Emission of formaldehyde by particleboard: effect of ventilation rate and loading on air-contamination levels

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Abstract

Dynamic tests for determining the formaldehyde emission behavior of UF-bonded boards involve the measurement of formaldehyde concentration in the air within a vessel whichcontains a specified board loading L (m^2 of board area per m^3 of vessel free volume) and is being ventilated at a specified air exchange rate N (hr.$^{-1}$). Such tests constitute a primary characterization of board emission behavior since they model conditions for air contamination in dwellings containing those boards. Many other board emission tests, e.g. Desiccator, Perforator, must be regarded as secondary tests since their results cannot a priori be related to use conditions; to be accepted as valid quality control tests, they must first be correlated with primary test results. Existing approaches toward explaining the dependence of formaldehyde concentration upon ventilation rate and loading in a dynamic test are reviewed Data for three particleboards are fit by a two-parameter equation in which the reciprocal of steady state formaldehyde Concentration is linearly related to the ratio of ventilation rate to loading, N/L. According to one interpretation, the intercept of a plot of reciprocal Concentration versus N/L is a measure of the equilibrium concentration (C_{eq}) in the absence of any air exchange, while the slope measures the rate of formaldehyde transport through the board-air interphase in terms of a mass transfer coefficient K. On this basis, reductions in board emission can be accomplished by reducing C_{eq}, e.g., by stabilizing the resin, and/or by reducing K, e.g., by coating the board surfaces. Examples are given to show how this equation can be used to evaluate the degree of improvement required for a board to meet permissible air contamination levels and for evaluating the utility of a board under various ventilation rate and loading conditions.

In recent years a great deal of effort has been devoted to the problem of formaldehyde liberation from wood products bonded with urea-formaldehyde (UF) adhesives. That effort was necessitated by increasing reports of discomfort and possible health effects induced by formaldehyde gas in the air within buildings constructed partially of such bonded wood products. Other materials, such as UF foamed insulation and resin-treated carpeting and fabrics, have been cited as potential sources for formaldehyde contaminated air.

Such reports have led to increasing attention from various governmental agencies and to the likelihood of government imposed limitations upon the permissible concentration of formaldehyde in the air. For example. the State of Wisconsin is expected to impose in 1981 a limit of 0.4 ppm of formaldehyde in the air of new mobile homes.

In a room exposed to wood products which emit formaldehyde, e.g., particleboard in floors or cabinets or hardwood plywood paneling, the air concentration of formaldehyde will be dependent at constant humidity and temperature upon the specific emissivity of the board (rate of formaldehyde emission per unit board weight or area), the amount, of board exposed to the room, the room volume, and the rate of ventilation with fresh air.

The need to understand the influence of these variables quantitatively is important for three reasons: 1) to translate laboratory measurements of board emission behavior into the potential consequences for air pollution levels in dwellings, thus providing meaningful evaluations of chemical or processing changes in terms of air pollution goals, 2) to balance the improvements in formaldehyde/air levels which might be achieved from increased ventilation rates against the resultant higher energy requirements, and 3) to balance

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the improvements which might be achieved from using lowered amounts of UF-bonded material against resultant higher costs or loss of desirable properties.

In this paper we describe current theories which attempt to relate formaldehyde air concentration to the variables mentioned. We evaluate those theories in the light of data obtained with three different particleboards and illustrate the potential utility of one equation which provides an excellent quantitative description of the data.

Background

There is abundant evidence that a constant rate of formaldehyde emission (mL/hr.–m^2) from UF-bonded particleboard into the well-mixed air of a chamber will lead to a constant level of formaldehyde (C in ppm formaldehyde, i.e., mL formaldehyde per 10^6 mL air at STP) in the chamber air (3. 5, 10, 14). The emitted formaldehyde results from residual “free” formaldehyde within the board and from formaldehyde produced by hydrolysis of the UF adhesive in the board. In the long run these sources will be depleted, leading to a decreased rate of emission and a lower level of formaldehyde contamination in the chamber air.

