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Foaming performance and bonding strength of a novel urea-formaldehyde foaming resin facilely prepared with thermo-expandable microspheres

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ABSTRACT

A novel urea-formaldehyde (UF) foaming resin to be used in low-density wood-based materials was facilely fabricated by mixing thermo-expandable microspheres (TEMs) with UF resin. The foaming performance and bonding strength of the new resin were investigated with specially designed paper/UF samples and bonded veneers joints, respectively. The developed resin had a good expansion effect, with an apparent density of less than 300 kg/m^3 and expansion ratios of 4.5–9.8, and a uniform cellular structure inside the resin was demonstrated by scanning electron microscope observations. However, the addition of TEMs resulted in a 47.5%–91.4% decrease in the shear strength of the resins. As the concentration of TEMs increased from 2.5% to 10.0%, the apparent density and shear strength showed similar decreasing trends. The introduction of glutaraldehyde (GA) increased the bonding strength of the resins to 69.8%–199.7%, while significantly increasing their apparent density. Increasing the concentration of TEMs would be an effective way to reduce the apparent density of the UF foaming resins, but there will be an accompanying degradation in their strength. GA could be a potential chemical modifier for TEMs/UF resins in terms of improving their bonding strength.

1. Introduction

Urea-formaldehyde (UF) resin and its derivatives, as one of the most important amino resins, have been used as adhesives with a long history of use in wood-based materials, and these resins are still the primary adhesive used in the wood panel industry, owing to their competitive price, high reactivity and good performance [1,2]. Medium density fiberboard (MDF) and particleboard are the most important wood products that use UF resins. Lowering the density of these products has obvious advantages in terms of material savings and transport costs. Additionally, fiberboards or particleboards with ultra-low density have particular sound and thermal insulation properties [3,4]. However, the relatively high density ($>$1000 kg/m^3) of the UF resins, which account for approximately 10% of the mass of MDF and particleboard, greatly limit the possibility of obtaining ideal “light-weight” products. Therefore, developing UF foaming resins with low density and adequate bonding performance will help overcome this problem.

UF foaming resins are usually prepared by adding low-boiling point alkanes (such as hexane [5], pentane [6,7]) as blowing agents. When the heating temperature exceeds the boiling point of the blowing agents, they boil to form cellular structures inside the resin. Owing to the complex factors affecting the vaporization process, the controllability of the foaming effect of the resin is relatively poor. Thermally expandable microspheres (TEMs) are special physical foaming agents with a core-shell structure formed by encapsulating blowing agents (low-boiling point alkanes) with thermoplastic polymers, such as polyacrylonitrile (PAN), poly(methyl methacrylate) (PMMA) and polyurethane (PU) [8–10]. When the heating temperature exceeds the glass transition temperature (T_g) of the thermoplastic shell, the shell softens, and the alkanes rapidly vaporize to form a hollow structure. The expansion of TEMs under heating is illustrated in Fig. 1. With the advantages of a high closed-cell ratio, a uniform pore size and high controllability of the foaming ratio [11,12], TEMs are widely applied in fields such as plastics [13], coatings [14], adhesives [15] and printing [16], and have great potential to be used as a new blowing agent for UF foaming resins.

Furthermore, high brittleness and low strength are the main drawbacks of UF foams, and these drawbacks are mainly attributed to the lack of flexible functional groups in the UF molecules [17,18]. With an increase in the amount of blowing agents added, the compressive strength of a UF foam decreases further, and the pulverization rate increases [5]. To increase the added value of UF foams, toughening units including

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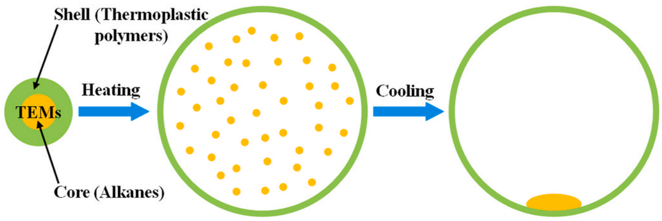

Fig. 1. Schematic diagram of TEMs expansion.

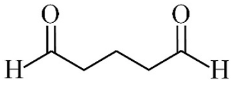

Fig. 2. Chemical structure of glutaraldehyde.

