Effects of Ethanol-Supplemented Phenol-Resorcinol-Formaldehyde Resin on Formaldehyde and Acetaldehyde Emissions from Glued-Laminated Timbers*

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Abstract

Aldehyde emissions from glued-laminated timbers bonded with phenol-resorcinol-formaldehyde (PRF) resin adhesives, some of which included ethanol as a solvent, were investigated. Four commercial ethanol-free and three laboratory-made PRF resins with varying ethanol content were prepared and used to manufacture sugi (*Cryptomeria japonica* D. Don) glued-laminated timbers. The aldehyde emissions from these timbers were measured by a Japanese Industrial Standard small chamber method over an extended period. Three test samples, each with a differently oriented sealed surface, were also prepared for discussion of the mechanism. No relationship was observed between formaldehyde emission and either ethanol content or the orientation of the exposed area. However, acetaldehyde emission was significantly influenced by both parameters. When a cross-sectional surface was exposed, acetaldehyde emission decreased rapidly; however, conversely, when a cross-sectional surface was sealed, acetaldehyde emission remained constant and even increased gradually over an extended period, which was attributed to excessive acetaldehyde. When ethanol was present in adhesives in excess of 5 percent of the resin weight, excessive acetaldehyde may have been produced by enzyme alcohol dehydrogenase as previously reported and may spread into the wood, whereupon the acetaldehyde may have been emitted mainly from the cross-sectional surface rather than the other sides.

Volatile organic compounds (VOCs), including aldehydes from wood-based materials, remain a topic of great concern as one of the causes of sick building syndrome (Takeda et al. 2009; Takigawa et al. 2009, 2010), although findings of VOC emission have declined year-by-year over the past decade. Formaldehyde emission has been successfully decreased by technical improvements of adhesives. However, because acetaldehyde was originally not used as a raw material of adhesives, the source of acetaldehyde in indoor air was not identified. Acetaldehyde has been identified as a possibly carcinogenic compound for humans by the International Agency for Research on Cancer (IARC 1999). Furthermore, an acetaldehyde concentration for indoor air of less than 48 $\mu g/m^3$ has been announced in the Japanese guidelines of indoor air quality (Ministry of Health, Labour and Welfare 2002). This value is, in fact,

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lower than the 100 μg/m^3 guideline for formaldehyde. Wood is one of the possible sources of acetaldehyde because some amount of acetaldehyde is produced naturally via a metabolic pathway (Kimmerer and MacDonald 1987; Kimmerer and Stringer 1988; Kreuzwieser et al. 1999, 2001). However, the acetaldehyde emission rates from several solid wood samples have been measured to be less than 5 μg/m^2/h after 7 days by a small chamber method (Tohmura et al. 2005b). In the heat treatment of wood during drying, emissions of various VOCs including acetaldehyde have been detected, and the amounts of VOCs have been found to increase in proportion to both drying temperature and time (Ishikawa et al. 2009). The authors reported that 40 mg of acetaldehyde was emitted from 1 m^3 of sugi (*Cryptomeria japonica* D. Don) during drying for 11 minutes at 160°C.

Acetaldehyde emissions from wood-based materials, particularly glued-laminated timber bonded by phenol-resorcinol-formaldehyde (PRF) adhesives, were found in indoor air, sometimes in extraordinary amounts (Matsuda et al. 2004, Tamura 2004, Yagi et al. 2004). It was discovered that the interaction between the wood and ethanol included as a solvent in some PRF adhesives triggered acetaldehyde generation (Tohmura et al. 2005a). Subsequently, we proposed the hypothesis of an enzymatic oxidation mechanism involving alcohol dehydrogenase (ADH) and clarified the existence of both original ADH existing in wood right after harvest and induced ADH on the surface of processed wood in response to reaction with the ethanol added as a solvent in adhesives or paints (Tohmura et al. 2012). Although most commercial PRF resins manufactured in Japan are ethanol free, some PRF resins still contain a small component of ethanol to enhance the smoothness of spreading and other benefits. However, the effect of ethanol content in the PRF resins on acetaldehyde emissions from wood-based materials remains unclear. Accordingly, we initially verified the absence of acetaldehyde emissions from glued-laminated timbers bonded with commercial ethanol-free PRF resins and subsequently attempted to elucidate the quantitative relationship between ethanol addition and acetaldehyde emission with ethanol-containing PRF resins. In this study, we evaluate the hypothesis that acetaldehyde emission from glued-laminated timber is not observed if ethanol is not contained in the adhesive. Furthermore, the pathway of acetaldehyde emission depending on the exposure direction is investigated.

