Eur. J. Wood Prod. (2011) 69: 311–316
DOI 10.1007/s00107-010-0446-6

ORIGINALS ORIGINALARBEITEN

Empirical correlations between test methods of measuring formaldehyde emission of plywood, particleboard and medium density fiberboard

Byung-Dae Park · Eun-Chang Kang · Sang-Bum Park · Jong Young Park

Received: 28 May 2009 / Published online: 18 May 2010

Abstract Since different test methods of measuring the formaldehyde emission (FE) from wood-based composite panels have been used for different countries and regions, this study attempted to establish empirical correlations between three test methods (i.e., 24-hour desiccator, 1 m^3 chamber, and perforator) for plywood (PLW), particleboard (PB), and medium density fiberboard (MDF), particularly emphasizing on correlations between the 24-hour desiccator and the 1 m^3 chamber method. The desiccator method found statistically high correlations with other two methods, resulting in regression coefficient values ranging from 0.96 to 0.88 for PLW, PB, and MDF samples. In particular, the desiccator method had an empirically high correlation with the 1 m^3 chamber method that had been adopted as the reference method of comparing regionally different test methods of measuring the FE of wood-based composite panels by the ISO/TC89.

Empirische Korrelationen zwischen Prüfmethoden zur Messung der Formaldehydabgabe von Sperrholz, Spanplatten und mitteldichten Faserplatten

Zusammenfassung Da in verschiedenen Ländern und Regionen unterschiedliche Prüfmethoden zur Messung der Formaldehydabgabe (FE) von Holzwerkstoffplatten verwendet werden, wurde in dieser Studie der Versuch unternommen, empirische Korrelationen zwischen den drei Prüfmethoden, 24-Stunden Desiccator, 1 m^3 Prüfkammer und Perforator, für Sperrholz (PLW), Spanplatten (PB) und mitteldichte Faserplatten (MDF) zu bestimmen. Dabei wurde besonderes Gewicht auf die Korrelation zwischen dem 24-Stunden Desiccator-Verfahren und dem 1 m^3 Prüfkammer-Verfahren gelegt. Das Desiccator-Verfahren war statistisch hoch mit den beiden anderen Verfahren korreliert. Die Regressionskoeffizienten der PLW-, PB- und MDF-Proben lagen zwischen 0,96 und 0,88. Insbesondere das Desiccator-Verfahren wies eine hohe Korrelation mit dem 1 m^3 Prüfkammer-Verfahren auf, das von ISO/TC89 als Referenzverfahren zum Vergleich regional unterschiedlicher Prüfverfahren zur Messung der Formaldehydabgabe von Holzwerkstoffen übernommen worden war.

1 Introduction

Various reconstituted wood panel products such as plywood (PLW), particleboard (PB), medium density fiberboard (MDF) and so on, have become increasingly popular, and are being used for manufacturing furniture, cabinets, or various building products. These products are mainly bonded with formaldehyde-based resin adhesives such as urea-formaldehyde (UF) resin, melamine-urea-formaldehyde (MUF) resin, phenol-formaldehyde (PF) resins, etc. In particular, UF resin possesses some advantages such as fast curing, good performance in the panel, water solubility and lower price. In spite of these advantages, two main disadvantages of using UF resin are formaldehyde emission (FE) from the panels and lower resistance to water. The FE of wood-based panel products has been

B.-D. Park (✉)
Department of Wood Sci. & Tech., Kyungpook National University, Daegu, 702-701, Republic of Korea
e-mail: byungdae@knu.ac.kr

E.-C. Kang
R&D Team, Sunchang Enterprise, Incheon, 604-050, Republic of Korea

S.-B. Park · J.Y. Park
Division of Environmental Wooden Material Engineering, Korea Forest Research Institute, Seoul, 130-712, Republic of Korea

received great attention from public as well as the wood industries since formaldehyde has been known as a toxic air contaminant. In fact, the International Agency for Research on Cancer (IARC), a part of the World Health Organization (WHO) reclassified formaldehyde as 'carcinogenic to human (Group 1)' from 'probable carcinogenic to human (Group 2A)' in June 2004. In addition, an effort to reduce energy losses in houses after the energy crisis in the 1970s subsequently resulted in an increase of air tightness of indoor environment, which accelerated the FE problem. In 1980s, the first guideline for FE was adopted in Germany (Marutzky and Margosian 1995), and its regulation has been implemented in many regions around the world such as Japan, Taiwan, and California in the United State of America.

