FORMALDEHYDE EMISSION FROM SOLID WOOD

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ABSTRACT

Formaldehyde parameters of five European wood species were determined using test methods applicable for wood-based materials. The five wood species studied showed differences in their formaldehyde emissions. Samples of green oak produced values of 9 parts per billion of formaldehyde (the highest value) and dry beech wood produced 2 parts per billion of formaldehyde (the lowest value). The emission values determined in the dry state were 1 to 2 units higher than those in the green state, with the exception of oak.

The demand for minimizing formaldehyde (HCHO) emissions from wood-based materials has led to the production of wood-based materials with very low formaldehyde levels. These products have formaldehyde emissions well below the limit of 0.1 parts per million (ppm) determined by the Regulation on the Prohibition of Chemicals (Germany) (2). Also, wood-based materials have been developed using formaldehyde-free binders, which results in a very low (but detectable) formaldehyde emission level. This detectable level can be expected because formaldehyde is a metabolism product that is always present in nature. Thus, a value of "zero" cannot be attained even though the sensitivity of the measurement equipment has dramatically improved.

While a fair amount of data exist for glued wood products, there is little data available on formaldehyde values for solid wood. In 1978, Mayer determined a perforator value of approximately 2.7 mg formaldehyde per 100 g of dry wood for spruce (4) according to the iodometric evaluation method. The photometric value might be lower. Roffael (5) compared the formaldehyde emission of glued and unglued particles using the perforator and the WKI bottle method (5). The analytical evaluation of both methods was carried out iodometrically. The following perforator values were determined: 2.6 mg formaldehyde per 100 g of dry, debarked pine wood and 4.8 mg formaldehyde per 100 g of dry oak wood with bark. The problematic nature of the iodometric evaluation has been pointed out several times in the literature (1).

In this study, formaldehyde parameters were determined according to gas analysis, perforator, and bottle methods (3- and 24-hr. values). Additionally, the analysis was modified in order to sensitively and accurately determine the formaldehyde values, which were expected to be relatively low. This work was not a serial systematic investigation of wood species, e.g., age, growth rate, etc., but rather an initial investigation to allow a general estimation of formaldehyde emissions from solid wood.

TEST MATERIAL

The investigation used freshly felled wood from the forests of the Braunschweig area (northern Germany) with an age range of 45 to 100 years. The following wood species were chosen: beech, Douglas-fir, oak, spruce, and pine. The different wood species were stored separately until testing under normal climatic conditions. After a defined drying period, the investigations were carried out immediately after sampling. After felling, moisture contents (MCs) ranged from 42 to 134 percent. After drying, MCs were between 7 and 9 percent (**Table 1**). Only debarked wood without resin galls or defects was investigated. The ratio of sapwood to heartwood was approximately 1:2 for spruce samples.

TEST METHODS

The following test methods for determining the formaldehyde parameters of wood-based materials were applied. In all cases the analysis was carried out using fluorimetry in order to register even the smallest quantities of formaldehyde.

TESTING IN THE 1-M^3 CHAMBER

The formaldehyde emission was determined in a specially designed chamber of 1 m^3. During testing, the temperature was 23°C ± 1°C and the relative humidity was 45 percent ± 5 percent. The air exchange rate was fixed at 1h^{-1}, the cleaning of the supplied compressed air was carried out by means of activated carbon filters (blind value of chamber: 1...2 parts per billion (ppb)). The chamber was loaded with four solid wood samples measuring 500 by 250 by 20 mm, giving a total surface area capable of emission of 1 m^2 (neglecting the edges). Thus, the

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ratio of air exchange rate to loading was 1. These test conditions are in accordance with specific provisions (2).

The formaldehyde concentration in the chamber was measured once a day until the steady state was reached. The formaldehyde analysis was carried out by sampling using a gas sampling apparatus. In this approach, air quantities of 100 L were fed through gas-washing bottles filled with bidistilled water. The sampling period was 50 minutes each. The absorbed formaldehyde was determined fluorimetrically according to the acetyl-acetone method. After transformation with acetyl-acetone and ammonia acetate to diacetyl-dihydrolutidine, the absorbed formaldehyde was determined fluorimetrically so that formaldehyde was detectable up to concentrations in the range of ppb. The tracing reaction based on the Hantz's reaction is intrinsic to formaldehyde.

The volume of the analysis air was converted to a temperature of 20°C and to an air pressure of 1013 hPa; the formaldehyde concentration was indicated in ppb (1 ppb corresponds to 0.001 ppm corresponds to 1.248 μg formaldehyde m^{-3}).

TABLE 1. — Moisture content and density of solid wood.

	Moisture content		
	Directly after cutting	After drying[a]	Density after drying[a]
	------ (%) ------		(kg/m³)
Beech	53	7	763
Douglas-fir	117	9	579
Oak	63	8	720
Spruce	42	7	416
Pine	134	8	538

[a] Dried for 14 days at 30°C with an air exchange of $1h^{-1}$.

GAS ANALYSIS METHOD

The determination of the formaldehyde emission was carried out as a quadruple analysis according to the gas analysis method DIN 52 368 and EN 717-2. The samples were 400 by 50 by 20 mm. During testing, the edges were not sealed. The samples were floated in a heated tube with a constant stream of cleaned and dried air. The temperature during testing was 60°C ± 0.5°C, the stream was 1 L/minute. In order to optimize the analysis, the formaldehyde emitted from the samples was absorbed into two gas washing bottles with 30 mL each of bidistilled water and directly evaluated using the acetyl-acetone method. The absorption period was 1 hour each with a total testing period of 4 hours. The mean value of the first and the fourth hour was calculated and converted to formaldehyde emission per surface area (ug formaldehyde per (hr. × m^2)).

