

The Effects of Sealing Treatment and Wood Species on Formaldehyde Emission of Plywood

Wubin Ding, Wenyan Li, Qiang Gao, Chunrui Han, Shifeng Zhang,* and Jianzhang Li *

Urea-formaldehyde (UF) resin (F/U: 1.25), melamine-urea-formaldehyde (MUF) resin (F/(U+M): 1.05), and three kinds of wood species including poplar (*Populus davidiana* Dode), beech (*Fagus engleriana* Seem.), and eucalyptus (*Eucalyptus robusta* Smith) were used to produce plywood. The effects of sealing treatment and wood species on the formaldehyde emissions were studied. The anatomical characteristics of different wood species were measured. The results showed that: (1) formaldehyde emission of plywood treated by surface sealing was higher than without treatment; (2) formaldehyde emission of nine-ply poplar plywood bonded by UF resin decreased by 74.4% to 1.98 mg/L after edge sealing treatment; (3) compared with beech and poplar plywood, the formaldehyde emission of five-ply eucalyptus plywood bonded by MUF resin was the lowest obtained, at 0.19 mg/L; (4) formaldehyde emission of poplar plywood from the surface changed slightly in spite of different layers. The contact angle and spreading-penetration coefficient, *K*, analyses showed that the cell arrangement of eucalyptus was dense. Scanning electron micrographs indicated that the pore sizes of eucalyptus samples were the smallest in contrast to poplar and beech.

Keywords: Plywood; Formaldehyde emission; Sealing treatment; Wood species; Contact angle device; SEM

Contact information: College of Materials Sciences and Technology, Beijing Forestry University, Beijing 100083, P. R. China; *Corresponding author: lijianzhang_bjfu@163.com; hnzsf142@126.com

INTRODUCTION

Currently, urea-formaldehyde (UF) resin is one of the most widely used synthetic resins in the wood composite industry thanks to its simple synthesis process, low cost, fast curing, water solubility, and good performance (Zhang *et al.* 2011; Park and Kim 2008; Gao *et al.* 2012). The production of plywood in China is the largest in the world (Qian 2012). More than 90 percent of plywood factories in China are using UF resin and modified UF resin, such as MUF resin, to manufacture plywood (Gao *et al.* 2011). However, the release of formaldehyde by formaldehyde-based adhesives during the manufacture and use of plywood is a matter of concern.

Formaldehyde is a well-established cause of indoor air pollution. It is classified as a carcinogen and hazardous to human health, causing eye and throat irritations as well as respiratory discomfort (Kim *et al.* 2006; He *et al.* 2011; Li *et al.* 2004; Gao *et al.* 2011). Tightened environmental regulations on formaldehyde emissions in the production and use of wood composites have created an urgent need for test methods and ways to limit formaldehyde emissions. The formaldehyde emission of plywood is measured mainly by the desiccator method in China. This is because the method is efficient (taking 24 h), and specialized and expensive equipment is not needed (Salem *et al.* 2012). The

formaldehyde emission of plywood from the surface and edge is also measured by the desiccator method, which is different from the formaldehyde emission behavior in a typical interior space (An *et al.* 2010).

In practice, edge sealing has been widely adopted to treat plywood used as furniture materials and floorings. Although the use of edge sealing treatment for the desiccator method is described in an ASTM test method (2000 ASTM), the Chinese National Standard has not specifically mentioned any sealing treatment in the desiccator method. Formaldehyde emission of plywood from the edge and from the surface is different because it is a heterogeneous material. Some studies were conducted on the effect of sealing treatment on formaldehyde emission of plywood. Li *et al.* (2005) concluded that formaldehyde emission of five-ply plywood with surface sealing treatment was higher than that of a corresponding sample without sealing treatment. Li *et al.* (2005) and Kim *et al.* (2006) reported that edge sealing treatment could reduce formaldehyde emission of plywood effectively. However, Kim *et al.* (2005) mainly focused on measuring the formaldehyde emission of wood-based panels with edge sealing treatment and had no discussion concerning the overall emission law of plywood. Li *et al.* (2006) did not research the effect of sealing treatment on formaldehyde emission of different layers plywood and neglected the effect of wood species on formaldehyde emission of plywood.

The effect of wood species on formaldehyde emissions of plywood is significant. Martinez and Belanche (2000) reported that the formaldehyde emission of plywood made from the veneers of wood having a large porosity value was lower than that of plywood made from veneers that were of low porosity after 4 weeks of production. However, the method used to calculate the porosities of wood species was not clear and accessible.

Research systematically studying the formaldehyde emission law of plywood and focusing on the effects of sealing treatment and wood species on formaldehyde emissions from different-layer plywood have been barely addressed. Such information will be helpful to understand the characteristics of formaldehyde emissions from plywood and subsequently treat specimens properly before testing by the desiccator method. With proper sealing treatment, the formaldehyde emission of plywood measured by the desiccator method is reliable, relatively close to the conditions used in practice, and able to avoid the overestimation of formaldehyde emissions from plywood. Understanding the microscopic structure of wood will explain the difference in formaldehyde emission of plywood from different wood species and will help us predict emission amount.

The objectives of this research were to research the formaldehyde emission law and characteristics of plywood and the influence effects on formaldehyde emission of plywood.

The goals of the experiments were conducted as follows: (1) to investigate the effects of sealing treatment on formaldehyde emission of different-layer plywood bonded by UF resin and MUF resin, respectively; (2) to study the effect of wood species on formaldehyde emission of five-ply plywood bonded by MUF resin and treated with sealing; (3) to quantitatively evaluate the porosities of three wood species by means of contact angle tests and measurements of the spreading-penetration coefficient K , and to utilize SEM to observe anatomical characteristics of different wood species.