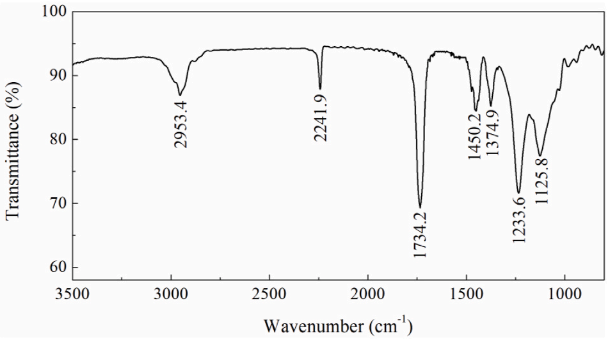

Fig. 3. FTIR spectra of the TEMs.

Table 1
Composition of the UF resins.

No.	Powdered UF (g)	Water (g)	TEMs (g)	GA (g)
1	11.00	9.00	0	0
2	10.75	8.25	0	1.00
3	10.50	7.50	0	2.00
4	10.50	9.00	0.50	0
5	10.00	9.00	1.00	0
6	9.50	9.00	1.50	0
7	9.00	9.00	2.00	0
8	9.75	8.25	1.00	1.00
9	8.75	8.25	2.00	1.00
10	9.50	7.50	1.00	2.00
11	8.50	7.50	2.00	2.00

polyether amine [7], graphene [19] and aramid fiber [6], have been introduced into the resin matrix, resulting in significant increases in the compressive strength and modulus as well as an obvious decrease in the pulverization rate. Additionally, polyethylene glycol (PEG) has been reported to significantly increase the toughness and impact strength and decrease the pulverization rate of phenol-urea-formaldehyde foam [20]. Poly vinyl alcohol (PVA) has also been used to enhance the toughness of unfoamed UF resin [21]. These approaches fall into two categories: physical filling and chemical modification. The later has attracted extensive attention for its notable toughening effect [22].

In this work, a novel UF foaming resin was facilely prepared by mixing TEMs with UF resin. The foaming performance (including the pore structure and apparent density) and bonding strength of the new resin were measured with specially designed paper/UF samples and bonded joints of veneers, respectively. Paper was selected as the substitute for the veneers to evaluate the foaming performance because their cellulose composition is similar to wood and because it is easy to form continuous, flat resin films on paper. The effect of the TEM concentration on the properties of the new resin was further investigated. Moreover, glutaraldehyde (GA, Fig. 2) was used as a new chemical modifier to improve the bonding strength of the UF foaming resin.

2. Materials and methods

2.1. Materials

A4 papers with a density of 80 g/m^2 were purchased from Artool Paper Co., Ltd. (Guangzhou) and were cut into sample pieces with a size of 110 mm × 60 mm. For each sample piece, a rectangular wireframe (80 mm × 30 mm) was printed in the middle of the paper to mark the gluing area. These paper pieces were collected into plastic ziplock bags with silica gel desiccant for at least 3 days.

Pinus massoniana veneers with a thickness of 2.0 mm were placed into a humid chamber (20 °C, 65% relative humidity) for one week to obtain a moisture content of nearly 12%, and then the samples were cut into veneers with a size of 50 mm × 25 mm (longitudinal × transverse). The average density of the veneers was measured to be 525.2 ± 41.9 kg/m^3. On the tight side of each sample veneer, a transverse straight line was drawn 20 mm from one end of the veneer to mark the 25 mm × 20 mm bonding area.

A powdered UF (Prefere 4405) was purchased from Dynea Co., Ltd. (Shanghai). Ammonium chloride (analytical reagent) and GA (biochemical reagent, 25 wt%) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Guangzhou). An embedding epoxy resin (SPI Chem™ Low Viscosity "Spurr" Kits) was purchased from Electron Microscopy China (Beijing).

TEMs (PG 12) with an expansion temperature range of 90–120 °C were provided by Beijing Hujin Technology Co., Ltd. The Fourier transform infrared (FTIR) spectra of the TEMs were recorded using a Thermo Scientific Nicolet iS5 (Thermo Fisher Scientific, USA) spectrophotometer. The samples were mixed with KBr powders, and were subsequently pressed to form a transparent tablet. The wavelength range varied from 4000 to 400 cm^{-1}. As shown in Fig. 3, the absorption bands at 2953.4 cm^{-1} (CH_2–CH_2 stretching vibrations), 1450.2 cm^{-1} (–CH_2 bending vibrations) and 1374.9 cm^{-1} (–CH_3 bending vibrations) are the apparent characteristic absorption peaks of the core alkane materials, while the absorption bands at 2241.9 cm^{-1} (–CN stretching vibrations), 1734.2 cm^{-1} ($C{=}O$ stretching vibrations), and between 1125.8 and 1233.6 cm^{-1} (C–O stretching vibrations) indicate that the TEM shells were copolymers of acrylonitrile and acrylate esters [23,24].

