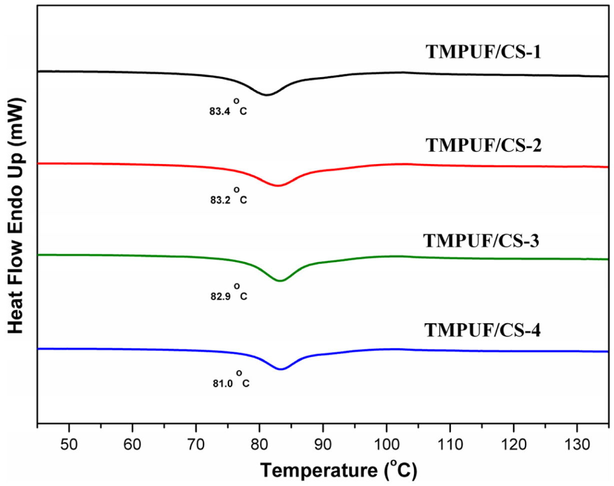


Figure 4. DSC thermograms during cure of TMPUF with different cure systems.

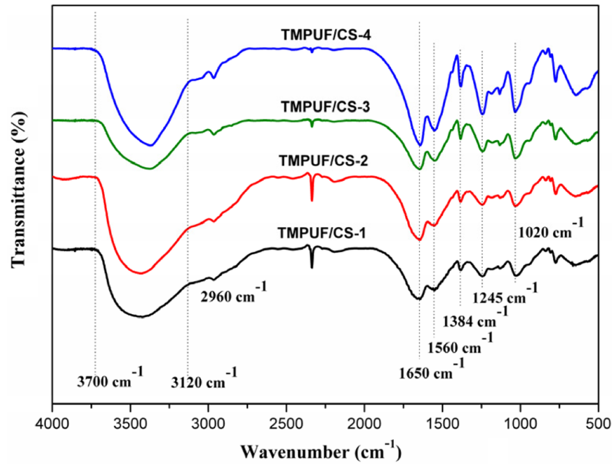


Figure 5. IR of the cured TMPUF resin with different curing hardeners.

type on the curing temperature of TMPUF resin, DSC has been used to characterize the curing temperature of TMPUF with different hardeners and the results are shown in Figure 4 and cure temperatures obtained from DSC analysis were marked in the Figure 4. It can be seen from Figure 4 that the curing temperature of TMPUF resin exhibits a little decreasing from 83.4 to 83.2 °C when the persulfate is used instead part of ammonium chloride. What's more, when oxalate and phosphoric acid are adopted instead part of ammonium chloride and persulfate, the curing temperature of TMPUF resin is drop to 82.9 and 81.0 °C respectively, indicating an increase in the resin curing reactivity. We can also note from Figure 4 that the curing temperature of TMPUF/CS-4 is lower than that of TMPUF/CS-3, because the pH value of CS-4 is lower than that of CS-3 at higher temperature (around 80 °C for DSC test and around 100 °C for gel time test). The results were consistent with the results of gel time.

IR Analysis of TMPUF Resins Cured with Different Hardeners. IR spectra of TMPUF cured with different hardeners at 100 °C for 10 min are shown in Figure 5 and the spectra were normalized using the 1650 cm^{-1} band intensity as the reference. All FTIR spectra exhibit the characteristic stretching vibration peaks of O-H and N-H between 3700 and 3120 cm^{-1}, stretching peaks of C-H at 2960 cm^{-1}, stretching vibration of amide carbonyl (C=O) at 1650 cm^{-1}, mechanical coupling of C-N stretching vibration and N-H deformation modes at 1560 cm^{-1}, bending vibration of CH_2 at 1384 cm^{-1}, stretching vibration of C-N and stretching vibration of C-O at 1245 and 1020 cm^{-1}, respectively.[29,30] The intensity of both stretching vibration and bending vibration of CH_2 at 2960 and 1384 cm^{-1} is increasing with the decreasing of pH value from TMPUF/CS-1 to TMPUF/CS-4. This may be due to that the reactivity of the TMPUF resin increases with the decreasing of the pH value from CS-1 to CS-4, and the content of CH_2 increased accordingly. Therefore, IR can also be used to explain the increased reactivity of TMPUF with the decreased pH value.