EXPERIMENTAL

Materials

The 98 wt% urea, 37 wt% formaldehyde solution, and 99.8 wt% melamine were purchased from China National Petroleum Corporation, Beijing Chemistry Company, and China Chemistry Company, respectively. Sodium hydroxide, formic acid, and ammonium chloride were of A.R grade from Beijing Chemistry Company, China. Wheat flour was bought from the Beijing Old Ship Food Company. The veneers of poplar, beech, and eucalyptus were obtained from Wen'an, Heibei province, China.

Methods

Preparation of UF resin

The UF resin was synthesized using formaldehyde and urea at a molar ratio of 1:0.8 in the laboratory. The urea was added into formaldehyde solution three times at the weight rates of 20:5:7. The formalin was placed in the reactor, adjusted to pH 8.0 with aqueous NaOH (40%wt. solution), and then the first amount of urea was added. The mixture was then heated to 90 °C under reflux for 1 h. The acidic reaction was brought by adding formic acid (30%wt. solution) to obtain a pH of about 5.0, and the condensation reactions were carried out until it reached a target viscosity. Then the mixture was adjusted to pH 8.0 by using NaOH, and the second amount of urea was added. After 0.5 h at 80 °C, final mole ratios of UF resins were adjusted by adding the third amount of urea and further stirring at 70 °C for 0.5 h. Then, the UF resin was cooled to room temperature, and later followed by adjusting the pH to 8.0.

Preparation of MUF resin

The MUF resin was synthesized using formaldehyde, melamine, and urea at a molar ratio of 1.1:0.05:1 in the laboratory. The urea and melamine were mixed and added into the formaldehyde solution three times at the weight rates of 26:5:16. The formalin was placed in the reactor, then adjusted to pH 8.0 with aqueous NaOH (40%wt. solution), and then the first amounts of urea and melamine were added. The mixture was then heated to 90 °C under reflux for 1 h. The acidic reaction was brought about by adding formic acid (30%wt. solution) to obtain a pH of about 5.0, and the condensation reactions were carried out until reaching a target viscosity. Then, the mixture was adjusted to pH 8 by using NaOH, and the second portions of urea and melamine were added. After 0.5 h at 85 °C, final mole ratios of MUF resins were adjusted by adding the third amounts of urea and melamine and further stirring at 70 °C for 0.5 h. Then, the MUF resin was cooled to room temperature, later followed by adjusting the pH to 8.0. The properties of resin are listed in Table 1. They were measured according to China National Standard (GB/T 14074-2006).

Table 1. Properties of Adhesive

Resin	F/U	Viscosity (cP)	Solid content	Gel time(s)	pH
MUF	1.05(M=4%)	146(20 °C)	54.50%	315	7.32
UF	1.25	106.5(20 °C)	52.20%	86	7.84

Preparation of plywood samples

Before adhesive application, the veneer was dried using an oven (100 °C ± 2 °C) to achieve a moisture content in the range 8% to 10%. The adhesive was applied to two

sides of veneer (400mm×400mm×1.6mm; moisture content, 8% to 10%) with 300 to 340 g/m² of the adhesive spread level. The coated veneer was stacked between two uncoated veneers with the grain directions of two adjacent veneers perpendicular to each other. The stacked veneers were hot-pressed at 1.0 MPa and 120 °C with a pressing time of 60 s/mm. Twenty grams of wheat flour and 0.5 g ammonium chloride were added into 100 g resin and stirred for 10 min at room temperature to form adhesive. After hot-pressing, the panel was stored at ambient environment for at least 24 h before it was treated with sealing and evaluated for its formaldehyde emission.

Sealing treatment

As defined in the Chinese National Standard desiccator methods, a 5 cm × 15 cm (L × W) sample, with the thickness not taken into account, was prepared. To analyze the formaldehyde emission of plywood from the edge and the surface, each sample was treated with surface sealing or edge sealing by polyethylene wax.

Formaldehyde emission measurement

The formaldehyde emission of plywood was determined using the desiccator method in accordance with the procedure described in China National Standard (GB/T 9848.3-2004). The plywood specimen was prepared with dimensions of 50 mm × 150 mm. Ten specimens per panel were put into a 9-11 L sealed desiccator at 20 ± 2 °C for 24 h. The emitted formaldehyde was absorbed by 300 mL deionized water in a container. The water was measured by a visible spectrophotometer to obtain the formaldehyde emission value. The principle for determining the concentration of formaldehyde absorbed in the deionized water is based on the Hantzsch reaction, in which the formaldehyde reacts with ammonium ions and acetylacetone to yield diacetyldihydrolutidine (DDL). The absorbance was measured at a wavelength of 412 nm for the emission of formaldehyde. The average value of formaldehyde emission was calculated from three panels.

Contact angle measurement

Contact angle was determined by OCA contact angle goniometer. The determination of balance contact angle was as follows. With deionized water (2 µL) dropping and gradually spreading on the sample surface, the contact angle got smaller and smaller before reaching an equilibrium value. At 140 s there was no further change in the contact angle. So the contact angle at 140 s was taken as the equilibrium contact angle. Thirty-six data points were taken for each recorded drop to obtain a curve of contact angle vs. time. Five replicates were averaged for each sample of wood species. Each wood species had three samples.

Wood penetration measurement

The equation of the wetting model is expressed as follows (Shi and Gardner 2001; Wei *et al.* 2012),

$$\theta = \frac{\theta_i \cdot \theta_e}{\theta_i + (\theta_e - \theta_i) \exp[-K(\theta_e/(\theta_e - \theta_i))t]} \quad (1)$$

where θ_i represents the instantaneous contact angle, θ_e represents equilibrium contact angle, θ represents contact angle at time t , and K is a constant referred to as the intrinsic relative contact angle decrease rate. The physical meaning of the K value represents how

fast the liquid spreads and penetrates into the porous structure of wood.