2.2. Preparation of the UF resins

The composition of the UF resins is given in Table 1. For all resins, the total weight and solid content were maintained at 20 g and 55%, respectively, and 1% (of the weight of the resins) ammonium chloride was added as a curing agent. Powdered UF and ultra-pure water were evenly mixed to prepare neat UF resin (No.1), and its viscosity, pH and curing time were 350 mPa s, 8.4 and 80 s, respectively. GA was added to the neat UF resin to obtain modified UF resins (No.2 and No.3). TEMs at concentrations of 2.5%, 5.0%, 7.5%, and 10.0% (of the final weight of the resin) were added to the neat resin to obtain UF foaming resins (No.4, No.5, No.6, and No.7). GA and TEMs were added to the neat resin to prepare modified UF foaming resins (No.8, No.9, No.10, and No.11).

2.3. Foaming performance test

TEMs (0.5 g) were placed in an oven (120 °C) for 5 min to acquire expanded TEMs. Optical micrographs of the control and expanded TEMs were obtained by an Eclipse 80i microscope (Nikon, Japan). The diameters of TEMs from the two experimental groups were measured using NIS-Elements D software, and more than 200 samples were measured for each experiment group.

The sample preparation process for the foaming performance test is

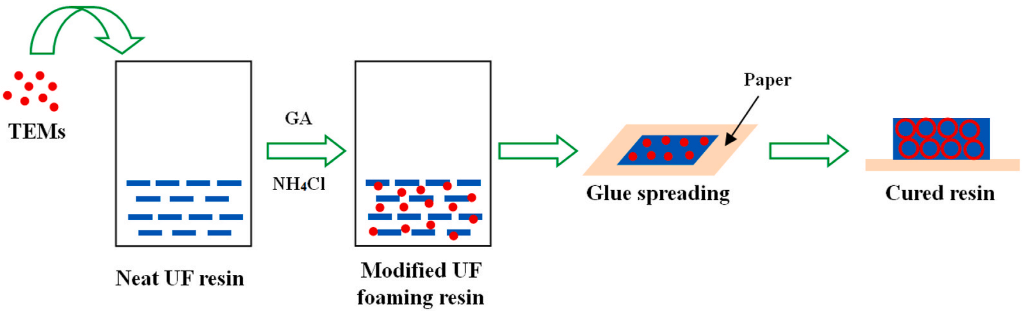

Fig. 4. Schematic diagram of the preparation of the modified UF foaming resin for the foaming performance test.

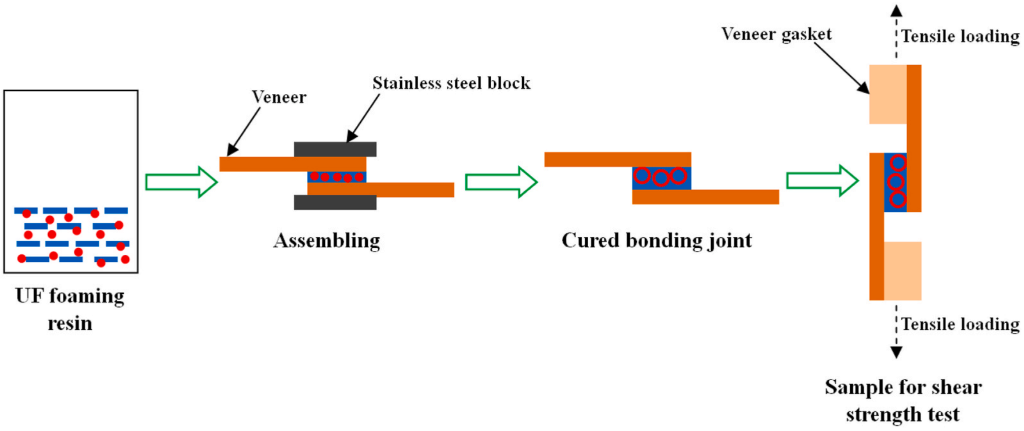

Fig. 5. Schematic diagram of the preparation of the samples for the bonding strength test.

