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Temperature and Humidity Dependence of Formaldehyde Release from Selected Building Materials

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The release of formaldehyde from a variety of consumer products has been measured by several laboratories using the Japanese Industrial Standard (JIS) desiccator procedure (Meyer 1979; Myers and Nagoka 1981a). This procedure involves the measurement of formaldehyde with the material under investigation at 100% relative humidity and at complete equilibrium with the environment. Although this method is very effective in producing relative release rate coefficients from materials of interest it suffers from some major drawbacks when attempts are made to apply such data to a real life situation such as those encountered in the residential setting. Static conditions with materials at 100% relative humidity in equilibrium are hardly every encountered in real life situations. These draw backs have been recognized and it has been suggested that information from dynamic chamber measurements is needed to describe more closely the situation observed in normal living environments (Pickrell et al. 1983; Myers and Nagoka 1981b; Pickrell et al. 1984). In a recent publication, dynamic chambers were used to measure the release of formaldehyde gas from a variety of materials common to a normal classroom environment over a temperature range from 20°C to 40°C.(Van Netten 1983) In another publication (Van Netten et al. 1988) the same chambers were used to monitor the release of formaldehyde and other volatile organic compounds from floors of different ages finished with a Swedish floor varnish. It was apparent from these studies that these controlled temperature dynamic testing chambers are an effective method of measuring formaldehyde release characteristics in real life situations as well as over a variety of conditions that simulate those encountered in the environment.

It is the intention of this studies to use this dynamic system to measure the formaldehyde release characteristics from a variety of suspected formaldehyde sources either because of their inherent formaldehyde content or because of previous exposure to urea formaldehyde foam used to insulate wall cavities.

It was also noted in the studies mentioned above that in certain trials formaldehyde release coincided with moisture release from the material under investigation and, as other studies have also stressed the importance of humidity (Pickrell et al. 1983) it was decided to measure formaldehyde release of each material at ambient as well as at elevated relative humidity levels.

MATERIALS AND METHODS

Samples of the materials to be investigated were cut, where possible, to a size of

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53.34 cm x 30.48 cm (i.e. 21" x 12"). A testing chamber, (dimensions 63.5 x 53.5 x 9.0 cm and total volume 30.57 litres) as described before (Van Netten 1983) was used to measure the temperature dependence of formaldehyde release. For those materials that were too large to fit inside the chamber mentioned before (in this case cement building block) a larger chamber was used (dimensions 63.5 x 53.5 x 37.8 cm, total volume 128 litres). The only modifications in both types of chambers was its stainless steel construction, rather than aluminum as used in the previous study as well as a small electric fan in order to ensure maximum mixing of the air. A continuous flow of air from outside the building was maintained at a rate of one litre per minute throughout all experiments. This was also the sampling rate and consequently corresponded to 1.96 and 0.47 air exchange per hour for the small and the big chambers respectively under empty conditions. The following procedure was followed for sample collection. A 15 minute control sample (chamber empty) was collected in 20 mls of a .1% sodium bisulfite impinger solution at 20C. The material to be tested was placed inside the chamber and allowed to equilibrate for 15 minutes after which a sample was collected for another 15 minutes. The temperature was raised to 30°C and, after an additional 15 minutes equilibration period, another 15 minute sample was taken. Again the temperature was raised by 10°C to 40°C followed by 15 minutes of equilibration and its corresponding 15 minute sample collection. The chamber was opened, and the material removed. After cooling and cleaning with 95% ethanol and distilled water the chamber was ready for the next testing regime but only after the control was taken. Formaldehyde analyses were made using the chromotropic acid method. (NIOSH 1977)

High humidity conditions within the chamber were achieved by moisturizing a second matched sample for each material with distilled water. All of these materials were kept under high humidity conditions for four to five hours prior to experimentation. Care was taken that no additional moisture was present during testing insuring a net transfer of moisture from the sample to the air within the chamber.