FE was one of the most important aspects of UF resin in the last few decades. Amino resins such as UF resin, MUF resin, etc. are mainly responsible for the formaldehyde emission resulting from wood-based composite panels bonded with them. A few excellent reviews on this matter have been published (Meyer and Hermanns 1985; Myers 1984). The susceptibility of UF resins to hydrolysis explains the formaldehyde emission of UF resins (Dunky 1998). The lack of water resistance of UF resin adhesives also limits the use of wood-based composite panels bonded with the UF resin adhesives to non-structural and interior applications.

There are many factors affecting the FE of wood panel products (Marutzky 1989). For example, many variables such as temperature, relative humidity, air exchange rate, loading ratio, etc. could affect the FE measurements of wood-based panel products (Myers and Nagaoka 1981; Myers 1983, 1984, 1985a, 1985b). Obviously, test methods such as the desiccator method, chamber method, perforator method, or gas analysis method provide different FE values for the same wood-based product (Myers 1983). The desiccator method has been widely used for the countries in the Asia-Pacific region, such as Korea, Japan, Malaysia, Indonesia, Australia, and New Zealand while the perforator method (also called extraction method) has been conventionally used in European countries. By contrast, a large chamber method is standardized in North America. Differences in details and procedures make it difficult to compare results between the methods. Thus, different test methods make it difficult for the manufacturers of wood-based composite panels to compare the FE values measured by other test methods.

So, many different attempts have been made to compare the FE or to establish correlations between methods (Lehmann 1982; Sundin et al. 1987; Risholm-Sundman and Wallin 1999; Wiglusz et al. 2000; Risholm-Sundman et al. 2005; Que et al. 2007; Que and Furuno 2007). Lehmann (1982) reported that the large chamber test was the most reliable and accurate method and was highly correlated with a small-chamber (19 L) method. But the author pointed out that there was need for a better method of determining FE for the quality control method on a day-to-day basis due to some limitations of the chamber methods. Sundin et al. (1987) also compared four different methods of testing the FE of particleboard, and found good relationships between the methods with correlation coefficients of greater than 0.9. In order to evaluate a simpler method, Risholm-Sundman and Wallin (1999) reported correlations between different methods (flask, desiccator, emission cell) and 1 m^3 chamber method with the best correlation between the emission cell method and the 1 m^3 chamber method. The authors also mentioned that the conditioning time should be maintained in a standardized way to obtain comparable results. Wiglusz et al. (2000) reported a significant variation in the FE measurements between laboratories, and concluded that the inter-laboratory bias was due to heterogeneities of chamber conditions such as volume, materials, sampling air, etc. Risholm-Sundman et al. (2005) reported that the variations between measured results were due to specific differences in test conditions. Bulian et al. (2003) also reported that the lack of certified reference material made it difficult to establish an inter-calibration between test methods.

Comparing six different test methods such as chamber (EN 717-1), gas analysis (EN 717-2), flask (EN 717-3), perforator (EN 120), desiccator (JIS A 1460), and small chamber (JIS A 1901), Risholm-Sundman et al. (2005) also pointed out the need for a harmonized testing method through an effort in the International Standardization Organization (ISO) activity. Que and Furuno (2007) established a correlation between the 24-hour desiccator and the large chamber (28.4 m^3) method, and confirmed that the formaldehyde concentrations of the chamber under conditions that simulated a mobile home environment were quite closely related to real formaldehyde levels.