Materials and Methods

Ethanol-free PRF resin adhesives

Four commercial ethanol-free PRF resins (denoted A, B, C, and D) were provided by the Thermosetting Plastic Resin Association of Japan and applied to kiln-dried–grade sugi laminas provided from a commercial product. The laminas were cut to dimensions of 400 mm (longitudinal [L]) by 120 mm (tangential [T]) by 20 mm (radial [R]) with a moisture content of approximately 11.5 percent. A single glued-laminated timber was manufactured for each test condition, and the test specimens were produced from five laminas. PRF resin was mixed with a curing reagent (15% to 30% relative to the resin weight, according to the instructions), mainly comprising paraformaldehyde and fillers. The PRF resin spread rate was 300 g/m^2. The closed assembly time, press pressure, pressing temperature, and pressing time were 15 minutes, 0.68 MPa, 25°C, and 24 hours, respectively. One control sample consisting of a glued-laminated timber without resin (GLS) was also prepared by assembling the laminas according to the exact same procedure without the application of an adhesive. The glued-laminated timbers were wrapped in aluminum foil and placed in a 20°C and 65 percent relative humidity air-conditioned room for 1 day, whereupon specimens were cut to a length of 108 mm and a width of 100 mm from each timber. Two cross sections of each specimen corresponding to each test were then sealed with an aluminum tape to form exposure areas of 43,200 mm^2 each (100 by 108 by 4 mm).

Ethanol-containing PRF resin adhesives

To investigate the effect of ethanol content on aldehyde emissions, ethanol-containing PRF resin adhesives were prepared in a laboratory using an ethanol-free PRF resin sample provided from Oshika Corporation as the base. Three grades of ethanol-containing PRF resins were prepared by adding and mixing pure ethanol at 1, 5, and 10 percent by weight to the ethanol-free PRF base resin. As for the ethanol-free PRF adhesives, sugi laminas were also used with a moisture content of approximately 11.5 percent and size of 400 (L) by 120 (T) by 30 (R) mm. The test specimens were made from four laminas. The original ethanol-free resin, used as a control, and the ethanol PRF resins were mixed with 15 percent (relative to resin weight) of curing agent, as previously described, and applied to the lamina with a spread rate of 300 g/m^2. The pressing conditions, the specimen size, and the manner of conditioning were the same as for the ethanol-free PRF adhesives. Two glued-laminated timber samples per condition were manufactured.

Preparing specimens with different exposure direction areas

Laminas were provided by Meiken Lamwood Corporation as kiln-dried sugi. Only knot-free sugi laminas were chosen to eliminate the effect of knots on the aldehyde emission pathways. The moisture content of the laminas was approximately 11.5 percent, and the laminas were cut to dimensions of 400 by 120 by 30 mm. The test specimens were made from four laminas. The 10 percent ethanol-containing PRF resin was mixed with the curing reagent as described previously and applied to the laminas with a spread rate of 300 g/m^2. The closed assembly time was 15 minutes. Cold pressing was performed at 30°C for 24 hours under 0.68 MPa. Two glued-laminated timber samples per condition were manufactured. Each product was cut as cubic specimens measuring 106 by 106 by 106 mm, whereupon the specimens were separately wrapped in a polyethylene bag and placed in a 20°C, 65 percent relative humidity, air-conditioned room for 1 day. In total six specimens, each with an exposure area of 45,000 mm^2, were prepared from clear laminas and sealed with aluminum tape in the three different exposure directions, cross section (CS) sealed, radial section (RS) sealed, and tangential section (TS) sealed, as shown in Figure 1.