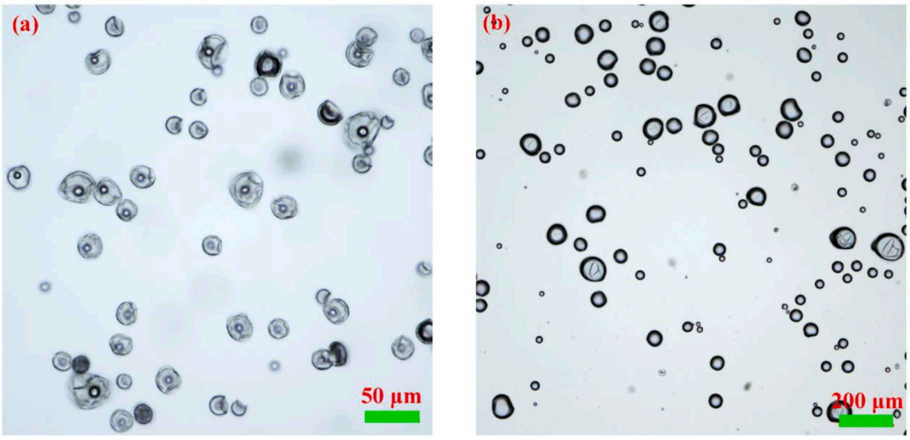

Fig. 6. Optical micrographs of the control (a) and expanded (b) TEMs.

shown in Fig. 4, using the modified UF foaming resin as an example. The resin was prepared as mentioned above, and an oil paint brush was used to paint the resin on the paper at a glue spread of 140 g/m^2. The painted papers were immediately placed into an oven (120 °C) to cure for 5 min, and the cured resin samples were placed in the atmosphere for 2 days. The resin samples were carefully cut into 20 mm × 20 mm pieces and subsequently placed into plastic ziplock bags with silica gel desiccant for at least 3 days. Six cured resin samples replicates were prepared for each experimental group, and three sample pieces were obtained for each cured resin sample. Neat papers were used as control samples.

The apparent density of the UF resins was measured according to ISO 845: 2006 “Cellular plastics and rubbers - Determination of apparent density” with some slight modification. As it was difficult to detach the UF resins from the sample pieces, the weight and volume of the resins were indirectly calculated based on the results from the sample pieces and control papers, rather than from direct measurements as required in the standard. The weight of the sample pieces was recorded. Their length and width were measured by a vernier caliper, and their thickness was measured with a 0.01 mm μm. The apparent density and expansion ratio of the UF resins were calculated using formula 1 and formula 2, respectively.

$$\text{Apparent density} = (m - l \cdot w \cdot d) / (l \cdot w \cdot t_1 - l \cdot w \cdot t_2) \tag{1}$$

where m is the weight of sample pieces; l and w are the length and width

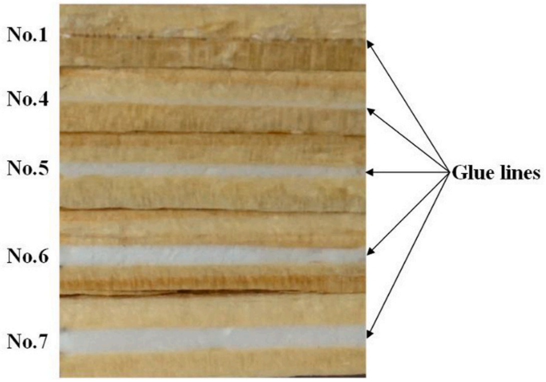

Fig. 7. Appearance of the glue lines on the bonded joints.

of sample pieces, respectively; d is the areal density of the neat papers, which was 79.4 g/m^2; t_1 is the thickness of the sample pieces; t_2 is the thickness of the neat papers, measured to be 0.096 mm.

$$\text{Expansion ratio} = \rho_1 / \rho_2 \tag{2}$$

where ρ_1 and ρ_2 are the apparent density of the neat UF resin and foamed resin, respectively.

Sample pieces from groups No.1, No.2, No.5, No.7, and No.9 were embedded using epoxy resin and were carefully cut by an EM UC6 ultrathin slicer (Leica, Germany) to form a flat paper/UF sample cross section. The processed samples were sputter-coated with a gold layer, and then observed using a Sigma 300 field emission scanning electron microscope (Zeiss, Germany), operating at an accelerating voltage of 5 kV.