An international effort to harmonize the comparison between test methods has been made by the Technical Committee 89 (TC89) of the International Standardization Organization (ISO) on wood-based composite panels. As a result, the 1 m^3 chamber method has been adopted as the reference method of measuring and comparing the FE of wood-based composite panels. So, this requires establishing correlations between the standards methods. This paper reports on a result of establishing empirical correlations between 24-hour desiccator methods and two different methods (1 m^3 chamber method and perforator) of measuring the FE of PLW, PB, and MDF panels.

2 Materials and methods

2.1 Materials

Wood-based composite panels used in this study were PLW, PB, and MDF, and they were supplied from local PLW, PB,

and MDF mills in the Republic of Korea. Three full-size (1200 mm × 2400 mm) panels of each PLW, PB, and MDF sample were used to select three different levels of FE of each panel type, which resulted in nine panels for each panel type. The total number of PLW, PB, and MDF were 27 panels. Namely, low (L), medium (M), and high (H) levels of FE values were in the range of below 0.5 mg/L (E_0 level), below 1.5 mg/L (E_1 level), and above 5.0 mg/L (E_2 level) based on the 24-hour desiccator method, respectively. The PLW panel used in this study was a concrete-forming PLW panel with 5-ply bonded with MUF resin adhesive, and had a great amount of the FE, which was used as the H level of the FE. Both the M and L levels of the FE of PLW samples were obtained by exposing the concrete-forming PLW samples to the controlled environment (20°C and 65% RH) for 1 month and 2 months, respectively. However, the FE levels of both PB and MDF samples were adjusted by using different UF resin formulations in their production lines in the mills. Nominal panel thicknesses of PLW, PB, and MDF were 12 mm, 12 mm, and 15 mm, respectively.

Test samples of each type of panel were cut into 500 mm × 500 mm samples at the mills, and then delivered to the laboratory. The delivered samples were wrapped with polyethylene film prior to further cutting into test specimens with a proper size for each test. The samples wrapped with the film were kept in a humidity-controlled room at 20°C and 65% relative humidity (RH). Depending on test methods, proper size and number of specimens were cut, and then these specimens were used for each test. But, the samples were directly used for the 1 m^3 chamber test without further cutting after conditioning without the film wrapping.

2.2 Test methods of formaldehyde emission

Methods used in this study were the 24-hour desiccator method (KS M 1998-4, 2005), perforator method (EN 120, 1991), and the 1 m^3 chamber method (ISO FDIS 12460-1, 2007). However, the perforator method was not used for the plywood samples, but for the PB and MDF samples.

2.2.1 24-hour desiccator method

For the 24-hour desiccator method (KS M 1998-4, 2005), nine specimens (150 mm × 50 mm) were placed on the specimen holder in a 10 L glass desiccator, which contained a crystallizing dish (12 cm × 6 cm) with 300 ml of distilled water. Then, the desiccator with the glass lid was allowed to stand for 24 hours at 20 ± 2°C. The dish was then removed and the water was used to analyze the formaldehyde concentration, which was expressed as mg/L of formaldehyde in water. Acetylacetone analysis procedures were employed to determine the formaldehyde concentration in the aqueous solution. The formaldehyde solution was mixed with the reagent of 25 ml acetylacetone-ammonium acetate solution. The resultant mixture was heated in a capped glass bottle for 10 min at 65°C, cooled to room temperature, and its absorbance was measured in a 1-cm cell at 412 nm with a UV–VIS spectrophotometer (Optizen 2120UV, Mecasys Co. Ltd., Seoul, Korea).

2.2.2 1 m^3 chamber method (ISO/FDIS 1246-1, 2008)

For the 1 m^3 chamber method, two specimens (0.5 m × 0.5 m × thickness) were conditioned in an environment-controlled room at 23 ± 1°C, 50 ± 5% relative humidity (RH) for 7 ± 2 days. These preconditioned test specimens with a total surface area of 1 m^2 were placed in a 1 m^3 chamber, which gave a loading ratio of 1.0 m^2/m^3. The edges of two specimens were sealed with aluminum foil to obtain a constant ratio of the length (l) of the open (unsealed) edges to the surface area (A), so that $l/A = 1.5$ m/m^2. The chamber was environmentally controlled by temperature (23 ± 0.5°C), RH (50 ± 3%), and the air exchange rate (1.0 h^{-1}). Formaldehyde emitted from the specimen test mixes with the air in the chamber. The air in the chamber is sampled periodically over a defined period. The formaldehyde concentration is determined by drawing air from the chamber through gas-washing bottles containing water, which absorbs the formaldehyde. The formaldehyde concentration in the water is determined. The concentration of formaldehyde in the chamber atmosphere is calculated from the concentration in the water in the gas washing bottles and the volume of the sampled air. It is expressed in mg/m^3. Sampling is periodically continued until the formaldehyde concentration in the chamber has reached a steady-state.