To obtain the K -value for the deionized water and wood system, a linear-fitting method can be used to fit the empirical data in Eq. (1). Reorganizing Eq. (1), one obtains the following expression:

$$\theta \cdot \theta_i + \theta(\theta_e - \theta_i) \cdot e^{K(\theta_e/(\theta_e - \theta_i))t} = \theta_i \cdot \theta_e$$

Then

$$e^{K(\theta_e/(\theta_e - \theta_i))t} = \frac{\theta_i(\theta_e - \theta)}{\theta(\theta_e - \theta_i)} \quad (2)$$

Logarithmic transformation of the Eq. (2) yields the following expression:

$$K \cdot \frac{\theta_e}{\theta_e - \theta_i} \cdot t = \ln \frac{\theta_i(\theta_e - \theta)}{\theta(\theta_e - \theta_i)} \quad (3)$$

In Eq. (3), $\theta_i(\theta_e - \theta)/\theta(\theta_e - \theta_i)$ was defined as y . With this change, Eq. (3) then can be express as shown in Eq. 4:

$$K \cdot t = \frac{\theta_e - \theta_i}{\theta_e} \ln y \quad (4)$$

By using the definition, $a = [(\theta_e - \theta_i)/\theta_e] \ln y$, a linear equation is obtained:

$$a = K \cdot t \quad (5)$$

With a mapping of t , different slopes of the straight line are defined, and the slope is the K -value.

Imaging with a scanning electron microscope (SEM)

Test samples were directly cut from veneer and then all the samples were placed on an aluminum stub. A coating of 10 nm Au/Pd film was applied to the samples. The coated samples were then examined and imaged using a S-3400N scanning electron microscope (SEM) (Hitachi, Japan)

Statistical analysis

Formaldehyde emission data were analyzed by one-way ANOVA of compare means using SPSS[®] statistical software (version 17.0). All comparisons were based on a 95% confidence interval. Differences between groups were considered significant at $P < 0.05$.

RESULTS AND DISCUSSION

Effects of Sealing Treatment on Formaldehyde Emission of Plywood

Figure 1 shows the effect of sealing treatment on formaldehyde emissions of poplar plywood having different numbers of layers bonded by UF and MUF resins. Formaldehyde emission of plywood without sealing treatment increased with an increasing number of plywood layers. Formaldehyde emission of plywood with surface sealing treatment was higher than that without treatment. The explanation is that formaldehyde mainly is emitted through vessels of veneer and the inner pressure of

formaldehyde gas increases after the surface sealing treatment, resulting in an increase of formaldehyde emission rate (He *et al.* 2011). This result is consistent with the previous research of Li *et al.* (2005). After edge sealing treatment, the formaldehyde emission of nine-ply plywood bonded by UF resin was measured as 1.98 mg/L, which was 0.04 mg/L higher than seven-ply plywood, 0.15 mg/L higher than five-ply plywood, and 0.76 mg/L higher than three-ply plywood (Fig. 1a). That bonded by MUF resin was measured as 0.84 mg/L, which was 0.13 mg/L higher than seven-ply plywood, 0.06 mg/L higher than five-ply plywood, and 0.04 mg/L higher than three-ply plywood (Fig. 1b).