This paper, however, is concerned with shorter time periods, e.g., weeks, wherein the emission rate and resultant air contamination areeffectively constant. We also consider here only conditions of constant temperature and humidity. It should be noted, however, that C will be increased approximately 10 percent for every 10 percent increase in humidity and approximately 180 percent for every 10°C increase in temperature (10).

Within such constraints, if the chamber is not ventilated a true equilibrium concentration of formaldehyde in the air (C_{eq}) will be achieved. With ventilation, however, a lower steady state concentration (C_s) will be established which is dependent upon: 1) the ventilation rate N (hr.$^{-1}$, i.e., the number of total air exchanges per hour), 2) the surface area A (m^2) of exposed board, and 3) the chamber free volume V(m^3).

The simplest relation that has been applied (12) to describe the dependence of C_s upon these variables is

$$C_s=\frac{kA}{NV}=k\cdot\frac{L}{N} \qquad [1]$$

where L is termed the loading (m^2/m^3) and k is the board’s specific emission rate (mL formaldehyde per hour per m^2 of exposed board).

In contrast, Andersen, Lundquist, and Molhave (1) have proposed the following empirical expression for particleboard at constant temperature and humidity,

$$C_s=\frac{kA}{k'A+NV}=\frac{kL}{k'L+N} \qquad [2]$$

which differs from Equation [1] by incorporating the term $k'L$, where k' has the units m/hr. Inversion of Equation [2] leads to the relation

$$\frac{1}{C_s}=\frac{k'}{k}+\frac{1}{k}\cdot\frac{N}{L} \qquad [3]$$

indicating that C_s is a function not of N or L separately, but of their ratio and that the quantity k/k' should be identical to the equilibrium concentration C_{eq} at zero ventilation rate Nor infinite loading L. Inversion of the simpler Equation [1] shows that itdiffers from Equation [3] by having a zero intercept, i.e., by the requirement that C_{eq} go to infinity at zero Nor infinite L.

Two groups (3, 6) have used somewhat different approaches to derive theoretical equations for both the transient and steady state conditions. Fujii, Suzuki, and Koyagashiro (3) assumed that formaldehyde is liberated into the chamber air space at a rate given by kA and is removed from the air space by two processes: ventilation at rate NVC, and reabsorption by the board at rate $k'AC$. At steady state, then,

$$NVC_s+k'AC_s=kA \qquad [4]$$

which rearranges to Equations [2] and [3]. This approach, therefore, explicitly incorporates a dynamic interchange of formaldehyde between board and air, and characterizes that interchange by the two rate constants k and k’. As noted, the ratio of the two opposing rate constants is the equilibrium concentration C_{eq}.

In contrast, Hoetjer (6) employs a material transport approach and assumes first that the driving force for liberation of formaldehyde into the air at any time is given by ($C_{eq}-C$), i.e., by the magnitude of the concentration difference from the maximum possible value C_{eq}.

This driving force is moderated by a board-air interphase material transfer coefficient or permeability K (m/hr. or m/s. in Hoetjer’s formulation) such that the rate of transport from board to gas is KA ($C_{eq}-C$). Formaldehyde is again lost from the system by ventilation at rate NVC. Therefore, at the steady state

$$NVC_s=KA\,(C_{eq}-C_s) \qquad [E]$$

which rearranges to

$$C_s=\frac{KC_{eq}}{K+\frac{N}{L}}=\frac{C_{eq}}{1+\frac{1}{K}\frac{N}{L}} \qquad [6]$$

or

$$\frac{1}{C_s}=\frac{1}{C_{eq}}+\frac{1}{KC_{eq}}\cdot\frac{N}{L} \qquad [7]$$

Thus Fujii’s k and k' are, respectively, Hoetjer’s KC_{eq} and K. Hoetjer points out that Equation [6] makes it clear that the effect of N/L is to reduce C_{eq} to an extent controlled by K.

In an attempt to define the underlying mechanisms more completely we have derived an equation which explicitly includes formaldehyde diffusion and reversible reaction processes within the board, in addition to reversible, unhindered interchange between formaldehyde in board and air. Additional details are not presented here because the transient equation is very cumbersome and does not predict any steady state conditions except for rather short-lived ones at very low ventilation rates.