2.4. *Bonding strength test*

The bonding strength of the UF resins was evaluated according to ISO 6237: 2017 "Adhesives - Wood-to-wood adhesive bonds - Determination of shear strength by tensile loading" with a slight modification. Since the joints bonded with the foamed UF resins in this study could have very low bonding strengths, an overlapping structure was designed for the bonded joints instead of a laminated pattern required in the standard to avoid damaging to the bonded joints from the subsequent addition of grooves to the veneers.

The preparation process of shear strength test samples is shown in Fig. 5. The bonding area of two sample veneers was evenly brushed with the UF resin at a glue spread of 200 g/m^2, and then assembled to form a

Table 2
Apparent density and shear strength of the UF resins with different TEM concentrations.

No.	TEM concentration (%)	Apparent density (kg/m^3)	Expansion ratio	Shear strength (MPa)
1	0	1281.8 ± 69.8	None	0.89 ± 0.09
4	2.5	287.1 ± 15.6	4.5	0.47 ± 0.05
5	5.0	213.5 ± 18.7	6.0	0.24 ± 0.02
6	7.5	151.6 ± 6.9	8.5	0.10 ± 0.01
7	10.0	130.9 ± 9.6	9.8	0.08 ± 0.01

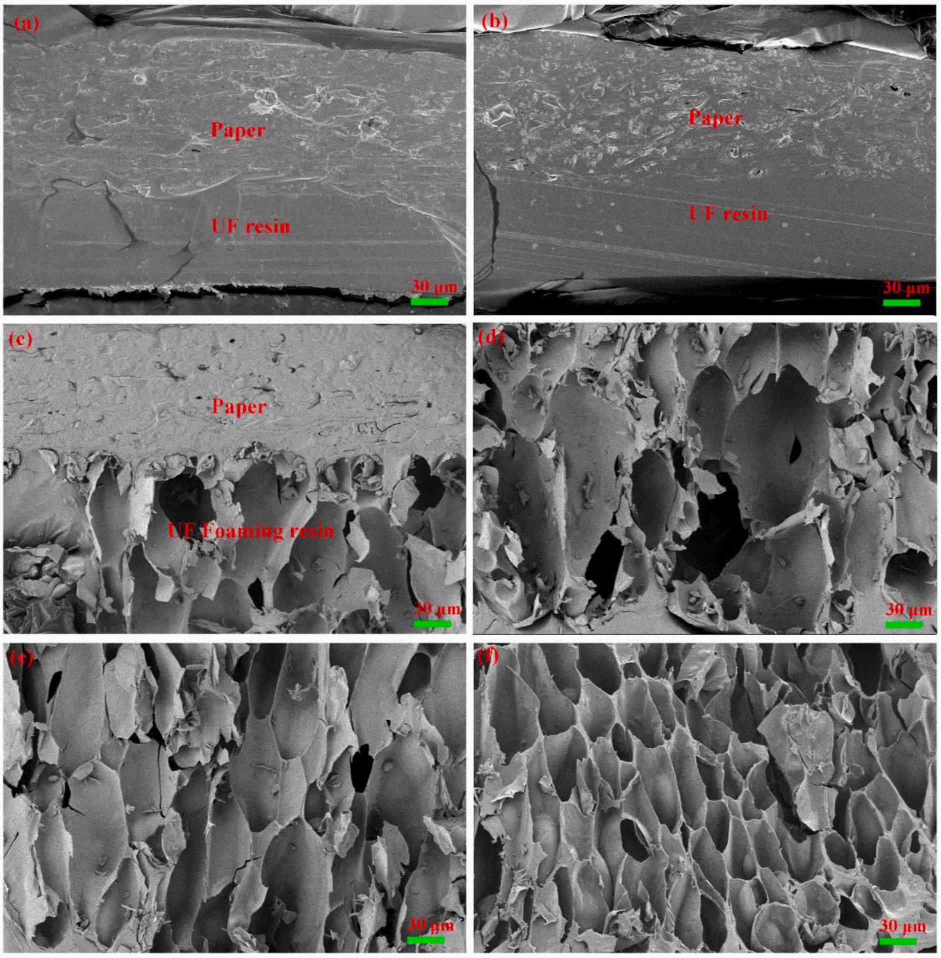

Fig. 8. Micro-morphologies of the paper/UF sample cross sections: No. 1 (a), No. 2 (b), No. 5 (c, d), No. 7 (e), and No. 9 (f).