Aldehyde emission measurement

Measurements of acetaldehyde emissions from the glued-laminated timbers were subjected to the Japanese Industrial Standard (JIS) A1901 small chamber method (JIS 2009). In

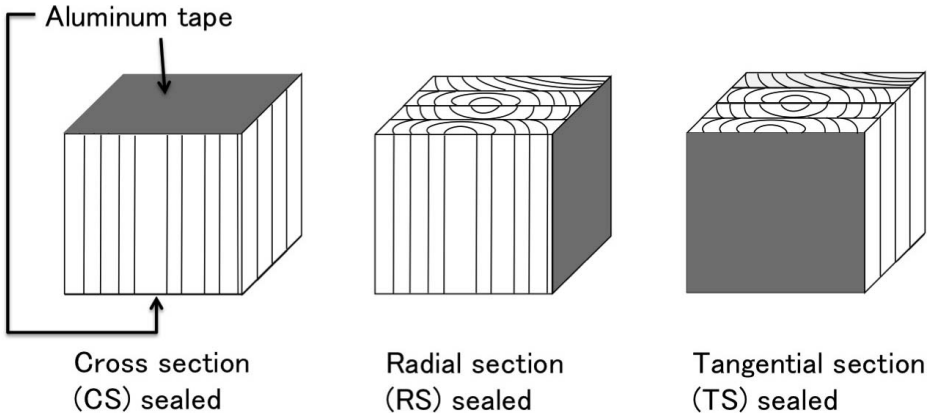

Figure 1.—Specimens of glued-laminated timber with different exposure direction areas for the small chamber method.

the 20-liter small chamber system (ADPAC system, ADTEC), the temperature was maintained at 28°C, the relative humidity at 50 percent, with a ventilation rate of 0.5 h^{-1}. The loading factor was 2.16 m^2/m^3 and 2.25 m^2/m^3 for ethanol-free PRF resin series and ethanol-containing PRF resin series, respectively. The 5 liters of air in the chamber was sampled through a 2,4-dinitrophenlyhydrazine (DNPH) cartridge (GL Science) using a sampling pump operating at 167 mL/min. Sampling was performed on days 1, 3, 7, 14, 28, and 125 after sample placement in the chamber.

Determination of aldehydes

Aldehydes were determined according to the International Organization for Standardization (ISO) standard method (ISO 2001). The aldehyde derivatives in the DNPH cartridge were dissolved and diluted with high-performance liquid chromatography (HPLC)–grade acetonitrile into a volumetric flask up to 5 mL. A 20-μL DNPH derivative solution was then injected into an HPLC system (Shimadzu, SIL-AD10vp HPLC system). HPLC separations were achieved on two octadecyl-silica (ODS) columns (STR-ODS II, 150 by 4.6 mm inside diameter, Shinwa Chemical Industries, Ltd.). The temperature of the column chamber in the HPLC system was maintained at 40°C, and a solvent gradient flowing at 1.0 mL/min was provided. For the first 5 minutes of the gradient program, the solvent was acetonitrile–distilled water (40/60). The acetonitrile content increased linearly to 100 percent during the next 25 minutes of the program and remained at 100 percent for 5 minutes. The acetonitrile–distilled water ratio then reverted to 40/60 for the next 1 minute and remained at this level until the end of the 50-minute analysis. The wave range of the ultraviolet detector in the HPLC system was set at 360 nm. The amounts of formaldehyde and acetaldehyde were determined by the standard 13-carbonyl compounds (Supelco, CARB 1004 DNPH Mix2).