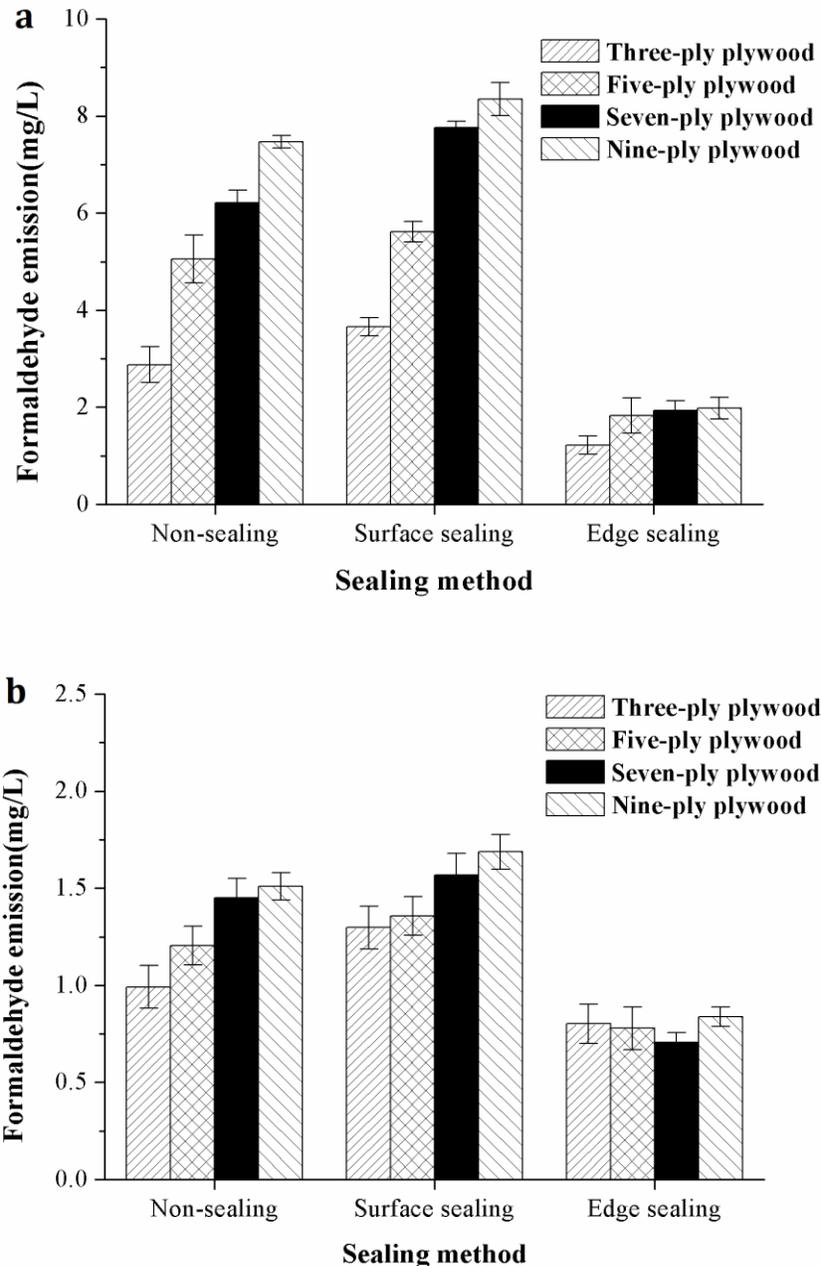


Fig. 1 a. Effect of sealing treatment on formaldehyde emission of different layers poplar plywood bonded by UF resin. Data are the means of 3 replications, and the error bar represents one standard error of mean. **b.** Effect of sealing treatment on formaldehyde emission of different layers poplar plywood bonded by MUF resin. Statistical details are the same as those in **a**.

The significance of sealing treatment to formaldehyde emission was analyzed by LSD_{0.05} (Least Significance Difference) method in SPSS. The variance analysis results are shown in Tables 2 and 3. Based on the comparison between non-sealing treatment and edge sealing treatment in Table 2, the *P* values of different layers plywood bonded by UF resins were less than 0.05, which were considered the differences between these two treatments were significant. Comparing with the plywood bonded by UF resin, the *P* values of plywood bonded by MUF resin were also less than 0.05 except for three-ply plywood, because formaldehyde emission of the one was small (Table 3). Formaldehyde emission decreased significantly through edge sealing treatment. In comparison to those without treatment, the reduction rates of formaldehyde emission of nine-ply plywood bonded by UF and MUF resins and treated with edge sealing were 74.4% from 7.74 mg/L to 1.98 mg/L and 44.5% from 1.51 mg/L to 0.84 mg/L, respectively.

Table 2. Variance Analysis Results of Formaldehyde Emissions of Poplar Plywood Bonded by UF resin

Plywood	Sealing Treatment		Mean Difference (I-J)	Std. Error	Sign. (<i>P</i>)	95% Confidence Interval	
						Low bound	Upper bound
Three-ply	1	2	-0.78	0.37689	0.084	-1.7022	0.1422
		3	1.66*	0.37689	0.005	0.7378	2.5822
Five-ply	1	2	-0.57	0.52464	0.319	-1.8537	0.7137
		3	3.22*	0.52464	0.001	1.9363	4.5037
Seven-ply	1	2	-1.54*	0.28083	0.002	-2.2272	-0.08528
		3	4.28*	0.28083	0.000	3.5928	4.9672
Nine-ply	1	2	-0.61	0.33005	0.114	-1.4176	0.1976
		3	5.76*	0.33005	0.000	4.9524	6.5676

* The mean difference is significant at the 0.05 level ($P < 0.05$)

1: Non-sealing; 2: Surface sealing; 3: Edge sealing.

Table 3. Variance Analysis Results of Formaldehyde Emissions of Poplar Plywood Bonded by MUF Resin

Plywood	Sealing Treatment		Mean Difference (I-J)	Std. Error	Sign. (<i>P</i>)	95% Confidence Interval	
						Low bound	Upper bound
Three-ply	1	2	-0.30667	0.13562	0.064	-0.6385	0.0252
		3	0.18667	0.13562	0.218	-0.1452	0.5185
Five-ply	1	2	-0.15333	0.1489	0.343	-0.5177	0.211
		3	0.42333*	0.1489	0.029	0.059	0.7877
Seven-ply	1	2	-0.12	0.12555	0.376	-0.4272	0.1872
		3	0.74333*	0.12555	0.001	0.4361	1.0505
Nine-ply	1	2	-0.17667	0.10129	0.132	-0.4245	0.0712
		3	0.67333*	0.10129	0.001	0.4255	0.9212

* The mean difference is significant at the 0.05 level ($P < 0.05$)

1: Non-sealing; 2: Surface sealing; 3: Edge sealing.

Table 4. Statistical Analysis of Formaldehyde Emissions of Poplar Plywood from Surface Bonded by UF Resin

Plywood	Mean Difference (I-J)	Std. Error	Sign. (P)	95% Confidence Interval		
				Low bound	Upper bound	
Nine-ply	Three-ply	0.76	0.35263	0.063	-0.0532	1.5732
	Five-ply	0.15	0.35263	0.682	-0.6632	0.9632
	Seven-ply	0.04	0.35263	0.912	-0.7732	0.8532

Table 5. Statistical Analysis of Formaldehyde Emissions of Poplar Plywood from Surface Bonded by MUF Resin

Plywood	Mean Difference (I-J)	Std. Error	Sign. (P)	95% Confidence Interval		
				Low bound	Upper bound	
Nine-ply	Three-ply	0.3333	0.10157	0.751	-0.2009	0.2676
	Five-ply	0.05667	0.10157	0.592	-0.1776	-0.2908
	Seven-ply	0.13000	0.10157	0.236	-0.1042	-0.3642

The mean difference is insignificant if $P > 0.05$.