Tensile Properties of Bamboo Particleboards Made with TMPUF/Hardeners. The hardener type greatly affected the tensile properties of particleboards as mentioned in the reference.[23] Here, the results for the tensile strength and elongation at break of bamboo particleboards bonded with TMPUF/hardeners are shown Figure 6. From Figure 6, it is observed that both tensile strength and elongation of bamboo

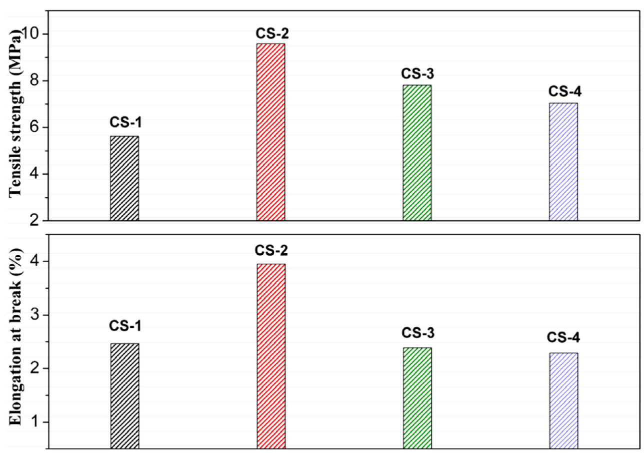


Figure 6. Tensile properties of bamboo particleboards made with TMPUF/hardeners.

particleboards increases when part of ammonium chloride is substituted by persulfate. It indicates that making the curing condition of TMPUF resin more acidic by persulfate can make the TMPUF in bamboo particleboards cure effectively. However, it is obviously that the tensile strength of bamboo particleboards made with TMPUF/CS-3 and TMPUF/C-4 is lower than that made with TMPUF/CS-2, implying that further reducing the pH value of TMPUF may impair the tensile strength of bamboo particleboards. This should be ascribed to the reason that reducing the pH value of TMPUF/hardeners may increase the formation rate of $–CH_2$- by condensation reaction between $–CH_2OH$ and –NH at room temperature, causing the reduction of $–CH_2OH$ in the resin that can react with hydroxyl groups of the cellulose of bamboo. On the other hand, the pressure applied would destroy the early formed cross-linked structures as mentioned by Costa *et al.*.[23] From Figure 1 and Figure 6, we maintain that the pH value range of TMPUF/hardener 6.0-7.0 is desirable for making the bamboo particleboards bonded with TMPUF/hardeners exhibit excellent tensile properties.

Fracture Morphology Analysis of Bamboo Particleboards Made with TMPUF/Hardeners. The morphology of tensile fracture surface of bamboo particleboards bonded with different hardeners was characterized by SEM and the micrographs are shown in Figure 7. It can be seen form Figure 7 that many cavities exit in the rupture surface of bamboo particleboards bonded with TMPUF/CS-1, while the rupture surface of bamboo particleboards made with TMPUF/CS-2 exhibits a relative compact morphology and little cavities, which means that those bamboo particleboards bonded with TMPUF/CS-2 have higher interfacial adhesion and exhibit better tensile strength shown in Figure 6. Besides, the structure of rupture surface of bamboo particleboards bonded with TMPUF/CS-3 and TMPUF/CS-4 is less compact compared with that of bamboo particleboards made with TMPUF/CS-2. This may be due to the reduction of bond strength of TMPUF adhesive caused by the lower pH value of TMPUF/CS-3 and TMPUF/CS-4.