Results and Discussion

Effect of ethanol-free PRF resin

The ethanol-free PRF resins provided by four different domestic glue companies were applied to the manufacture of sugi glued-laminated timbers. We attempted to clarify in this experiment whether or not ethanol alone is a trigger of acetaldehyde emission. The ethanol-free PRF sample emission rates of formaldehyde and acetaldehyde are shown in Figures 2 and 3, respectively. The code GL refers to glued-laminated timber, while the letters A, B, C, and D refer to the commercial PRF resin from each resin manufacturer, and S refers to sugi lamina laminated without adhesive as a control.

Formaldehyde emissions from GLB and GLD specimens were initially 680 and 583 $\mu g/m^2/h$, respectively; they rapidly decreased to below 100 $\mu g/m^2/h$ within 3 weeks as shown in Figure 2. This was probably because the formaldehyde emissions were trapped immediately following production by wrapping the timber specimens. Formaldehyde emissions from GLA and GLC specimens behaved nearly the same as that from the GLS specimen. The observed differences in the two classes of emission levels might be related to the curing rate. A practical industrial manufacturing process involves much more postcure conditioning time after pressing; therefore, the extent of formaldehyde emission should be considerably lower than the values determined in the present study.

On the other hand, although acetaldehyde emissions from all ethanol-free adhesive–bonded samples remained under 5 $\mu g/m^2/h$ and were all higher than the GLS sample, the level is very low and near the detection limit of 3 $\mu g/m^2/h$, as

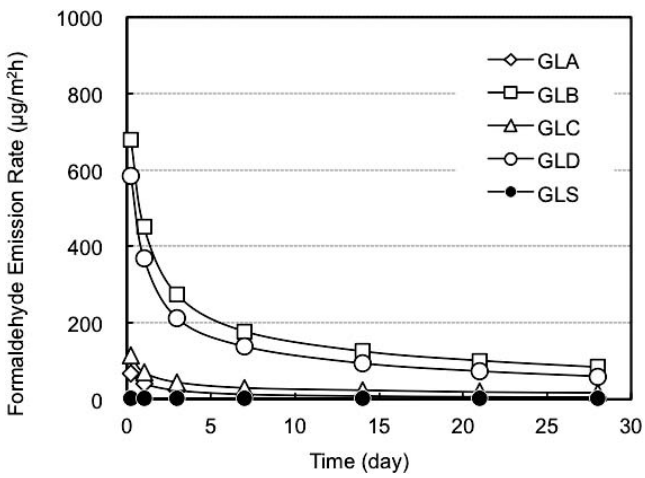

Figure 2.—Eeffect of ethanol-free phenol-resorcinol-formaldehyde (PRF) resins on the formaldehyde emission rate of glued-laminated timber. GLA = glued-laminated timber for which the PRF resin of company A's product was used, etc.; GLS = glued-laminated timber without adhesive.

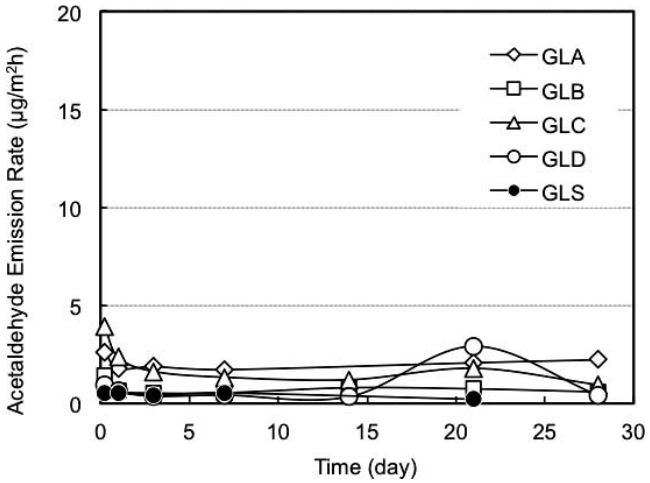

Figure 3.—Effect of ethanol-free phenol-resorcinol-formaldehyde (PRF) resins on the acetaldehyde emission rate of glued-laminated timber. GLA = glued-laminated timber for which the PRF resin of company A's product was used, etc.; GLS = glued-laminated timber without adhesive.

