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RAPID COMMUNICATION

Shin-ichiro Tohmura · Kohta Miyamoto · Akio Inoue

Acetaldehyde emission from glued–laminated timber using phenol–resorcinol–formaldehyde resin adhesives with addition of ethanol

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Introduction

Volatile organic compounds (VOCs) from wood-based materials have been a topic of great concern as one of the causes of sick building syndromes for decades. According to an investigation of indoor air quality of new residences by the Japanese Ministry of Land, Infrastructure and Transport, about 9% of all residences exceeded the guideline value of 48 μg/m^3 (0.03 ppm) of acetaldehyde concentration.[1] Other studies have also shown excessive acetaldehyde concentration in indoor air in wooden houses.[2,3] Most natural wood has a certain level of acetaldehyde emission (5–130 μg/m^{2}h after 1 day) measured by small chamber methods.[4,5] The amount of acetaldehyde emission is considered to depend on the wood species, although the mechanism of acetaldehyde emission has been unclear. Recently there have been multiple reports in which extraordinary acetaldehyde emissions (600–1500 μg/m^{2}h) from glued–laminated timbers and laminated veneer lumbers bonded with phenol–resorcinol–formaldehyde (PRF) resin adhesives have been measured using small chamber methods.[2,6,7] These values are much higher than the natural acetaldehyde emission from wood. However, as yet, we have not found any feasible answer regarding the origin of the acetaldehyde. It is necessary to elucidate the source of the extraordinary acetaldehyde emission as soon as possible. In general, two types of commercial PRF resins containing methanol and/or ethanol have been used. In addition, radio frequency or cold press techniques are commonly used in the laminating process in the industry. In this study, therefore, the effect of PRF type, press condition, and wood species on acetaldehyde emission from glued–laminated timber was investigated using the small chamber method.[8]

S. Tohmura (✉) · K. Miyamoto · A. Inoue
Forestry and Forest Products Research Institute, Tsukuba 305-8687, Japan
Tel. +81-29-873-3211; Fax +81-29-874-3720
e-mail: tomura@ffpri.affrc.go.jp

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Materials and methods

Sample sizes and parameters used in this study are summarized in Table 1. The laminas used were made of kiln-dried sugi (*Cryptomeria japonica*) from Kyushu in Japan and Douglas fir (*Pseudotsuga menziesii*) from the United States. Lamination numbers of sugi and Douglas fir were four and five, respectively. The methanol-added PRF (M-PRF) and ethanol-added PRF (E-PRF) resins were prepared by adding 7% of methanol or ethanol to an alcohol-free PRF resin synthesized by Oshika Corporation. The alcohol-added PRF resins were mixed with a curing reagent (15% to the resin), and then spread. The spread amount was 300 g/m^2. Cold pressing was performed at 25°C for 16 h under 0.98 MPa and 1.18 MPa for sugi and Douglas fir, respectively. In the radio frequency heat press (0.3 A for 6 min), press pressure was the same as that used in the cold press. All laminas and glued–laminated timbers were conditioned in a ventilated room at 25°C for 1 week. Each product was wrapped separately in a polyethylene bag and placed in a 20°C conditioned room for about 1 month until the emission tests. Determination of acetaldehyde emission from the glued–laminated timbers was performed according to the JISA 1901 small chamber method.[8] In the small chamber system (ADPAC system, Adtec), the temperature was 28°C, the relative humidity was 50%, and ventilation rate was 0.5 1/h. Before the test, the surface of the timber was cut and planed to the appropriate size for each method (Table 1). Cross sections of the glued–laminated timber and laminas were sealed by aluminum tape and the total exposure area was fixed at 432 cm^2. Samples of the air were