After edge sealing treatment, the statistical analysis of formaldehyde emission of plywood from surface bonded by UF and MUF resins are shown in Tables 4 and 5. In spite of UF and MUF resins, the formaldehyde emission differences among four groups (three-ply, five-ply, seven-ply, and nine-ply plywood) were found to be insignificant, because all P values of surface formaldehyde emission of different layers plywood bonded by UF and MUF resins, respectively, were more than 0.05. So the formaldehyde emission of different layers plywood with edge sealing treatment was statistically the same. This is because the veneer acted like a barrier, resulting in inhibition of formaldehyde emission from the surface.

Effects of Edge Sealing Area on Formaldehyde Emission of Plywood

Figure 2 shows the effect of edge sealing area on formaldehyde emission of five-ply plywood made from poplar veneers and bonded by MUF resin. Results show that the formaldehyde emission decreased with an increase in edge sealing area. After edge sealing treatment, the formaldehyde emission of plywood treated by surface sealing decreased 72.5% from 1.42 mg/L to 0.39 mg/L and that of plywood with non-surface sealing treatment decreased 29.9% from 1.14 mg/L to 0.8 mg/L. However, when edge sealing area was in the range of 25.1% to 50.2% of total edge area of five-ply plywood, formaldehyde emission of plywood decreased very slightly. This is because the rate of emission increases as a result of an increase of inner formaldehyde pressure by edge sealing treatment, which compensates for a decrease of exposed edge area.

Effects of Wood Species on Formaldehyde Emission of Plywood

Figure 3 shows the effect of sealing treatment on formaldehyde emission of five-ply plywood made from three wood species. The formaldehyde emission of five-ply plywood made from poplar veneers and bonded by MUF resin was 1.13 mg/L, which was

146.2% (0.67 mg/L) more than beech plywood (0.46mg/L) and 500% (0.94 mg/L) more than eucalyptus plywood (0.19 mg/L).

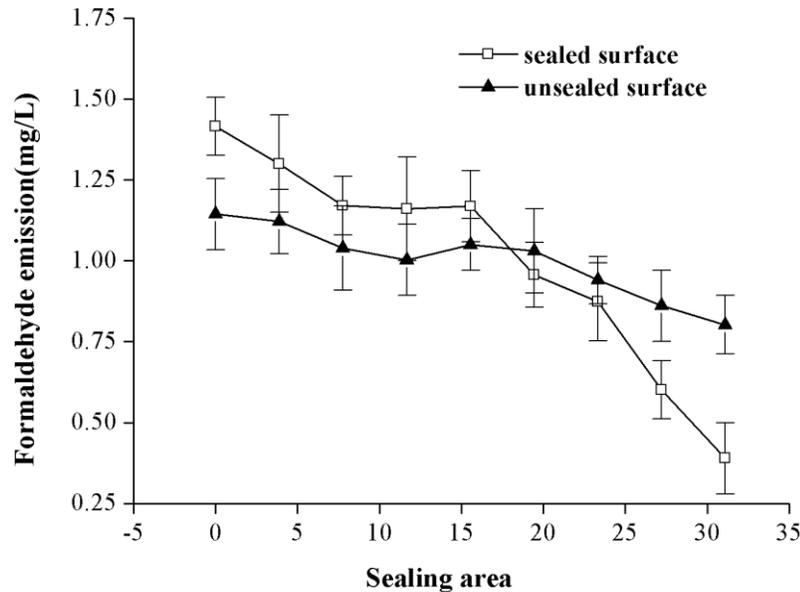


Fig. 2. Effect of edge sealing area on formaldehyde emissions of 5-ply plywood made from poplar

The formaldehyde emissions from the three kinds of plywood increased with surface sealing treatment. After all plywood specimens underwent edge sealing treatment, the five-ply plywood made from poplar veneers and bonded by MUF resin had the highest formaldehyde emission values (0.73 mg/L) compared with the other two kinds of plywood. The formaldehyde emission of eucalyptus plywood was lower than that of beech plywood. The effects of wood species on formaldehyde emission of plywood are attributed to the physical properties of wood species, such as density, extractives content, and anatomical characteristics. There are three main reasons to explain the differences between the formaldehyde emissions. The first one is that formaldehyde in the adhesive interacts in a distinct way with each wood species. The formaldehyde can form compounds with the cellulose of wood under the pressing conditions (high temperature and acid medium produced by the catalyst); the second one is that formaldehyde emission is affected by the anatomy of the respective wood species, such as porosity and density (Martinez and Belanche 2000); the third one is that certain wood extractives react with formaldehyde and hence act as a formaldehyde scavenger (Schafer and Roffael 2000). However, cellulose content of different wood species is very close in value. Moreover, the moisture content and wood pH values of different wood species are similar. For these reasons there is only slight probability that formaldehyde interaction with each wood species has an influence on formaldehyde emission (Martinez and Belanche 2000). So the anatomy and extractives contents of wood species are the major factors that may affect formaldehyde emissions. The porosity of beech was low (Martinez *et al.* 2000). Compared with beech and eucalyptus plywood, the density of poplar plywood was the lowest, and its anatomy was simple (Bohm *et al.* 2012). These characteristics of poplar allowed formaldehyde to be emitted easily from the panel during the testing, and the poplar plywood exhibited the highest formaldehyde emissions of three kinds of plywood. The beech and eucalyptus wood have a lot of extractives. These represent barriers to the

emission of formaldehyde from the panel (Martinez and Belanche 2000; Jiang *et al.* 2002).