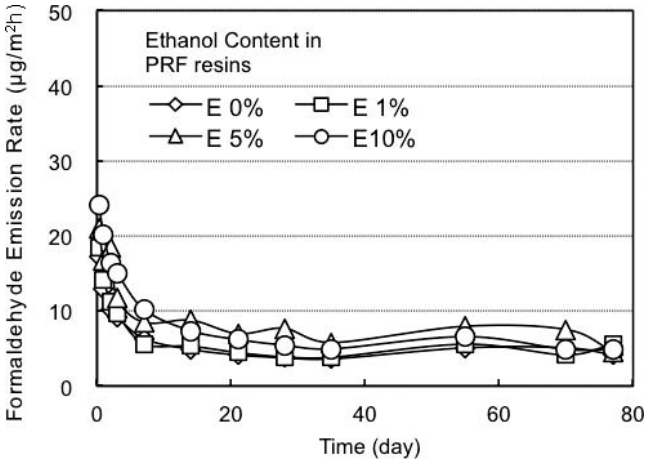

Figure 4.—Time intervals of formaldehyde emission rate of glued-laminated timber manufactured with the ethanol-supplemented phenol-resorcinol-formaldehyde (PRF) resins by the chamber test.

shown in Figure 3, indicating no substantial difference from the GSL solid sample. Therefore, we reconfirmed previous findings that ethanol-free PRF resins produce no acetaldehyde in glued-laminated timber (Tohmura et al. 2005a, 2012).

Effect of ethanol content of PRF resins

Three ethanol-supplemented PRF resins were prepared to elucidate the effect of the ethanol content on acetaldehyde emission. Glued-laminated timber bonded with ethanol-supplemented PRF resins was measured using the same small chamber process. The emission behaviors of formaldehyde and acetaldehyde are shown in Figures 4 and 5, respectively. The values are the average of two specimens. Although formaldehyde emission rates varied from 10 to 25 $\mu g/m^2/h$ in the early stages of the experiment, they decreased within a week. Furthermore, all emission rates were less than 10 $\mu g/m^2/h$ within 2 weeks, while continuing to decline to as little as 5 $\mu g/m^2/h$ over an extended period, as shown in Figure 4.

In contrast, the sample with no added ethanol demonstrated emission rates of acetaldehyde that were mainly zero, as shown in Figure 5. This is the same result as the case of ethanol-free PRF resins. Similarly, the sample laminated with PRF resin with 1 percent added ethanol also remained less than 5 $\mu g/m^2/h$. The results suggest that the addition of up to 1 percent ethanol maintains a low acetaldehyde emission level.

The acetaldehyde emission rates from the 5 and 10 percent ethanol–containing PRF resin samples were initially 125 and 142 $\mu g/m^2/h$, respectively, and decreased rapidly within a week. However, they subsequently increased and remained at approximately 70 to 100 $\mu g/m^2/h$ for an extended period. The behavior of the acetaldehyde emission looks similar for both 5 and 10 percent ethanol–containing PRF resin samples. This suggests that the addition of 5 percent ethanol may represent the saturation amount for the capacity of producing an ADH action. We assume that ADH-converting action occurs inside the wood on or in close proximity to an adhesive bondline of a glued-laminated timber. It is expected that the acetaldehyde gradually diffuses to the sample edge. Migration of acetaldehyde in the radial and tangential sections of the wood might be slower than that in the cross section of the wood, as discussed below. When the ADH reaction occurs near the adhesive bondline, most of the generated acetaldehyde is quickly emitted to the outside along the bondline. A reaction occurring inside the wood would require more time to diffuse. Additionally, the diffusion of acetaldehyde from lamina to lamina is probably blocked by the cured adhesive layer after adhesive curing.