Materials that had been in contact with urea formaldehyde insulation (UFFI) in houses insulted with this material were cleaned with a steel brush in order to duplicate the method used by commercial UFFI removal companies. UFFI remnants were cleaned from the cement building block by means of sandblasting the internal and external surfaces. After cleaning all materials were stored in sealed plastic bags which were opened prior to analysis. The samples of the materials had the following sizes and description. Carpet, nylon 48.5 x 30.4 x 0.95 cm; ceiling tile, fibre board used, 5.8 x 30.5 x 1.27 cm; gypsum board, new, 55.8 x 30.5 x 1.27 cm; gypsum board, UFFI exposed, 57.8 x 30.5 x 1.27 cm; shiplap, fir, UFFI exposed, 57.15 x 36.8 x 1.9 cm; plywood, new, spruce exterior grade, 58.4 x 30.5 x 1.9 cm; terra cotta brick, UFFI exposed, 9.4 x 5.08 x 6.2 cm; lath and plaster, UFFI exposed, 33.0 x 33.0 x 1.58 cm; cement building block (3 hole), UFFI exposed, 41 x 20 x 10 cm; K-3 particle board, new, 58.4 x 30.5 x 1.9 cm. Although duplicate trials were performed on most of the materials i.e. carpet, plywood gypsum board (new), ceiling tile, this was not always possible for the UFFI exposed materials due to a lack of sample material. When duplicate samples were taken the values obtainer were within ±5% of the set of values reported here. During the analyses all materials were elevated from the bottom of the chamber by four aluminum support rings (2.54 cm high, 2.54 cm in diameter and with a .63 cm wall).

RESULTS AND DISCUSSION

All data gathered at ambient and high humidity levels are shown in graphical form in Figures 1 - 9. The relative humidity measurements for each of these trials is shown in a second graph below the one illustrating the formaldehyde measurements. The total surface area (T.S.A.), i.e. the sum of all external surfaces, is shown on the top graph for each of the materials.

Carpet. The nylon carpet sample investigated did not release formaldehyde in quantities higher than the detection limit, (0.1 ppm) and no graph was constructed. The carpet under investigation is therefore not a significant source of formaldehyde under the conditions described.

Ceiling Tile. At the two sets of humidities shown in Figure 1 there is an increase in formaldehyde release with an increase in temperature. The effect of humidity on formaldehyde release is insignificant at 20°C but doubles at the higher temperatures. The higher humidity conditions are associated with increased formaldehyde release.

Gypsum Board. As indicated in Figure 2 there is an increase in formaldehyde release with increasing temperature at both sets of humidities. In this case, however, high humidity results in a lower release of formaldehyde.

Gypsum Board (UFFI exposed). This material released considerably more formaldehyde (Figure 3) than the new gypsum board indicating that exposure to the foam will contaminate gypsum board. Again a high humidity results in a lower release of formaldehyde than what is observed at the ambient humidity levels.

Shiplap (UFFI exposed). Figure 4 shows that very little formaldehyde is released from shiplap under the conditions shown. Although the humidity levels at 40°C are identical, the formaldehyde release is quite different, illustrating the effect of moisture movement from sample to chamber (high humidity curve) on formaldehyde release. The slight increase in the lower humidity curve is probably due to an increase in humidity of outside air which is pumped through the chamber, allowing some transfer of moisture from atmosphere to sample.

Plywood (new exterior grade). This material had not been exposed to UFFI and any formaldehyde released by this material is entirely due to the use of formaldehyde based resins in its production. In this case (Figure 5), and in contrast to what was observed with the gypsum board trials, high humidity releases considerably more formaldehyde than the lower humidity.

Terra Cotta Brick (UFFI exposed). There were only a limited number of UFFI exposed half bricks available, for this reason the same bricks were used for both of the humidity trials. Although the formaldehyde release appears to be very low (Figure 6) this is not the case when one considers the small total surface area (TSA), 0.0275 m^2, compared to, for instance, plywood (TSA 0.3902 M^2). To compare these two items one tends to multiply small changes many times which increases the uncertainty. Better comparative results can be obtained with larger samples of comparable surface area. In these trials there appears to be a trend to release less formaldehyde at the higher humidity levels with increasing temperatures.

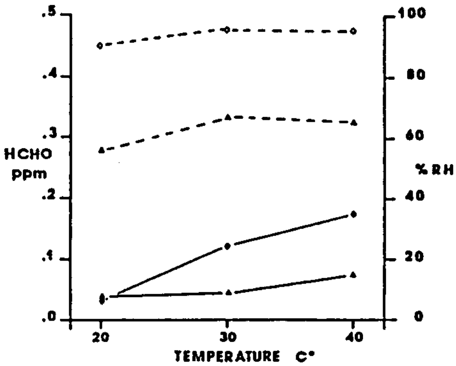

Figure 1. Formaldehyde release from ceiling tile.