It is interesting to speculate in this connection that, as suggested by others (8), there may be different emission rate-limiting steps at low and very high ventilation rates. At high ventilation rates the formaldehyde may be removed from the boards so rapidly that its concentration within the board becomes

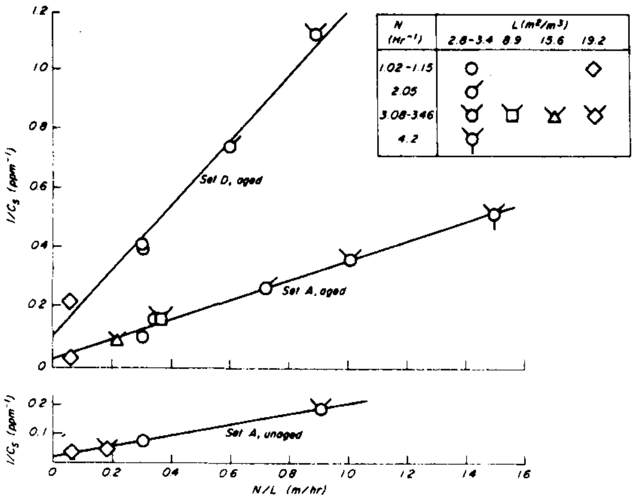

Figure 1. – Test of Equation [7] (*N,* ventilation rate (air exchange/hr.); *L.* loading (m^2 of panel/m^3 free air volume); and C_s, steady state level of formaldehyde contamination in air (ppm)).

depleted, the rate-limiting step becomes formaldehyde diffusion or generation within the board, and a steady state concentration in air cannot be maintained.

This situation may, therefore. correspond to the conditions underlying our own derivation. Its occurrence would be more likely if the board possessed an inherently low supply or generation rate of formaldehyde. In contrast, at low ventilation rates the emission rate-limiting process may be formaldehyde diffusion through a thin gaseous layer immediately adjacent to the board surface (6); thus the formaldehyde concentration is not depleted within the board and a steady state concentration can be maintained in the air.

This last interpretation is consistent with the observation (10) that at a quite low value of *N/L* (0.060 m/hr.), the amount of formaldehyde lost by some particleboard samples is a very small fraction of the potentially available amount (~0.2% of total original content) even after dynamic experiments have continued for 10 weeks.

Moreover, the interpretation is consistent with Hoetjer's view (6) that the emission driving force is moderated by an interphase permeability or material transfer coefficient K. For this reason, the ensuing discussion will emphasize Hoetjer's Equations [6] and [7] with parameters K and C_{eq} rather than the Fujii Equations [2] and [3] and parameters *k* and *k'*.

Experimental

Details of particleboard preparation and themethod of measuring dynamic ($N>0$) formaldehyde air concentrations have been described (10). Board set A was prepared with a Douglas-fir furnish using 7 percent (dry furnish basis) of a commercial UF resin containing 1.5 percent free formaldehyde; emission data were obtained before and after board aging for 30 days at 60°C, low humidity, and high air exchange rates. Board set D was prepared with the same furnish and the same total amount of the UF resin to which 10 percent urea and 5 percent melamine had been added; emission data reported here were obtained only for aged D boards.

Dynamic tests were conducted in a nonsystematic order for *N* from about 1 to 4 air exchanges/hr., for *L* from about 3 to 19 m^2/m^3, and for *N/L* from about 0.06 to 1.5, using sealed-edge specimens (50 by 125 mm) contained in glass vessels (10). Steady state formaldehyde concentration (C_s) values reported for the various N/L are the means of several analyses of the effluent air stream over periods of at least 10 days after attaining steady state. Some of the data points reported (10) were obtained without stirring the gas in the vessel.

The rather large fluctuations about the mean C_s values observed under these conditions were subsequently reduced by stirring the chamber atmosphere with a magnetically driven paddle. All experiments reported here were performed at 40°C and 75 percent relative humidity (RH). The formaldehyde concentrations in the effluent air stream were dotermined by absorption of the formaldehyde into water and analyzing the water solution with either Purpald or acetylacetone procedures (10).