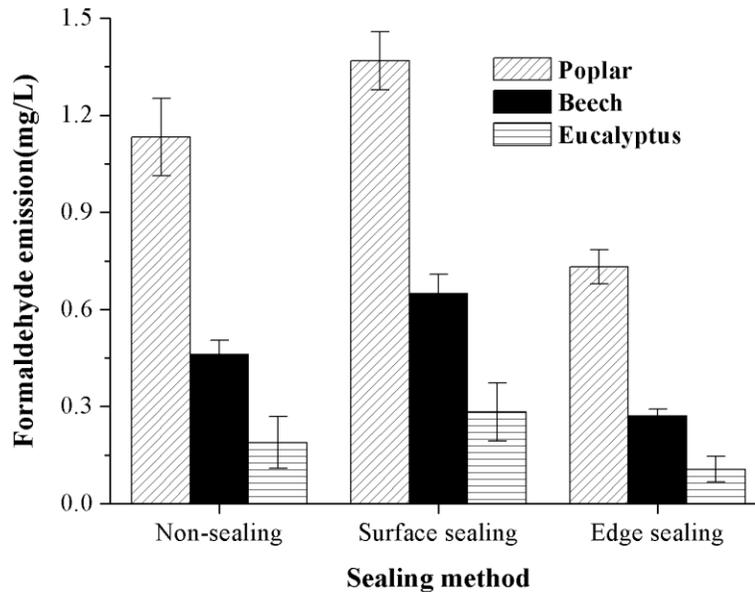


Fig. 3. Effects of wood species and sealing treatment on formaldehyde emission of five-ply plywood

Penetration Properties of Plywood

In order to investigate the anatomical characteristics of different wood species, a contact angle goniometer was used to test the contact angle as a function of time on the surface of the veneer. Based on Eq. (5), the spreading-penetration parameter K was calculated by contact angle. Figure 4 shows the change of contact angle as a function of time of contact on different surfaces of plywood. The results show that the contact angles of the same size of deionized water droplet (2 μL) on three surfaces of veneer were all declining evidently and trending to equilibrium values at 140 s. Finally, equilibrium contact angles of three surfaces were obtained. From the comparison curves plotted in Fig. 4, it is apparent that wood species had a significant effect on the water spreading-penetration process. The instantaneous contact angle of deionized water on the surface of beech was comparable to that of eucalyptus, which was higher than that of poplar. The equilibrium contact angles on poplar, beech, and eucalyptus veneer surface were 31.2° , 82° , and 69.1° , respectively. It can also be seen in Fig. 4 that poplar exhibited the largest percent decrease in contact angle from initial to equilibrium when compared to that of beech and eucalyptus (70.4% vs. 30.5% vs. 42.6%). In addition to dynamic contact angle of three wood species, the spreading-penetration coefficient K of the same deionized water droplet on the surfaces of three wood species was determined. According to the calculation method of K -value in the Eq. (5), values at different times (t) were obtained firstly, and then the slopes of fitted to straight lines, yielding the K -values. Figure 5 shows the a vs. t curves for veneer of different wood species. Results show the spreading-penetration coefficient K on the surface of poplar was 0.02846: 169.3% greater than that of beech (0.01057) and 68.4% greater than that of eucalyptus (0.0169). The greatest K -values of deionized water indicated its faster penetration and spreading process on the poplar surface compared to beech and eucalyptus. Therefore, the density of poplar was

small, arrangement of cells in poplar was loose, and its porosity was relative large. These characteristics led to the smallest equilibrium contact angle, the highest rate of decrease of the contact angle, and the highest spreading-penetration coefficient K . So formaldehyde was easily emitted through the veneer, which confirmed the result that the formaldehyde emission of poplar plywood was the highest compared with eucalyptus and beech plywood. The formaldehyde emission of plywood made from beech and eucalyptus were low due to their large equilibrium contact angles and low spreading-penetration coefficients K . The results of porosity measurement of three wood species were consistent with previous studies (Martinez and Belanche 2000; Bohm *et al.* 2012).

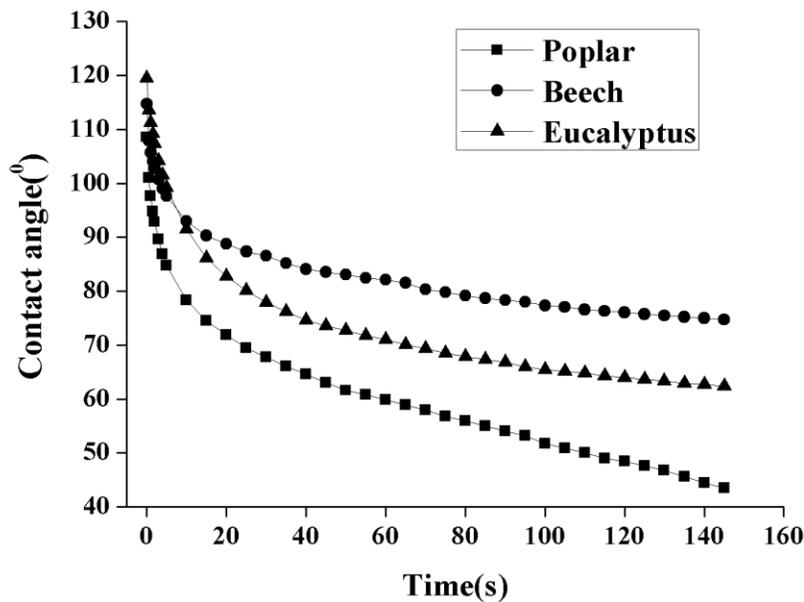


Fig. 4. The change of contact angle as a function of time on different surfaces of veneer