Table 1. Sample code and manufacturing condition of glued–laminated timbers

Sample code	Wood name	Adhesive type	Press method	Sample size L/W/D (cm)[a]
SL	Sugi	–	–	14.0/12.5/3.0
SMR	Sugi	M-PRF	Radio frequency	12.0/11.5/9.2
SMC	Sugi	M-PRF	Cold press	12.0/11.5/9.2
SER	Sugi	E-PRF	Radio frequency	12.0/11.5/9.2
SEC	Sugi	E-PRF	Cold press	12.0/11.5/9.2
DL	Douglas fir	–	–	15.2/12.0/2.3
DMR	Douglas fir	M-PRF	Radio frequency	12.0/11.5/9.2
DMC	Douglas fir	M-PRF	Cold press	12.0/11.5/9.2
DER	Douglas fir	E-PRF	Radio frequency	12.0/11.5/9.2
DEC	Douglas fir	E-PRF	Cold press	12.0/11.5/9.2

PRF, phenol–resorcinol–formaldehyde resin; M-PRF, methanol-PRF; E-PRF, ethanol-PRF
[a] Shows dimensions in length/width/depth

Table 2. Acetaldehyde emission factor from laminas and glued–laminated timbers bonded by PRF resins

Time	Sugi					Douglas fir				
	Lamina	M-PRF		E-PRF		Lamina	M-PRF		E-PRF	
	SL	SMR	SMC	SER	SEC	DL	DMR	DMC	DER	DEC
1 day	0	5	30	1006	863	11	26	14	552	224
3 days	1	4	17	759	723	4	17	9	408	147
7 days	1	4	12	626	627	2	13	8	314	104
14 days	1	4	8	629	595	1	10	7	278	89

Emission factors given in $\mu g/m^2h$

taken by 2,4-dinitrophenylhydrazine cartridge (LpDNPH-S10, Supelco) at 1, 3, 7, and 14 days after sample setting. The amounts of formaldehyde and acetaldehyde were determined by a high performance liquid chromatography (HPLC) on a SIL-AD10vp HPLC system (Shimadzu).

For additional experiments to investigate the interaction between ethanol and various materials, sugi lamina, Douglas fir lamina, filter paper (No.1 240 mm, Advantec), and glass (petri dish 100 mm) were selected. Distilled ethanol (0.8 g) was spread on the surface of the materials using a paintbrush. Immediately after painting, each sample was tested by the chamber method. An air sample (1.0 liter) was collected after 1, 3, 18, 50, and 170 h.

Table 3. Acetaldehyde emission factor after ethanol addition to the surface of various materials

Time	Wood		Filter paper	Glass
	Sugi	Douglas fir		
1 h	8 989	272	5	9
3 h	16 504	484	5	5
18 h	12 246	388	8	1
50 h	4 430	238	0	0
170 h	475	103	0	0

Emission factors given in $\mu g/m^2h$. Ethanol addition: 0.8 g

Results and discussion

Acetaldehyde emission factors of laminated timbers determined by the small chamber method are shown in Table 2. Extraordinary amounts of acetaldehyde, 800–1000 $\mu g/m^2h$ (sugi) and 200–500 $\mu g/m^2h$ (Douglas fir), were observed after 1 day in the case of the glued–laminated timber using ethanol-PRF resin. These phenomena were noticeably independent of the species and press condition.

Questions remain as to when and how ethanol converts to acetaldehyde, or whether acetaldehyde already exists in the PRF resin before curing. PRF resins in this study were diluted and reacted with 2,4-dinitrophenylhydrazine solution, then directly analyzed by HPLC. An extremely small amount of acetaldehyde was detected in the PRF resin itself. This means that acetaldehyde did not exist before the spreading of the PRF resin. Therefore, acetaldehyde production is most likely caused by the interaction between ethanol and wood during the curing process.

Some evidence indicating acetaldehyde production by the combination of ethanol and wood is shown in Table 3. We found that a huge amount of acetaldehyde is produced when ethanol is added to the surface of wood and no other materials. In addition, acetaldehyde production rapidly takes place within a few hours after setting in the chamber.

Therefore, we conclude that the source of the extraordinary acetaldehyde emission from PRF resin adhesive is ethanol contained in the PRF resin. The production of acetaldehyde appears to be caused by a certain interaction (possibly oxidation) between the ethanol and the wood.

Further experiments to elucidate the mechanisms of the acetaldehyde production from ethanol and wood are now being conducted.

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