Water Absorption of Bamboo Particleboards Made with TMPUF/Hardeners. Cellulose is hydrophilic because it contains numerous hydroxyl groups, which generally leads to a poor water resistance of bamboo products. Generally, the water absorption ratio of the bamboo fiber is high (above 50%). Here, the water absorption ratio and swelling ratio of bamboo particleboards made with TMPUF/hardeners have been characterized and the results are shown in Figure 8. It can be seen from Figure 8 that all the water absorption of bamboo particleboard bonded with TMPUF/hardeners are increased quickly during the initial 4 h and then it is almost leveled-off. In addition, hardeners have no apparent influence on the water absorption ratio of bamboo particleboards bonded with TMPUF/hardeners. However, compared with the water absorption ratio of bamboo particleboards made with TMPUF/CS-2, the water absorption ratio of bamboo particleboards made with TMPUF/CS-4 showed a slight increase, which may be ascribed to more un-compacted structure and cavities exited in bamboo particleboards made with TMPUF/CS-4. This is in consistent with the result of tensile strength analysis and morphology analysis. What's more, we can note from Figure 8 that all the swelling ratio of bamboo particleboards (24 h) is around

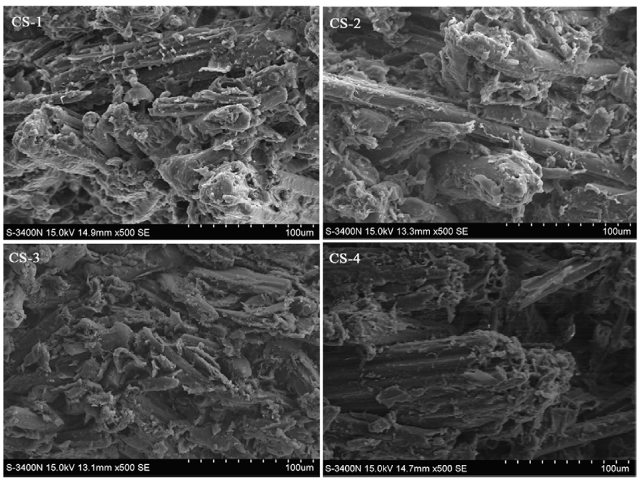


Figure 7. Morphology of the fracture surface of bamboo particleboards made with TMPUF/hardeners.

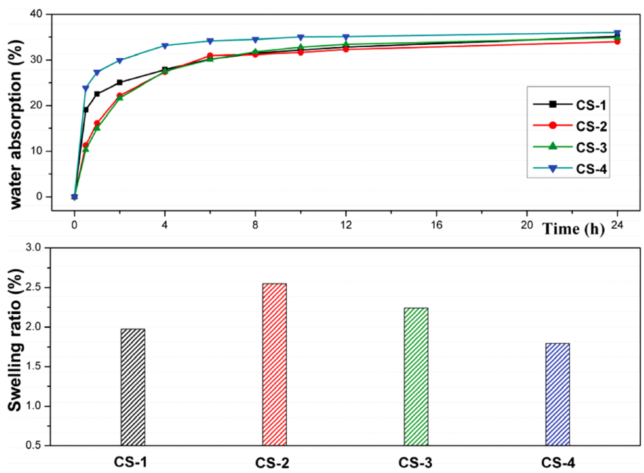


Figure 8. Water absorption ratio and swelling ratio of bamboo particleboards made with TMPUF/hardeners.

2.25%. According to these results, it can be concluded that hardeners studied in the experiment had no significant influence on water absorption ratio and swelling ratio of bamboo particleboards.

Conclusions

This work provides three alternative hardeners to ammonium chloride for curing TMPUF resin with low formaldehyde content. When using ammonium persulfate, oxalate or phosphoric acid instead of partial ammonium chloride, the pot-life, gel time and curing temperature of TMPUF/hardeners shortened and reduced accordingly. Besides, the intensity of stretching vibration -CH_2 shown in IR spectra increased as the pH value of TMPUF/hardeners reduced. What's more, tensile strength of bamboo particleboards made with TMPUF/hardeners increased when the part of ammonium chloride was replaced by ammonium persulfate to cure the TMPUF adhesives. However, further reducing the pH value of TMPUF/hardeners by oxalate or phosphoric acid may impair the tensile strength of the particleboards. On the other hand, hardeners studied in the experiment had no significant influence on water absorption ratio and swelling ratio of bamboo particleboards. The study presented suggested that the pH value range 6.0-7.0 is desirable for making the UF resin with low formaldehyde content cure effectively and bamboo particle boards exhibit excellent tensile properties.

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