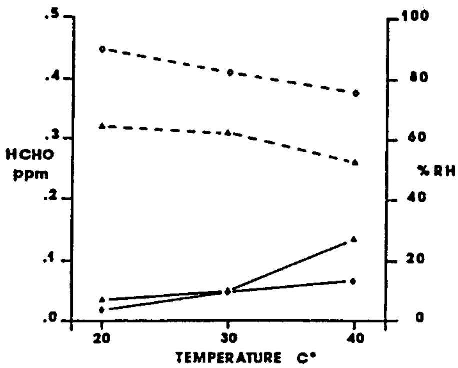

Figure 2. Formaldehyde release from gypsum board.

In this, and all subsequent figures the solid lines and symbols (▲ ♦) refer to the formaldehyde (HCHO) concentration measured. The dotted lines and open symbols (Δ ◊) refer to the percent relative humidity (% RH) measured at the time of the formaldehyde determinations. The closed and open symbols of the same type belong to the same trial, i.e. ▲Δ and ♦◊.

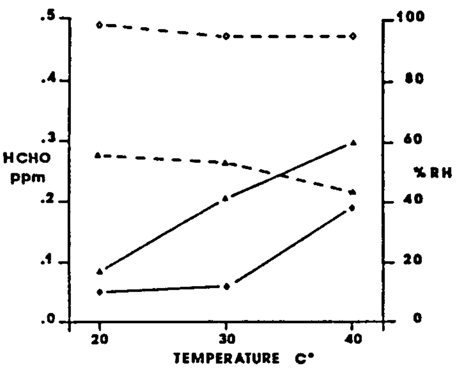

Figure 3. Formaldehyde release from from exposed gypsum board.

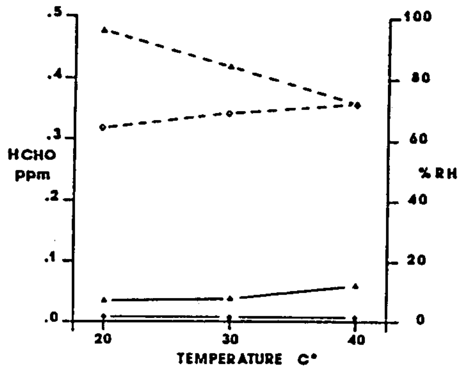

Figure 4. Formaldehyde release from UFFI exposed shiplap boards.

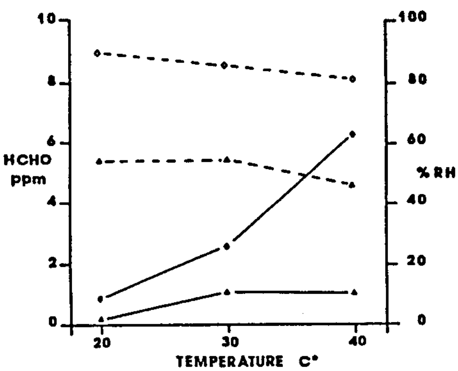

Figure 5. Formaldehyde release from new plywood panels.

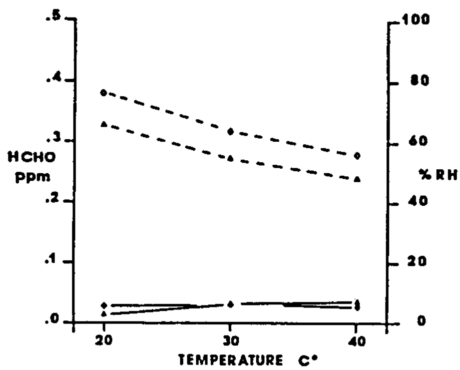

Figure 6. Formaldehyde release from UFFI exposed terra cotta building bricks.