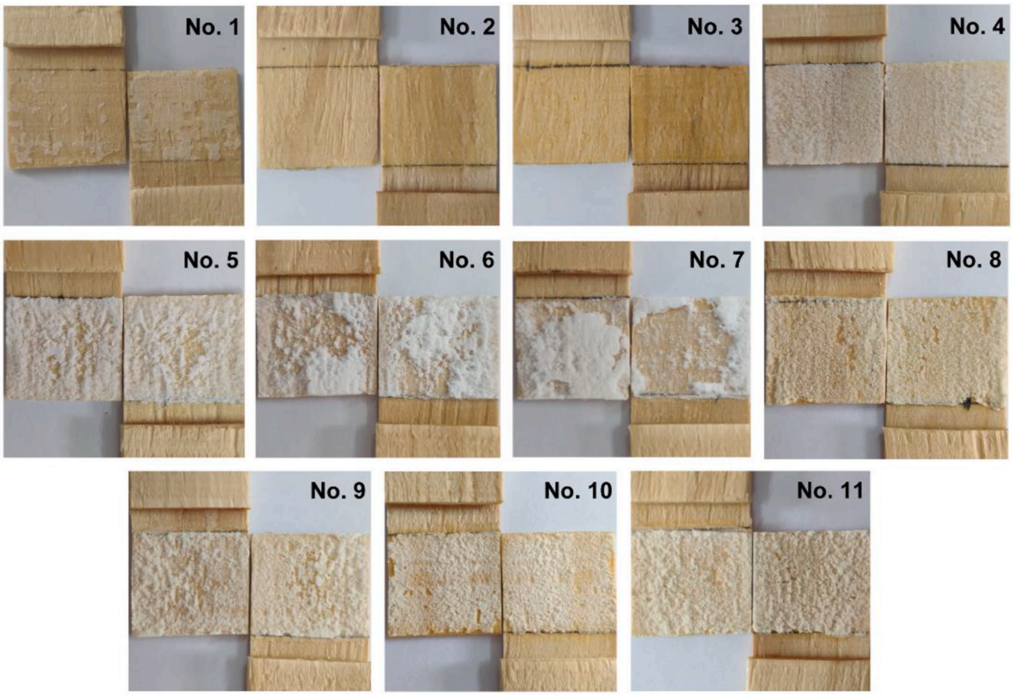

Fig. 9. Appearance of the bonded joint fracture surfaces during the bonding strength test.

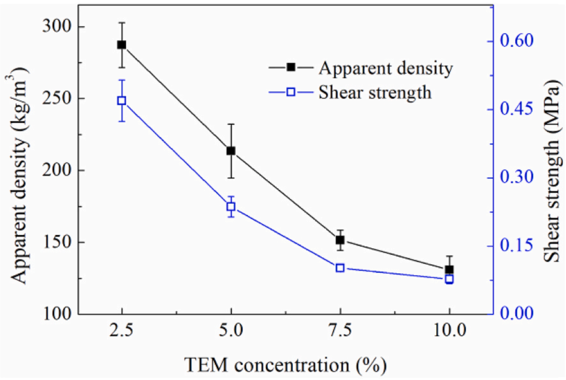

Fig. 10. Effects of TEM addition on the apparent density and shear strength of UF foaming resins.

two-layered bonded joint sandwiched by two stainless steel blocks (28 mm × 28 mm × 2 mm). The unit pressure on the bonded joints was calculated to be 0.00015 MPa, which not only stabilized the bonded joint but also had no significant influence on the UF resin foaming process. The assembled samples were immediately placed in the oven (120 °C) to cure for 8 min, and 10 replicates were used for each experimental group.

The samples were placed into a humid chamber (20 °C, 65% relative humidity) for one week before testing. Two veneer gaskets (25 mm × 20 mm) were applied near the ends of samples using double-sided tape (Fig. 5). The shear strength was measured by a UTM5504 universal testing machine (SUNS, Shenzhen, China), with a loading speed of 0.25 or 0.5 mm/min and a loading time of 60 ± 30 s.