Results and discussion

Comparison of data with theory

The Fujii (3) and Hoetjer (6) approaches predict that $1/C_s$ should be a linear function of the ratio *N/L*, i.e., air exchange rate over loading, as in Equation [7]. Figure 1 shows the data for all three board sets plotted in that fashion. The lines through the data result from linear regression analysis; values of the various parameters are summarized in Table I. The Hoetjer equation obviously provides an excellent fit to the steady state formaldehyde concentration data.

Using the regression parameters, the predicted dependence of steady state concentration upon *N/L* has been calculated from the Hoetjer equation and the resultant curves are shown in Figure 2. The nonzero intercepts $1/C_{eq}$ in Figure 1, and the corresponding noninfinite intercepts (C_{eq}) in Figure 2, demonstrate the improvement of this equation over Equation [1]. The insert in Figure 2 illustrates the potential difference between C_s calculated by Equations [1] and [6].

Change in Hoetjer parameters with board type

In the last three columns of Table 1 the magnitudes of the derived parameters are given and are also shown relative to the values for unaged board set A. This set was prepared using a commercial UF resin containing 1.5 percent free formaldehyde. Aging of that set for 30 days at 60°C resulted in only moderate reductions in the transport parameter *K* and in C_{eq}. Additon of 10 percent urea and 5 percent melamine to the resin, however, caused much greater lowering of KC_{eq} and C_{eq}. Hoetjer (6) indicates that values for *K* between 0.04 and 2.0 m/hr. have been observed in his laboratory for uncoated particleboards.

The Hoetjer parameters are further compared in Table 2 with calculated values corresponding to several standard formaldehyde characterization tests in order

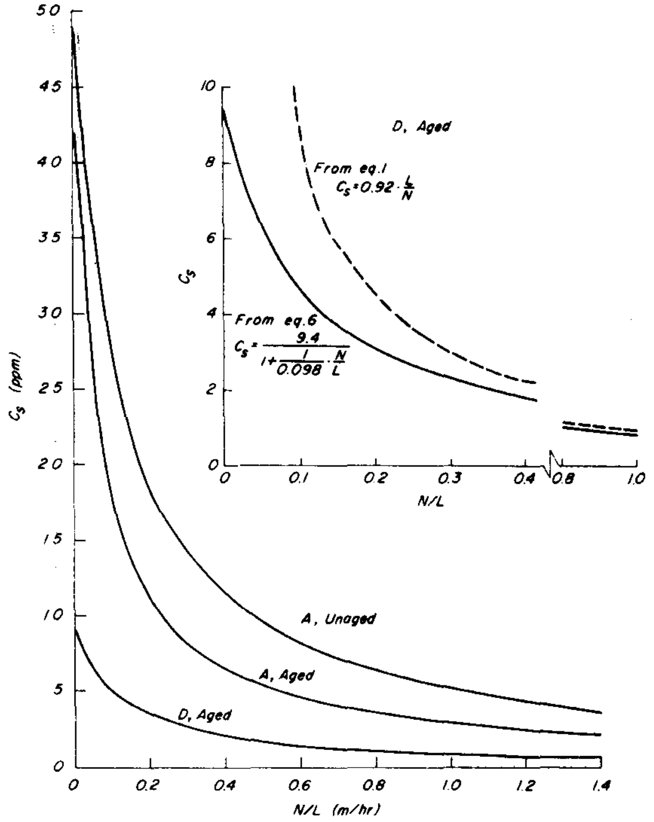

Figure 2. - Calculated dependence of C_s on *N/L* (*N*, ventilation rate (air exchange/hr.); *L*, loading (m^2 of panel/m^3 free air volume); and C_s, steady state level of formaldehyde contamination in air (ppm)).

to provide emission characteristics that are perhaps more familiar.

In previous work (1-), for example, it was shown that C, values at $N/L = 0.060$ were approximately linearly correlated with resin free formaldehyde content and with Japanese Industrial Standard (JIS) Desiccator (7) results; subsequent measurements (11) have in turn shown approximate linear logarithmic correlations between the JIS Desiccator values and both Perforator (13) and so-called Equilibrium Jar (4) values. These various correlations were employed to calculate the "test results" given in Table 2. The emission parameter C_{eq} in Table 1 appears to follow the patterns exhibited by the more conventional tests. However, *K* does not follow that pattern; to establish whether this is real or is due to experimental error will require much more data.