To emphasize the effects of the ethanol content on formaldehyde and acetaldehyde emission, Figures 6 and 7 were composed from the same data given in Figures 4 and 5. The figures clarify that ethanol-containing PRF resins only affected acetaldehyde emissions and not formaldehyde emissions. The amount of acetaldehyde emission rose with the increasing content of ethanol in the PRF resins. The PRF resin containing a minimal ethanol content of 1 percent had no effect on acetaldehyde emissions, but an ethanol content

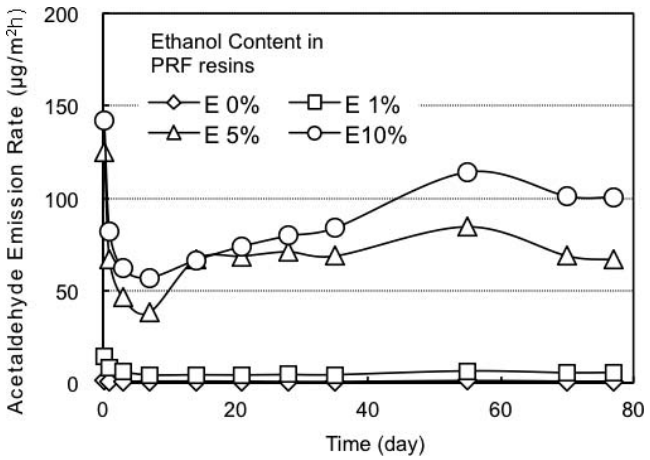

Figure 5.—Time intervals of acetaldehyde emission rate of glued-laminated timber manufactured with the ethanol-supplemented phenol-resorcinol-formaldehyde (PRF) resins by the chamber test.

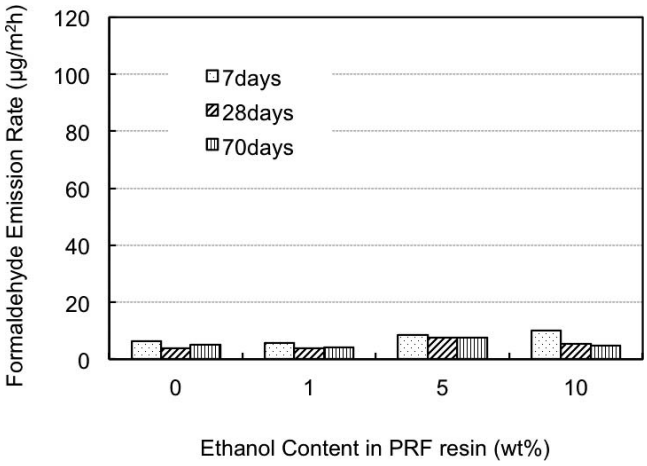

Figure 6.—Effect of ethanol content of phenol-resorcinol-formaldehyde (PRF) resin on the formaldehyde emission rate of glued-laminated timber.

exceeding 5 percent caused acetaldehyde emissions to rise and remain at a high level for an extended period. Acetaldehyde emission increased continuously even over 70 days. In general, aldehyde and VOC emissions decrease over time in a ventilated chamber. The continuously increasing emission rate resulted because the supply amount exceeded the exhaust amount in the ventilation system. The gradual, long-term emission of acetaldehyde is considered to be related to emission pathways in the glued-laminated specimens.