3. Results and discussion

3.1. Appearance and pore structure of the UF foams

Optical micrographs of the control and expanded TEMs are shown in Fig. 6, and the TEMs of the two experimental groups were nearly spherical. The TEMs showed good dispersity in water due to their hydrophilic shells. This result is consistent with the fact that the UF foaming resins with evenly distributed TEMs were easily prepared by simple mechanical stirring. The diameter of the control and expanded TEMs were 18.5 ± 5.5 μm and 47.8 ± 17.3 μm, respectively. The core-shell structure was apparent for the control TEMs but difficult to distinguish for the expanded TEMs, mainly due to the thinning of the shell during the expansion. The diameter expansion ratio of the TEMs was nearly 3:1, which is consistent with the reported results of PMMA/pentane TEMs [8] and PU/n-hexane TEMs [10].

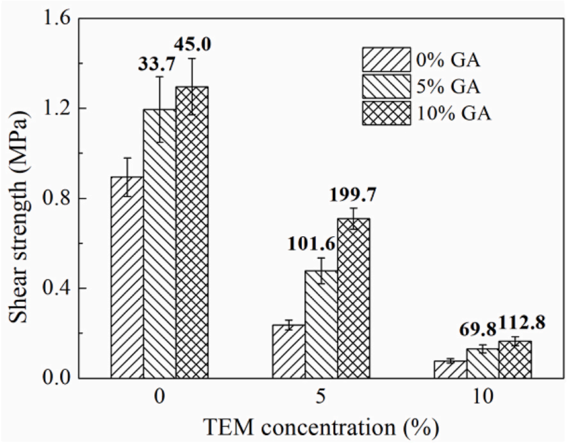

Fig. 11. Effects of GA addition on the shear strength of the UF foaming resins. Numbers above the error bars represent the increase (%) in the modified resins compared with the control resins.

The appearance of the glue lines in the bonded joints is shown in Fig. 7. The glue line was too thin to distinguish for the joint (No. 1) bonded with the neat UF resin, while apparent white glue lines were observed in the joints bonded with the UF foaming resins. Furthermore, as the TEM content increased from 0.5 g to 2.0 g (No. 4 to No. 7), the

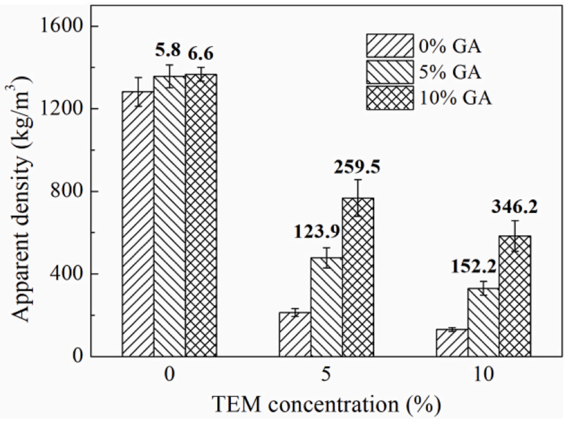

Fig. 12. Effect of GA addition on the apparent density of UF foaming resins. Numbers above the error bars represent the increase (%) in the modified resins compared with the control resins.

thickness of the glue lines obviously increased, indicating the enhanced expansion effect of the resins. In sum, a good expansion effect was preliminary found for the TEMs/UF resins.

SEM micrographs of the cross sections of the paper/UF samples are given in Fig. 8. A typical layered fiber structure was found in the paper cross section, and a smooth surface appeared in the unfoamed UF resins (No. 1 and No. 2). For both UF foaming resins (No. 5 and No. 7) and the modified UF foaming resin (No. 9), uniform cellular structures similar to those reported for UF foams [5–7] and phenol-urea-formaldehyde foam [25] were observed, indicating a good foaming ability of the resins. The pore size was approximately 20–100 μm, which is consistent with the diameter of the expanded TEMs. The profile of some pores was approximately oval rather than circular, probably due to the squeezing deformation during resin curing.

3.2. *Effects of the TEM concentration on UF foaming resin properties*

The apparent density was used to quantitatively describe the foaming performance of the UF resins. As shown in Table 2, the apparent density of the resins sharply decreased from 1281.8 kg/m^3 to less than 300 kg/m^3 after adding the TEMs, with expansion ratios of 4.5–9.8. The addition of TEMs was confirmed to be an effective way to reduce the resin density with the goal of producing "light-weight" wood-based materials. The apparent density (130–288 kg/m^3) of the foams was greater than that of reported UF foams (80–120 kg/m^3) [5], possibly owing to the larger pore size (283–556 μm) of the later foams. UF foams with lower apparent densities could be acquired by selecting TEMs with larger expansion diameters.