Board set A, either unaged or aged, is extremely poor in terms of its formaldehyde emission behavior, and the 1.5 percent free formaldehyde resin would undoubtedly not be employed currently to produce particleboard for use in mobile homes. The aged set D probably should be classified as marginal.

The JIS, for example, sets 5 µg/mL as a limit for the Desiccator value (7) while maximum permissible Perforator values range from 10-50 mg/100 g in Europe (15). The extent to which such a marginal board might be made acceptable by controlling loading or ventilation rate is considered next.

Practical utilization of Hoetjer equation

Figure 2 demonstrates that C_s falls rapidly as *N/L* goes from zero to about 0.2; thereafter the curves are relatively flat. The region of practical interest is actually from $N/L = 0.2$ to about 1.5 because: a) *L* for mobile homes, for example, is approximately 0.5-1.2 m^2/m^3 (2), and b) observed *N*'s for residences are generally in the range of 0.2-1.2 exchanges per hour (9), and there appears to be a growing tendency to accept approximately 0.5 $hr.^{-1}$ as a desirable compromise.

Table 3 presents some C_s values which were calculated for 25°C/75 percent RH within these ranges

TABLE 1. – *Parameters from Equation* [6].[a]

	Regression parameters					Derived parameters[b]		
Board set	Intercept $=1/C_{eq}$	Std. error of intercept	Slope $=1/KC_{eq}$	Std. error of slope	r^2	KC_{eq} (mL/m^2)	K (m/hr.)	C_{eq} (ppm)
A, unaged	0.0196	0.00540	0.180	0.0111	0.992	5.56 (1.0)	0.109 (1.0)	(51.0 (1.0)
A, aged	0.0229	0.00885	0.330	0.0120	0.992	3.03 (0.54)	0.069 (0.63)	43.7 (0.86)
	0.106	0.0350	1.09	0.0668	0.989	0.917 (0.16)	0.098 (0.90)	9.4 (0.18)

[a] $C = \frac{C_{eq}}{1 + \frac{1}{K}\frac{N}{L}}$

C_s = steady state concentration of formaldehyde in air (ppm)
N = ventilation rate ($hr.^{-1}$)
L = loading (m^2/m^3)
C_{eq} = equilibrium concentration (ppm) at $N = 0$
K = formaldehyde transfer coefficient (m/hr.).

[b] Numbers in parentheses are values relative to those for A, unaged.

TABLE 2. – *Calculated formaldehyde characteristics of boards.*

Board set	Resin	C_s at 40°C/75% RH and N/L=0.060[a]	C_s at 25°C/75% RH and N/L=0.060[b]	Hypothetical free formaldehyde in resin[c]	JIS Desiccator value[d]	Perforator value[e]	"Equilibrium" Jar value[f]
		(ppm)	(ppm)	(%)	(μg/mL)	(mg/100 g) board	(ppm)
A, unaged	UF	33	8.1	1.6	37	200	7.4
A, aged	Above	23	5.7	1.3	26	135	5.5
D, aged	Above + 10% urea + 5% melamine	5.8	1.3	0.81	5.8	27	1.5

[a] From Equation [6] of this paper.
[b] From equation: C_s (25°/75%) = −0.193 + 0.250 C_s (40°/75%); Ref. (10).
[c] From equation: Free formaldehyde = 0.652 + 0.122 C_s (25°/75%); Ref. (10).
[d] From equation: JIS = 0.14 + 4.50 C_s (25°/75%); Ref. (10).
[e] From equation: Log (Perforator) = 0.551 + 1.116 log (JIS); Ref. (11).
[f] From equation: Log (Jar) = −0.473 + 0.855 log (JIS); Ref. (11).