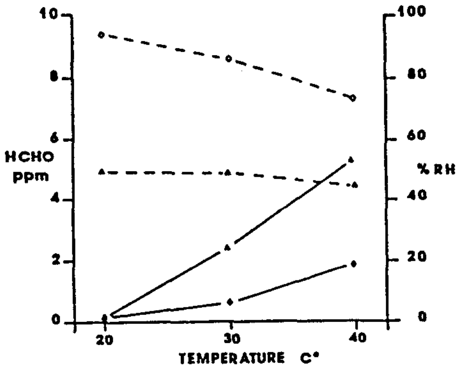

Figure 7. Formaldehyde release from UFFI exposed lath and plaster.

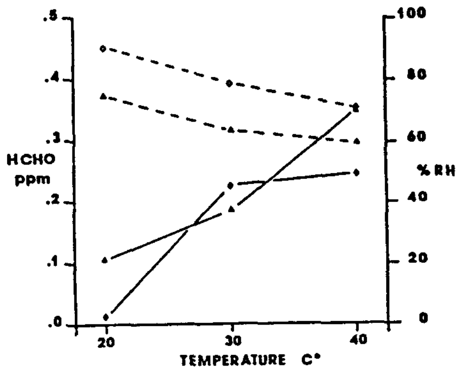

Figure 8. Formaldehyde release from cement building block previously exposed to UFFI.

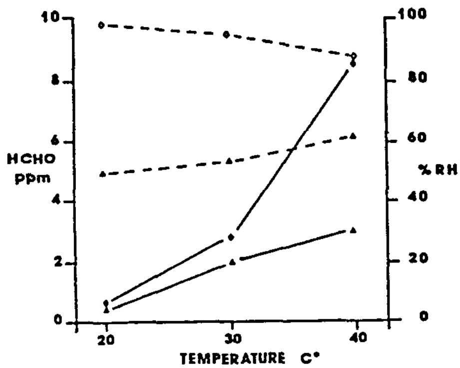

Figure 9. Formaldehyde release from new K-3 particle board panels.

Lath and Plaster (UFFI exposed). The relatively small samples (TSA 0.1005 M^2) were formidable sources of formaldehyde (Figure 7). Although the inner surface of this material was thoroughly steel brushed it is virtually impossible to remove all foam from the porous spaces within and between the lath and plaster. The practice of brushing out wall cavities therefore appears inadequate. Sandblasting might prove to be more effective. Again an increase in humidity coincides with a decrease in formaldehyde release. As wood derived products as noted in figures 1, 4, 5 show the opposite effect with high humidity it appears that the plaster rather than the lath, is the major source of formaldehyde.

Cement Building Block (UFFI exposed). The blocks were completely sandblasted in order to remove any of the foam which was present in its internal cavities. Figure 8 shows the formaldehyde release. High humidity again results in an average decrease in formaldehyde release. In this case remnant foam contamination of this material is very unlikely due to the sandblasting treatment. It appears, therefore, that the cement material of the block has absorbed considerable amounts of formaldehyde. For this reason it is questionable if even the method of sandblasting wall cavities is adequate when dealing with cement products.

K-3 Particle Board (new). Consistent with the results so far observed for wood based materials an increase in humidity increases the formaldehyde release, Figure 9. Again, as illustrated by the humidity curves, the net transfer of moisture from sample to the air and vice versa is important in controlling the extent of formaldehyde release. As indicated in this figure this material is a relatively high emission of formaldehyde.

The short terms testing regime used here measures two parameters: a). formaldehyde release under equilibrium or near equilibrium conditions. This information is obtained from the 20°C (room temperature) measurements. b). Formaldehyde release potential. This information is provided by the 30°C and 40°C release measurements. Under the conditions of these short term tests, the 30°C and 40°C release values do not, and are not expected to, describe equilibrium conditions but are an indication of formaldehyde release with increasing temperature, i.e. conditions which are often encountered in the normal living environment. A 10°C or, under certain conditions, a 20°C increase in temperature can be expected in situations where, for instance, the central heating is turned up or when solar radiation strikes a southern exposed wall, heating up the wall cavity and its contents.

The resulting composite curves simulate the two conditions encountered in the living environment i.e. equilibrium or steady state formaldehyde release at a relatively low temperature as well as formaldehyde release over various temperature gradients above the base level temperature. The curves obtained are the combined result of the various physical, chemical and possible biological action on formaldehyde sources including the formaldehyde which is physically absorbed onto the surface of the material, absorbed by the moisture held by the material and chemically bound either in a free form or in a polymerized form. The release from these sources is further influenced by, among others, concentration gradient, porosity, humidity differential geometric configuration and the bulk of the material under investigation.