The formaldehyde concentration in the chamber air is measured repeatedly until a constant concentration is reached. The formaldehyde is collected in a gas-washing bottle and the concentration is determined photometrically by the acetylacetone method. The results were expressed as the formaldehyde (mg) in the air (m^3). The test usually takes two weeks, one week of conditioning and one week for testing.

2.2.3 Perforator method (EN 120)

For the perforator test (European Standard (EN) 120, 1991), approximately 100 g of 25 × 25 mm specimens were extracted with boiling toluene for 2 hours. Before refluxing toluene returned to the boiling pot, it was bubbled through distilled water that extracted any dissolved formaldehyde. The acetylacetone analysis procedures were also used to determine the formaldehyde concentration in water, which provided the perforator value, expressed as milligrams of free formaldehyde per 100 g of dry board. Testing parameters for different test methods are presented in Table 1.

Table 1 Test parameter comparison of different test methods for formaldehyde emission
Tab. 1 Vergleich der Versuchsparameter der verschiedenen Prüfverfahren zur Messung der Formaldehydabgabe

Parameters	Test method		
	Desiccator	1 m^3 chamber	Perforator
Total surface area (m^2)	1.8	1.0	100 g[a]
Edge exposed	Yes	Partly yes	Yes
Temperature (°C)	20	25	–
Relative humidity (%)	65	50	–
Air exchange rate (h^{-1})	–	1.0	–
Loading ratio (m^2/m^3)	–	1.0	–
Analysis method	Acetylaceton	Acetylaceton	Acetylaceton

[a]Dry mass of the specimen

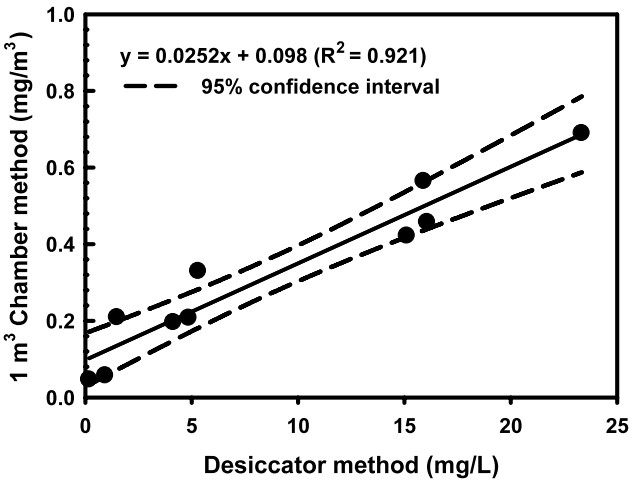

Fig. 1 Empirical correlation between the desiccator and 1 m^3 chamber method for plywood (Sample size, $n = 10$)
Abb. 1 Korrelation zwischen dem Desiccator-Verfahren und dem 1 m^3 Prüfkammer-Verfahren bei Sperrholz (Probenumfang, $n = 10$)