Effect of exposure direction

According to a previous article (Tohmura et al. 2012), when ethanol came into contact with wood, acetaldehyde was produced. This was attributed to an alcohol dehydrogenase enzymatic action mechanism. When the ethanol was applied to the adhesive as a solvent, the acetaldehyde was produced from the adhesive bondline and remained in the glued-laminated timber for an extended period. Aldehyde measurement from glued-laminated timber is usually performed using the small chamber method and by sealing

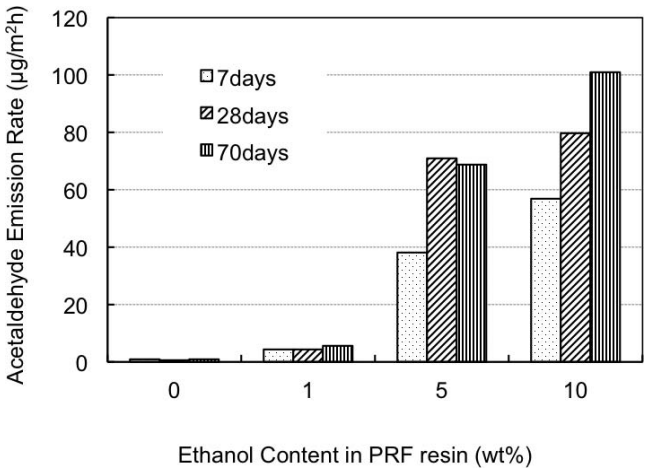

Figure 7.—Effect of ethanol content of phenol-resorcinol-formaldehyde (PRF) resin on the acetaldehyde emission rate of glued-laminated timber.

both cross-sectional sides (Matsuda et al. 2004, Tamura 2004, Yagi et al. 2004). That is because it is assumed that areas other than the cross-sectional sides will be exposed to room interiors when a glued-laminated timber is used indoors. In general, moisture and gas inside the wood move in the direction of the fibers and are then emitted from the cross-sectional area of the timber to the outside. Therefore, if the cross section is sealed completely, acetaldehyde should remain in the wood, and emissions will likely decline.

We therefore attempted to clarify the diffusion route of acetaldehyde occurring inside wood by sealing the different exposure areas of cubic glued-laminated timber samples. The three samples used were the CS, RS, and TS samples shown in Figure 1.

The effect of the exposure direction of glued-laminated timbers on the emission behavior of formaldehyde is shown in Figure 8. The values are the average of two specimens. Formaldehyde from RS and TS samples exhibited emissions of 164 and 231 $\mu g/m^2/h$, respectively, on the first day and then similarly decreased by one-half within 3 weeks. The CS sample exhibited an emission of 58 $\mu g/m^2/h$ at the beginning and decreased below 15 $\mu g/m^2/h$ after 3 weeks. These results indicate that formaldehyde is emitted more easily in the case of the cross-sectional side than the radial- and tangential-sectional sides.

In contrast, the effect of the exposure direction of glued-laminated timbers on acetaldehyde emission behavior is shown in Figure 9. The acetaldehyde emission rate of the specimen for which the cross-sectional side was sealed increased gradually over time, peaked, and then gradually declined. Conversely, acetaldehyde emission rates of the specimens for which the cross-sectional side was not sealed, the TS and RS samples, exhibited completely opposite behavior, where an initially large emission declined dramatically and then gradually decreased over an extended period. It is clear that the acetaldehyde generated by conversion of ethanol in the adhesive bondline tended to decrease promptly via diffusion from the cross-sectional side. Moreover, when the cross-sectional side is sealed, the