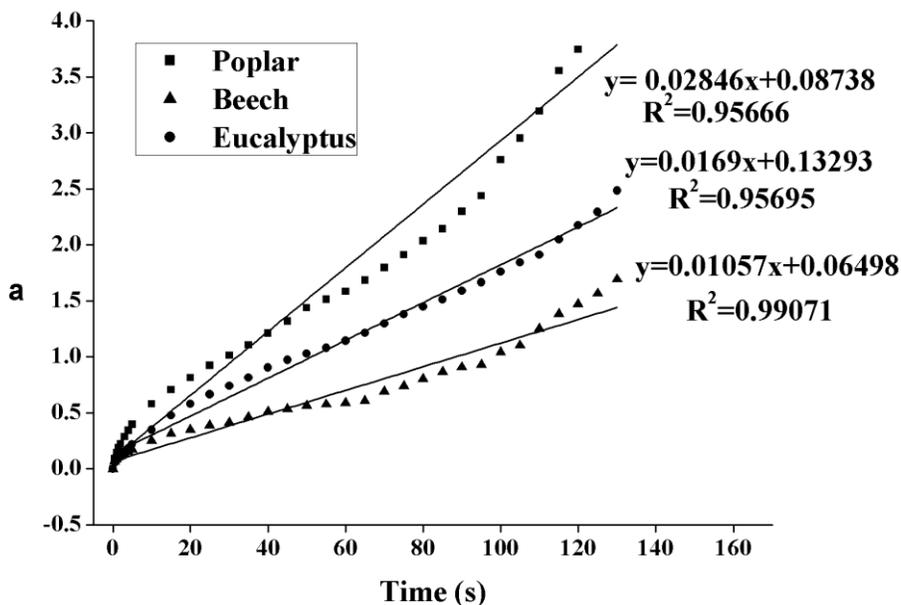


Fig. 5. The case of the parameter a as a function of t on veneer of different veneer wood species

Anatomical Characteristics of Three Wood Species

Figure 6 shows SEM images comparing the microscopic structure of the surfaces of veneers of the three wood species. The anatomical characteristics of the different wood species are shown to be dramatically different. Many pit apertures were observed on the surface of vessels of poplar and the diameter of the pit aperture was large (Fig. 6A). Considering these factors, formaldehyde and deionized water could easily pass through the veneer by these pit apertures. This result confirmed that the pore sizes of poplar veneer were large. This result also explained why the spreading-penetration coefficient K of poplar plywood on the surface and formaldehyde emission of poplar plywood was the highest compared to the other two kinds of plywood. The diameter of pit aperture on the surface of vessels in beech was relatively smaller than that of poplar and the pits were covered by some substance. The vessels of beech veneer were partially covered by spiral thickening (Fig. 6B).

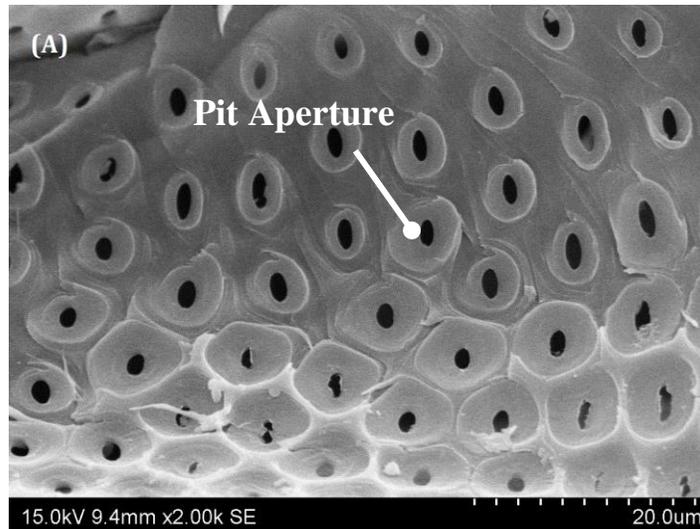


Fig. 6a. The microscope structure of veneer surface: (A) poplar



Fig. 6b. The microscope structure of veneer surface: (B) beech

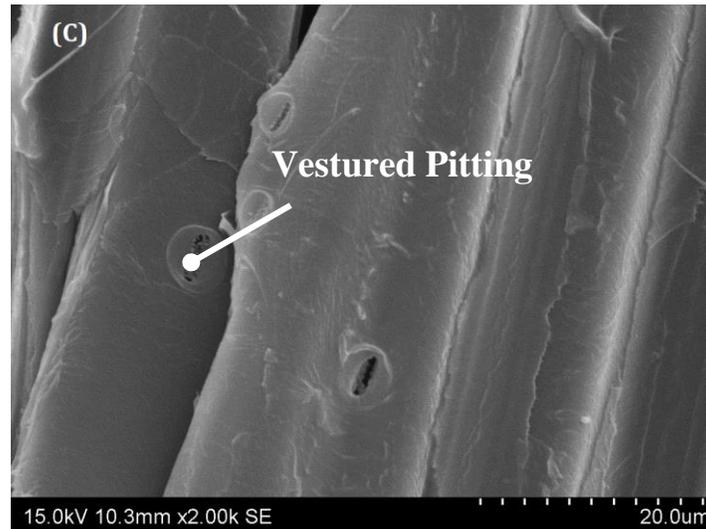


Fig. 6c. The microscopic structure of veneer surface: (C) eucalyptus

The vessel structure of beech tended to impede the passage of formaldehyde and deionized water. Compared with poplar and beech, there were a smaller number of pit apertures in eucalyptus (Fig. 6C). Pits in eucalyptus exhibited vestured pitting, and the vessels of eucalyptus contained more tyloses (Jiang *et al.* 2002), which inhibited the emission of formaldehyde from the plywood. This confirmed the result that the formaldehyde emission of plywood made from eucalyptus veneers was the lowest.