Unsurprisingly, the shear strength of the UF foaming resins was significantly lower than that of the control resin (Table 2). As the concentration of TEMs increased from 2.5% to 10.0%, the shear strength of the resin decreased by 47.5%–91.4%. The hollow structure inside the UF foams significantly reduced their mechanical strength. Additionally, as shown in Fig. 9, the failure of the bonded joints from all experiment groups appeared at the bond lines (with a wood failure ratio of 0%), which is mainly attributed to insufficient UF resin penetration into the wood because the pressure was too low [26].

The relationship between the TEM concentration and the UF foaming resin properties is further shown in Fig. 10. The apparent density and shear strength showed a similar decreasing trend. It is worth noting that the rate of change gradually decreased with an increase in the TEM content of the resins, and the difference in the properties between the 7.5% and 10.0% experimental groups was obviously less than that between the former experiment groups, which means the properties of the UF foaming resins were not simply linearly related to the TEM concentration. In summary, a lower apparent density within a certain range could be easily obtained for UF foaming resins by increasing the TEM content, but the resin would suffer from strength degradation. Similar trends were also found in UF foams with hexane used as the blowing agent, where the apparent density and compression strength gradually decreased with an increase in the amount of hexane added [5].

3.3. *Effects of GA modification on properties of UF foaming resin*

As discussed previously, the TEMs/UF resin performed well in terms of foaming, whereas its bonding strength was unsatisfactory. Therefore, GA, with high reactivity to urea [27], was used as a chemical crosslinking agent with the goal of improving the resin bonding strength. The effects of GA addition on the shear strength of UF foaming resins with different TEM concentrations are shown in Fig. 11. It is interesting that the UF resins with GA possessed significantly higher shear strength compared with the unmodified control samples. This result is similar to the improvement in the toughness and impact strength of phenol-urea-formaldehyde foams owing to PEG modification [20]. The active aldehydes in GA were apt to cross-link with the amino and hydroxyl groups in UF [28,29] to form a more stable network structure inside the resin and, thus, improving the bonding strength of the joints.

The rates of increase for the foaming UF resins (69.8%–199.7%) were greater than that of the unfoamed control resins (33.7%–45.0%), indicating that the enhancing effect on the shear strength was stronger for the foaming resins. Additionally, the UF resins with a higher GA concentration (10%) showed a better shear strength, while the increase rate was higher for resins with a lower TEM concentration (5%). Although the increase rate varied following the change in the TEMs or GA concentration, GA was confirmed to be a potential chemical modifier for TEMs/UF foaming resins in terms of improving their bonding strength.

However, the apparent density of the modified UF foaming resins experienced an unexpected increase, as shown in Fig. 12. The apparent density of the unfoamed resins showed a slight increase (5.8%–6.6%), whereas the rate of increase for the foaming resins reached 123.9%–346.2%. The rate of increase for the resins with 10% GA was significantly higher than that of resins with 5% GA. It appears that the GA crosslinking reaction restricted the expansion of the TEMs in the UF resins, although an evenly distributed cellular structure was formed inside the modified resins (Fig. 8). Nevertheless, a different conclusion was found in a previous study in terms of toughening UF foam using polyether amine (PEA) as the chemical modifier [7], and in this previous study, the addition of PEA not only improved the deformability of the foam but also led to a decrease in the apparent density. The mechanism of GA influence on the properties of UF foaming resins is worthy of further research.

4. Conclusions

In this research, a new foaming resin was successfully fabricated by adding TEMs to a UF resin. The developed resin showed good expansion, and the cured resin foams possessed an apparent density of less than 300 kg/m^3 and expansion ratios of 4.5–9.8. A uniform cellular structure inside the resin was demonstrated by SEM observations. However, the addition of TEMs to the UF resins resulted in a 47.5%–91.4% decrease in their shear strength.

As the concentration of TEMs increased from 2.5% to 10.0%, the apparent density and shear strength of the resins showed a similar decreasing trend with gradually reduced rates of change. This result indicates that increasing the TEM content could be an effective way to reduce the apparent density of UF foaming resin, but there would be an accompanying degradation in the resin strength. The introduction of GA to the UF foaming resin significantly improved its bonding strength, and the rate of increase varied from 69.8% to 199.7% following the change in the concentration of TEMs and GA. The addition of GA also increased the apparent density of the resin, possibly due to GA's crosslinking reaction restricting the TEM expansion.

Acknowledgements

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