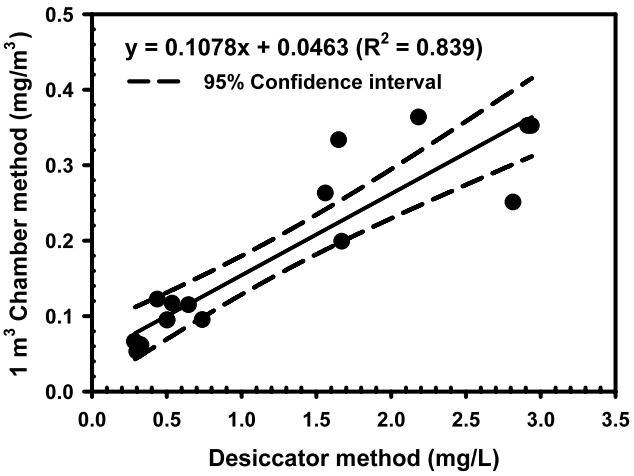

Fig. 2 Empirical correlation between the desiccator and 1 m^3 chamber method for PB (Sample size, $n = 17$)
Abb. 2 Korrelation zwischen dem Desiccator-Verfahren und dem 1 m^3 Prüfkammer-Verfahren bei Spanplatten (Probenumfang, $n = 17$)

3 Results and discussion

3.1 PLW panel

Figure 1 shows an empirical correlation between the desiccator method and the 1 m^3 chamber method for PLW panels. The perforator method was not used for the PLW samples because this method has been mainly used for PB and MDF in general. The result indicates that the FE value measured by the 1 m^3 chamber method corresponded to about 12% of the one measured by the desiccator method. The chamber method is quite different from the desiccator method in terms of air exchange. In other words, the chamber method measures the formaldehyde emitted from the surface of specimens under dynamic air circulation, or air exchange, which simulates an environment of indoor air conditioning.

As discussed below, the FE values of PLW samples measured by the 1 m^3 chamber method was lower than those of the PB and MDF samples. The FE levels of PLW samples shown in Fig. 1 were much greater than those of PB and MDF because concrete-forming PLW panels were used.

3.2 PB panel

Figure 2 shows an empirical correlation between the desiccator method and the 1 m^3 chamber methods for PB panels. Although four measurements were outside the 95% confidence interval of the linear regression, a positive correlation with a R^2 value of 0.839 was found between the desiccator method and 1 m^3 chamber (ISO) method. The FE values of PB measured by the desiccator method were about ten times greater than those of the ISO method. In other words, about 1 mg/l level of the desiccator measurement corresponded to about 0.1 mg/m^3 level of the ISO method, which gave a linear correlation.

An empirical correlation between the desiccator method and the perforator method for PB panels is presented in Fig. 3. The formaldehyde content measured by the perforator method was about five times greater than those FE values of the desiccator method. These differences could be at-

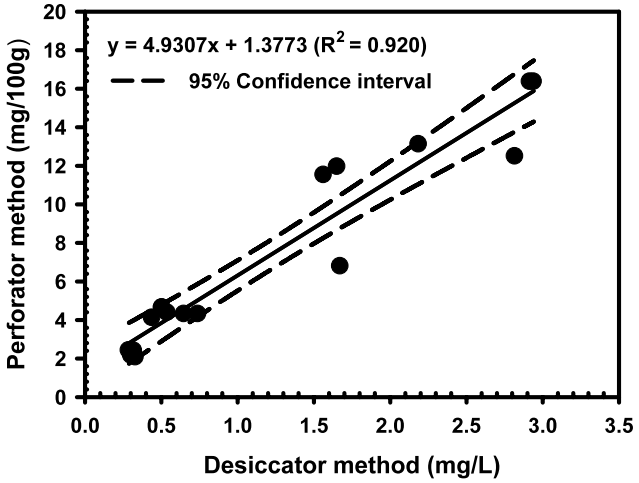

Fig. 3 Empirical correlation between the desiccator and perforator method for PB (Sample size, $n = 17$)
Abb. 3 Korrelation zwischen dem Desiccator-Verfahren und dem Perforator-Verfahren bei Spanplatten (Probenumfang, $n = 17$)