Based on the response of the various materials to high and low humidity, over the time span used, there appear to be two general categories; one that increases formaldehyde release with increasing humidity i.e. the wood based products, ceiling tile, shiplap, plywood, and K-3 particle board. The other class includes gypsum board, lath and plaster, terra cotta brick and cement block. The materials belonging to the latter category decrease formaldehyde release with increasing humidity. Preliminary experiments indicate however that this trend reverses approximately 4 hours after these cement and plaster products are kept at a particular temperature and humidity under dynamic conditions of 1 liter per minute.

Judging from the data obtained from lath and plaster and cement block it appears that brushing out cavities lined with these materials is totally inadequate. Even sandblasting, although efficient in removing UFFI, is by itself not capable of eliminating formaldehyde emission. In houses where the removal of UFFI is contemplated it might be wise, based on this data, to encourage the removal of those walls which contain lath and plaster and similar products rather than the walls made from shiplap. Further investigations on lath and plaster cleaned by the method of sandblasting should provide more information.

In order to obtain an index that compares the relative emissions from each of the samples an average release can be calculated. It has been shown that release rate coefficients based on surface area is a better measure of potential release than based on weight (Pickrell et al. 1983; Myers and Nagoka 1981a,b). For this reason the coefficients based on surface area is a better measure of potential release than based on weight (Pickrell et al. 1983; Myers and Nagoka 1981a,b). For this reason the relative emission each item has been calculated based on external surface area. Table 1 shows the average emission of the items used. The values shown have also been standardized per m^2 of total surface area. No attempt has been made to standardize for relative humidity measurements, instead the two broad categories of low percentage relative humidity and high percentage relative humidity are reported.

As can be noted in Table 1 the trend of lower humidity resulting in higher release of formaldehyde from gypsum and cement products is clearly seen. The values for brick did not reflect this effect and is most likely due to the small samples which were available fo these experiments as well as the narrow range between humidities used (see Figure 6).

Table 1, Average Formaldehyde Release in ppm. per M^2 over the 20 to 40 degree C range during 1.5 Hours.*

	Low Relative Humidity	High Relative Humidity
Carpet (nylon)	L**	L
Ceiling Title (used)	.17	.31
Gypsum Board (new)	.21	.13
Gypsum Board (UFFI exp.)	.52	.13
Shiplap (UFFI exp.)	L	.22
Plywood (new)	.23	.85
Cement Block (UFFI exp.)	.71	.56
Terra Cotta Brick (UFFI exp.	.95	1.32
Lath and Plaster (UFFI exp.)	11.30	9.81
K-3 Particle Board (new)	4.74	10.37

* The average values for this time period are based on the total emission of the 20°C, 30°C and 40°C readings divided by 3.

** L Indicates too low to measure (< 0.01 ppm).

Of the samples thus investigated, UFFI exposed lath and plaster and K-3 particle board were at least 10 times more potent formaldehyde sources than the next category which include UFFI exposed, terra cotta brick, cement block and gypsum board as well as plywood, and close to 100 times more potent than the low category formaldehyde emitters which include carpet, ceiling tile, new gypsum board and UFFI exposed shiplap.

One of the advantages of these dynamic formaldehyde release measurements over the desiccator procedure is that real time release is measured. If, for instance, a material releases most of its stored formaldehyde in the first hour as measured by the dynamic system and none thereafter, this would not show up in the desiccator procedure used to measure 24 hour release coefficients and a coefficient of an average release would be reported (Pickrell et al. 1983). This could give the false impression that release continues day after day at such a rate.

Although the loading characteristics (i.e. surface area of material being tested per free volume of chamber) (Myers and Nagoka 1981a,b) were in most cases kept similar this was not possible for all materials specifically terra cotta brick and cement block. The data reported here, therefore, cannot be expected to provide an absolute value of relative emission rates but they do provide, however, a good and realistic approximation of formaldehyde release characteristics that can be expected to occur from the various building materials when present in a particular living environment.

The findings reported here also provide some insight into the reasons why there have been continuing complaints from home owners regarding elevated formaldehyde levels even after UFFI has been removed from their houses.

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