CONCLUSIONS

1. Formaldehyde emission of plywood is higher with surface sealing treatment than with non-sealing treatment. After edge sealing treatment, by contrast, the reduction rate of formaldehyde emission of plywood made from poplar and bonded by UF resin ranged from 57.6% to 74.4% and that bonded by MUF resin ranged from 19% to 44.5%.
2. Formaldehyde emission of plywood from the surface was statistically the same in spite of layer numbers and types of resin. Based on these findings, formaldehyde emission of plywood specimen treated by edge sealing will be much lower than without sealing treatment, which is closed to the real situation. However, surface sealing treatment will tend to cause an over-estimation of formaldehyde emission levels of plywood.
3. The formaldehyde emission decreased with an increase of edge sealing area. However, the emission decreased very slightly when the edge sealing area was in the range of 25.1% to 50.2% of total edge area.
4. Wood species had a great effect on the formaldehyde emission of plywood. The formaldehyde emission of five-ply plywood bonded by MUF resin and made from poplar was 1.13 mg/L, which was 146.2% (0.67 mg/L) more than beech plywood and

500% (0.94 mg/L) more than eucalyptus plywood.

5. Poplar plywood had the smallest equilibrium contact angle and the highest spreading-penetration coefficient K because of its low density, loose arrangement of cells, and large porosity compared with that of beech and eucalyptus plywood.
6. SEM results show that fewer pit apertures on the vessel surface of eucalyptus were observable in contrast to what was observed with poplar and beech. Eucalyptus vessels contained more tyloses, and most of pits exhibited vestured pitting.

ACKNOWLEDGEMENTS

The authors are very grateful for financial support from the Special Fund for Forestry Research in the Public Interest (Project 201204702) and National Natural Science Foundation of China (Project 31000268/C160302).

REFERENCES CITED

- An, J. Y., Kim, S., Kim, H. J., and Seo, J. (2010). "Emission behavior of formaldehyde and TVOC from engineered flooring in under heating and air circulation systems," *Build. Environ.* 45, 1826-1833.
- ASTM. (2000). "Standard test method for determining formaldehyde levels from wood products using a desiccator," The American Society for Testing and Materials, Philadelphia.
- Bohm, M., Salem, M. Z. M., and Srba, J. (2012). "Formaldehyde emission monitoring from a variety of solid wood, plywood, blockboard and flooring products manufactured for building furnishing materials," *J. Hazard. Mater.* 221-222, 68-79.
- Gao, Q., Shi, S. Q., Li, J. Z., Liang, K. W., and Zhang, X. M. (2011). "Soybean meal-based wood adhesive enhanced by modified polyacrylic acid solution," *BioResources* 7(1), 946-956.
- Gao, Q., Shi, S., Zhang, S. F., Li, J. Z., and Liang, K. W. (2011). "Improved plywood strength and lowered emission from soybean meal/melamine urea-formaldehyde adhesives," *Forest Prod. J.* 61(8), 688-693.
- Gao, Q., Shi, S. Q., Zhang, S. F., Li, J. Z., Wang, X. M., Ding, W. B., Liang, K. W., and Wang, J. W. (2012). "Soybean meal-based adhesive enhanced by MUF resin," *J. Appl. Polym. Sci.* 125, 3676-3681.
- He, Z. K., Zhang, Y. P., and Wei, W. J. (2011). "Formaldehyde and VOC emissions at different manufacturing stages," *Build. Environ.* 28, 23-25.
- Jiang, Z. H., Fei, B. H., Wang, X. M., Zhao, R. J., and Liu, J. L. (2002). "Study on reduction of collapse of eucalyptus wood in drying," *China Wood Ind.* 16, 3-6.[Chinese]
- Kim, S., Kim, J. A., Kim, H. J., Lee, H. H., and Yong, D. W. (2006). "The effects of edge sealing treatment applied to wood-based composites on formaldehyde emission by desiccator test method," *Polym. Testing* 25, 904-911.
- Li, K. C., Peshkova, S., and Geng, X. L. (2004). "Investigation of soy protein-kymene[®] adhesive systems for wood composites," *JAOCS* 81, 487-491.
- Li, K. F., Hou, X. J., Xu, S. H., and Peng, W. X. (2005). "Effect of test methods on

- plywood formaldehyde emission content,” *China Wood Ind.* 19, 30-32.[Chinese]
- Martinez, E., and Belanche, M. I. (2000). “Influence of veneer wood species on plywood formaldehyde mission and content,” *Holz. Roh. Werkst.* 58, 31-34.
- Park, B. D., and Kim, J. W. (2008). “Dynamic mechanical analysis of urea–formaldehyde resin adhesives with different formaldehyde-to-urea molar ratios,” *J. Appl. Polym. Sci.* 108, 2045-2051.
- Qian, X. Y. (2012). “Transforming and upgrading of the Chinese wood-based panel industry,” *China Wood Ind.* 26, 1-6.[In Chinese]
- Salem, M. Z. M., Bohm, M., Srba, J., and Berankova, J. (2012). “Evaluation of formaldehyde emission from different types of wood-based panels and flooring materials using different standard test methods,” *Build. Environ.* 49, 86-96.
- Shi, S. Q., and Gardner, D. J. (2001). “Dynamic adhesive wettability of wood,” *Wood Fiber Sci.* 33, 58-68.
- Schafer, M., and Roffael, E. (2000). “On the formaldehyde release of wood,” *Holz. Roh. Werkst.* 58, 259-264.
- Wei, S. Y., Shi, J. Y., Gu, J. Y., Wang, D., and Zhang, Y. H. (2012). “Dynamic wettability of wood surface modified by acidic dyestuff and fixing agent,” *Appl. Surf. Sci.* 258, 1995-1999.
- Zhang, H., Zhang, J., Song, S. P., Wu, G. F., and Pu, J. W. (2011). “Modification of nanocrystalline cellulose from two kinds of modifiers used for improving formaldehyde emission and bonding strength of urea-formaldehyde resin adhesive,” *BioResources* 6, 4430-4438.

Article submitted: January 6, 2013; Peer review completed: February 22, 2013; Revised version received and accepted: March 31, 2013; Published: April 5, 2013.