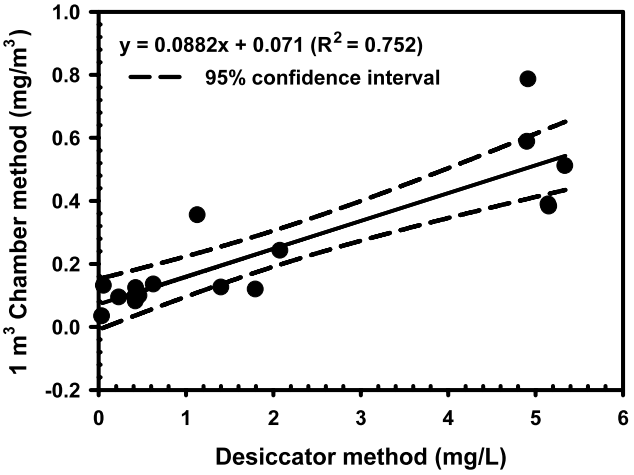

Fig. 4 Empirical correlation between the desiccator and 1 m^3 chamber method for MDF (Sample size, $n = 17$)
Abb. 4 Korrelation zwischen dem Desiccator-Verfahren und dem 1 m^3 Prüfkammer-Verfahren bei MDF (Probenumfang, $n = 17$)

tributed to an inherent difference between the two test methods. In other words, the perforator method measures the total extractable content of formaldehyde present in the samples, while the desiccator method and two other chamber methods measure the amount of formaldehyde emitted out of the panel surfaces. In spite of these differences between two methods, a strong positive correlation was found for PB panels. A linear regression of the formaldehyde values measured by these two methods had a R^2 value of 0.92 with four measurements located outside the 95% confidence interval. Compared to the ISO method, the perforator method showed a narrower 95% confidence interval and higher R^2 value, which indicated a good correlation.

3.3 MDF panel

Figure 4 shows an empirical correlation between the desiccator and the 1 m^3 chamber method for MDF panels. A linear regression with a R^2 value of 0.734 resulted in an empirical correlation between two methods. This result indicates that a 1 mg/l level of FE measured by the desiccator method is equivalent to about 0.089 mg/m^3 level of the 1 m^3 chamber method. As discussed later, the regression coefficient of this correlation was relatively lower than those of other correlations.

The perforator method also found a positive correlation with the desiccator method for MDF samples (Fig. 5). A linear regression also provided an empirical correlation with a regression coefficient of 0.954. This result indicates that about 1 mg/l level of the FE value measured by the desiccator method was equivalent to about 8.0 mg/100 g of the perforator method. This could be explained by the differences between the two methods. In other words, the desiccator method measures the formaldehyde emitted from the

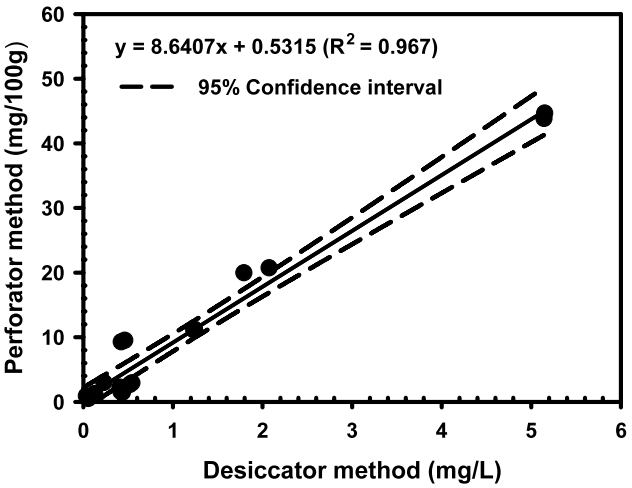

Fig. 5 Empirical correlation between the desiccator and perforator method for MDF (Sample size, $n = 17$)
Abb. 5 Korrelation zwischen dem Desiccator-Verfahren und dem Perforator-Verfahren bei MDF (Probenumfang, $n = 17$)

surfaces of specimens and dissolved in the distilled water, while the perforator method measures the total formaldehyde content in the specimen. This difference could explain the reason why the formaldehyde content by the perforator method was much greater than those FE values of the desiccator method.

4 Conclusion

This study showed all positive and empirical correlations between the desiccator and two other methods for PLW, PB and MDF panels. An emphasis was placed on the correlation between the 24-hour desiccator and the 1 m^3 chamber

method which was adopted as the reference method by the ISO/TC89.