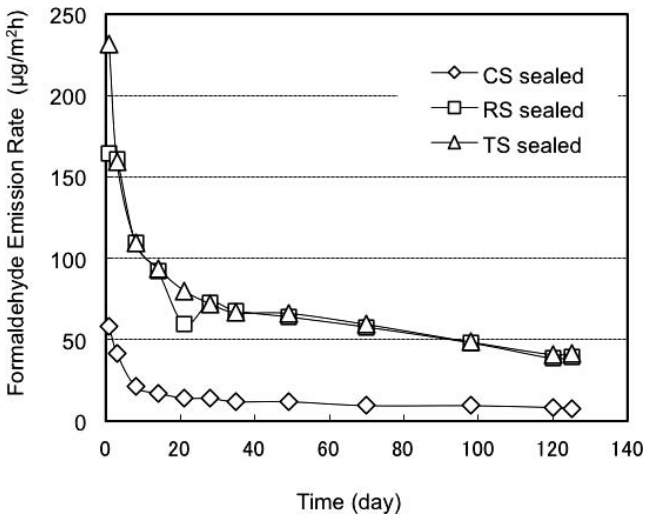

Figure 8.—Effect of the exposure direction on the formaldehyde emission rate of glued-laminated timber. The ethanol content of the phenol-resorcinol-formaldehyde resin was 10 percent. CS = cross section; RS = radial section; TS = tangential section.

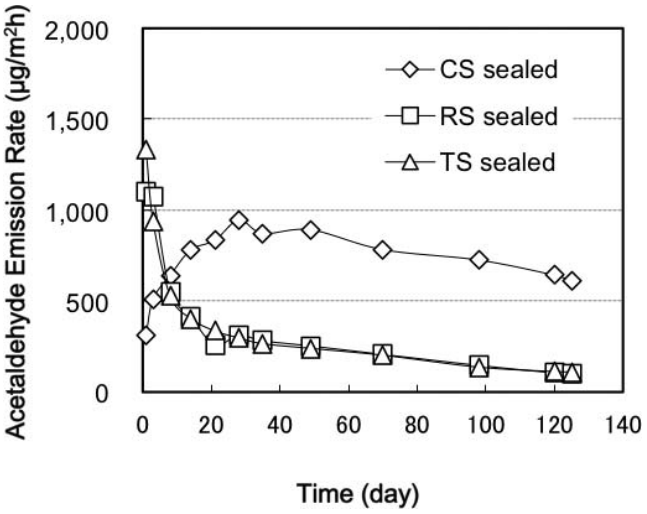

Figure 9.—Effect of the exposure direction on the acetaldehyde emission rate of glued-laminated timber. The ethanol content of the phenol-resorcinol-formaldehyde resin was 10 percent. CS = cross section; RS = radial section; TS = tangential section.

acetaldehyde generated inside is trapped and remains in the wood for an extended period. Although initial emissions are low, acetaldehyde gradually diffuses through the wood composite over time, whereupon emissions will continue gradually from the tangential section and/or the radial section sides over an extended period.

The maximum acetaldehyde emission rate values of 1,300 μg/m^2/h at the initial stage shown in Figure 9 and of 142 μg/m^2/h at the initial stage shown in Figure 5 represent substantial differences for samples glued with the same 10 percent ethanol PRF resin. This is probably because the wood samples differed in the degree of ADH. This indicates that an estimation of acetaldehyde emission for constant ethanol amounts is unreliable. Some effective treatments for reduction in the acetaldehyde emission induced by ethanol addition have been reported. One method involves heat treatment (Ishikawa et al. 2009, 2014) and another is a sterilization treatment (Tohmura et al. 2012). Both treatments enable deactivation of ADH.

Conclusions

We verified that acetaldehyde was detected when ethanol-containing adhesives were used to manufacture glued-laminated timber from sugi wood. The effect was completely different from the case of formaldehyde emissions. No relationship between the orientation of the exposed wood surfaces of the glued-laminated timbers and formaldehyde emission was observed. When a cross section of the glued-laminated timber was exposed, acetaldehyde decreased quickly. Conversely, when a cross section was sealed, acetaldehyde remained constant and even gradually increased over an extended period, which was attributed to excessive acetaldehyde. If an emission resource such as ethanol exists in adhesives by an amount of 5 percent or more relative to resin weight, excessive acetaldehyde may be produced by enzyme ADH, as previously reported, and spread into the wood. In this case, the acetaldehyde might be emitted mainly from the cross section and not from the other sides.

Acknowledgments

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