TABLE 3. – *Calculated C_s (ppm) at 25°C/75% RH.*[a]

Board set	Loading L(m^2/m^3)	Ventilation rate, N (hr.$^{-1}$)				
		0.2	0.5	1.2	3.0	10
A, unaged	0.5	2.7	1.3	0.55	0.23	0.07
A, aged	0.5	1.6	0.71	0.31	0.12	0.04
D, aged	0.5	0.46	0.21	0.09	0.04	0.01
A, unaged	1.2	5.0	2.6	1.3	0.53	0.14
A, aged	1.2	3.2	1.6	0.71	0.29	0.08
D, aged	1.2	0.87	0.45	0.21	0.09	0.02

[a] One-fourth of values calculated using Equation [6] and 40°C/75% parameters in Table 1.

of Nand L and at two higher N's (3 and 10 hr.$^{-1}$), using the Hoetjer parameters and equation with the additional fact that C_s values at 25°C/75 percent RH are approximately one-fourth those at 40°C/75 percent RH (10).

The lowest formaldehyde concentration which has been seriously proposed as a regulatory goal appears to be 0.1 ppm (14); that value, therefore, provides the most extreme test of the boards reported on here. Table 3 shows that at a ventilation rate of 0.5 hr.$^{-1}$ even the aged D set would require a loading well below 0.5 m^2/m^3 to achieve 0.1 ppm concentration.

Alternatively, the goal could be met at 0.5 m^2/m^3 loading by increasing the ventilation rate from 0.5 hr.$^{-1}$ to 1.2 hr.$^{-1}$; that achievement would entail an energy sacrifice, however, since airflow heat losses would be approximately doubled (9). At a loading of 1.2 m^2/m^3 energy losses would be further magnified by the necessity for a 3.0 hr.$^{-1}$ ventilation rate.

Thus, the previous conclusion that the best of the three board sets must be regarded as marginal is verified by these calculations. How much then must the aged D set be improved upon to meet the goal of 0.1 ppm at reasonable Nand L? Table 3 indicates that an improvement factor of 4.5 is required, for example, at $N = 0.5$ and L=1.2.

Following Hoetjer's viewpoint, there are two general means of achieving that improvement (6). In the first, the transfer coefficient K would be kept constant while C_{eq} was reduced by a factor of 4.5, e.g., by stabilizing the resin. In the second, C_{eq} would be kept constant while the transfer coefficient was reduced by a factor of 5.2, e.g., by coating the board surfaces with a chemically inert barrier.

Summary and conclusions

From the data reported herein, plus the observations of others (1, 3, 6, 14), it appears that formaldehyde air contamination by UF-bonded particleboard is dependent upon ventilation rate N and loading L through their ratio N/L. In the steady state, that dependence is very well described by the simple two parameter Hoetjer equation.

The extent of applicability of the dynamic air ventilation test and the Hoetjer equation needs to be established by examining a wider variety of UF-bonded boards, including hardwood plywood. It seems possible, for example, that boards possessing very low capacity to emit formaldehyde (low C_{eq}) may not produce steady state concentrations in air at reasonable N/L values. Once broadly established, this procedure and equation can be very useful in several ways:

a) They provide a means for evaluating, under admittedly idealized conditions, the potential benefits or sacrifices entailed in altering loading or ventilation rate.

b) They offer guidelines via the Hoetjer interpretation as to the mechanism of formaldehyde emission and directions for improvement.

c) They provide a method for characterizing board emission behavior in terms that have relatively direct significance for air contamination.

Dynamic tests are inherently more meaningful than are the simpler, quicker tests, e.g., Desiccator, which are performed under conditions that are far removed from those in actual service. These latter types of tests should properly be regarded as "secondary board characterization" methods, whereas the dynamic test constitutes a "primary board characterization."

However, it is highly probable that one of the "secondary" types of test will be adopted as a standard quality control test within the board manufacturing industry. To interpret those "secondary" data in terms of their consequences for air contamination, and thereby evaluate properly the ability of a board to allow a mobile home to meet an internal air standard, it will be essential to develop correlations between the "secondary" and the "primary" test results for a wide variety of boards.

Obviously, some caution must be exercised in extrapolating the results of the idealized dynamic test to actual dwellings where conditions may vary widely. Nevertheless, the dynamic test affords an essential bridge between "secondary" quality control tests and air contamination levels in real dwellings.

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