The 24-hour desiccator method value showed good correlations with the 1 m^3 chamber method for PLW, PB and MDF samples, resulting in lower emission values for the chamber method than those of the desiccator method. Both 1 m^3 chamber method for PLW samples provided FE values corresponding to about 2% of the desiccator method, while the chamber method for PB samples resulted in FE values corresponding to about 11% of the desiccator method. For MDF samples, FE values of the 1 m^3 chamber method corresponded to about 14% of the desiccator method. The perforator method for both PB and MDF samples gave much greater FE values than those of the desiccator method. Although this study found good empirical correlations, theoretical correlations between test methods are necessary.

Acknowledgement The authors appreciate partial financial support from the Ministry of Environment, Republic of Korea for a part of this work (plywood experiment).

References

Bulian F, Battaglia R, Ciroi S (2003) Formaldehyde emission from wood based panels: The importance of intercalibrating the test methods. Holz Roh- Werkst 61:213–215

Dunky M (1998) Urea-formaldehyde (UF) adhesive resins fro wood. Int J Adhes Adhes 18:95–107

Lehmann WF (1982) Correlations between various formaldehyde tests. In: Proc of the 16th international symposium on particleboard. Washington State University, Pullman, pp 35–44

Marutzky R (1989) Release of formaldehyde by wood products. In: Pizzi A (ed) Wood adhesives: chemistry and technology, vol 2. Dekker, New York, pp 307–387

Marutzky R, Margosian R (1995) Measuring formaldehyde emissions from pressed wood products: an international perspective. In: Measuring and controlling volatile organic compound and particulate emissions from wood processing operations and wood-based products. The Forest Products Society, Madison, pp 62–73

Myers GE (1983) Formaldehyde emission from particleboard and plywood paneling: measurement, mechanism and product standards. For Prod J 33(5):27–27

Myers GE (1984) Effect of ventilation rate and board loading on formaldehyde concentration: a critical review of the literature. For Prod J 34(10):59–68

Myers GE (1985a) Effect of temperature and humidity on formaldehyde emission from UF-bonded boards: a literature critique. For Prod J 35(9):20–31

Myers GE (1985b) Effect of separate conditions to furnish or veneer on formaldehyde emission and other properties: a critical review. For Prod J 35(6):57–62

Meyer B, Hermanns K (1985) Formaldehyde release from pressed wood products. In: Turoski V (ed) Formaldehyde: analytical chemistry and toxicology. Advances in chemistry series, vol 210. American Chemical Society, Washington, pp 101–116

Myers GE, Nagaoka M (1981) Emission of formaldehyde by particleboard: effect of ventilation rate and loading on air-contaminating levels. For Prod J 31(7):39–44

Que Z, Furuno T (2007) Formaldehyde emission from wood products: relationship between the values by the chamber method and those by the desiccator test. Wood Sci Technol 41:267–279

Que Z, Furuno T, Katoh S, Nishino Y (2007) Evaluation of three test methods in determination of formaldehyde emission from particleboard bonded with different mole ratio in the urea-formaldehyde resin. Build Environ 42:1242–1249

Risholm-Sundman M, Wallin N (1999) Comparison of different laboratory methods for determining the formaldehyde emission from three-layer parquet floors. Holz Roh- Werkst 57:319–324

Risholm-Sundman M, Larsen A, Vestin E, Weibull A (2005) Formaldehyde emission—a comparison of different standard methods. In: Proc Indoor Air, pp 1925–1930

Sundin EB, Mansson B, Endrody E (1987) Particleboard with different contents of releasable formaldehyde: a comparison of the board properties including results from four different formaldehyde tests. In: Proc of the 21st international particleboard/composite materials symposium. Washington State University, Pullman, pp 139–186

Wiglusz R, Jarnuszkiewicz I, Sitko E, Nikel G (2000) Interlaboratory comparison experiment on the determination of formaldehyde emitted from mineral wood board using small test chambers. Build Environ 35:53–57