PERFORATOR METHOD

The determination of the formaldehyde content was carried out according to the perforator method DIN EN 120 (August 1992) as a quadruple analysis. For this purpose, approximately 110 g of the solid wood cut into cubes (25 by 25 by 20 mm) was extracted during 2 hours in a perforator apparatus using boiling toluene in a recycling flow. The emitted formaldehyde was collected in a water recipient and then determined using the acetyl-acetone method and related to the

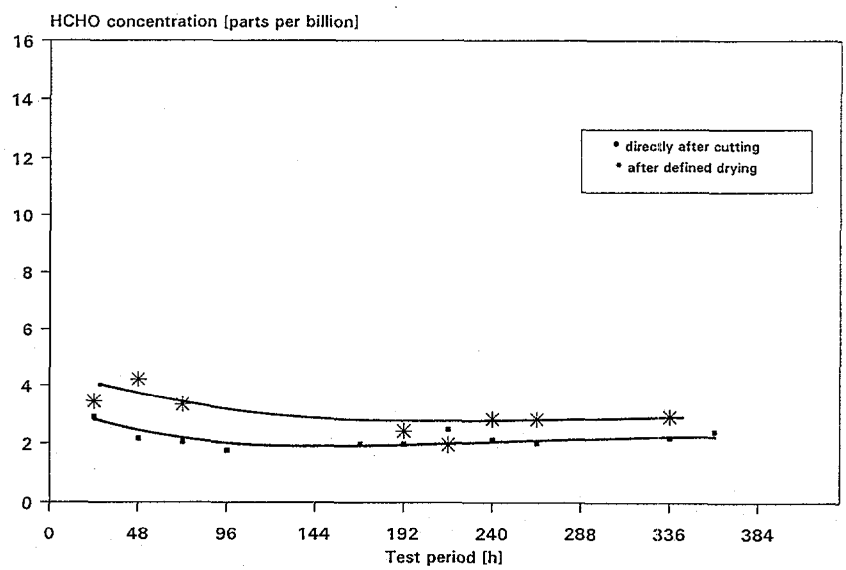

Figure 1. — Course of the formaldehyde concentration in the 1-m³ chamber of solid beech wood.

TABLE 2. — Formaldehyde parameters of the five species of solid wood tested.[a]

		Testing in the 1-m³ chamber				Flask value	
	Moisture content	Testing period	HCHO concentration	Gas analysis value	Perforator value	3 hr.	24 hr.
	(%)	(hr.)	(ppb)	(μg HCHO per (h×m²))	------ (μg HCHO per 100 g dry board) ------		
Beech	53	360	2	114	359	2	22
	7	336	3	34	155	8	12
Douglas-fir	117	384	4	397	517	4	55
	9	240	5	82	207	6	75
Oak	63	360	9	431	597	17	80
	8	360	4	51	188	6	44
Spruce	42	384	3	133	334	2	9
	7	336	4	71	277	19	132
Pine	134	240	3	195	217	2	18
	8	360	5	86	233	16	80

dry weight of the material (μg formaldehyde per 100 g dry board).

BOTTLE METHOD

For this purpose, approximately 15 to 30 g of the solid wood cut into cubes (25 by 25 by 20 mm) was stored in polyethylene bottles of 500 mL over 50 mL bidistilled water for 3 and for 24 hours in a cabinet conditioned to 40°C. The emitted formaldehyde was absorbed into the aqueous solution and analyzed using the acetyl-acetone method. The mean value from four determinations was calculated and related to the dry weight of the material (μg formaldehyde per 100 g dry board).

DETERMINATION OF MC

The wood MC was determined according to DIN 52 361 and EN 322s. For this purpose, approximately 40 g of the solid wood cut into cubes (25 by 25 by 20 mm) was stored in a cabinet conditioned to 103°C until constant weight was attained.

WOOD DRYING

Because the MCs directly after cutting were between approximately 40 and 135 percent, the samples were dried for 14 days in a 1-m^3 glass chamber at a temperature of 30°C with an air exchange rate of 1 h^{-1}. The low drying temperature was chosen because it has been demonstrated (on wood particles) that drying under industrial conditions causes the formation of formaldehyde (3). In order to avoid contaminating the solid wood samples with formaldehyde from the ambient air, dry and cleaned compressed air was let into the chamber. The wood moisture after 14 days was 7 to 9 percent (**Table 1**).

RESULTS

The results of the determination of the formaldehyde parameters are compiled in **Table 2. Figures 1** to **5** show the curve of the formaldehyde concentration in the 1-m^3 chamber in the green state directly after cutting, as well as after drying, which was carried out after the first chamber test.

It is clear that the individual wood species differ from each other in relation to their formaldehyde emissions. The steady state concentrations vary between 2 to 9 ppb after a test period of 240 to 384 hours. Oak wood in the green state showed the highest formaldehyde emission, with 9 ppb, beech wood had the lowest, with 2 ppb. The values for Douglas-fir, spruce, and pine were between 3 and 4 ppb. In the dry state, the determined formaldehyde values were 1 to 2 units higher, except for oak. Obviously, the formation of formaldehyde took place even when the wood was dried at a low temperature (30°C) (3). The value of 9 ppb determined in the green state for oak decreased to 4 ppb in the dry state. With values of about 3 to 5 ppb at a moisture of 7 to 9 percent, the differences between the wood species can be regarded as relatively slight. In general, the results obtained according to other methods of materials testing confirmed the test chamber measurements (**Table 2**). Thus, the highest values were measured on green oak wood.

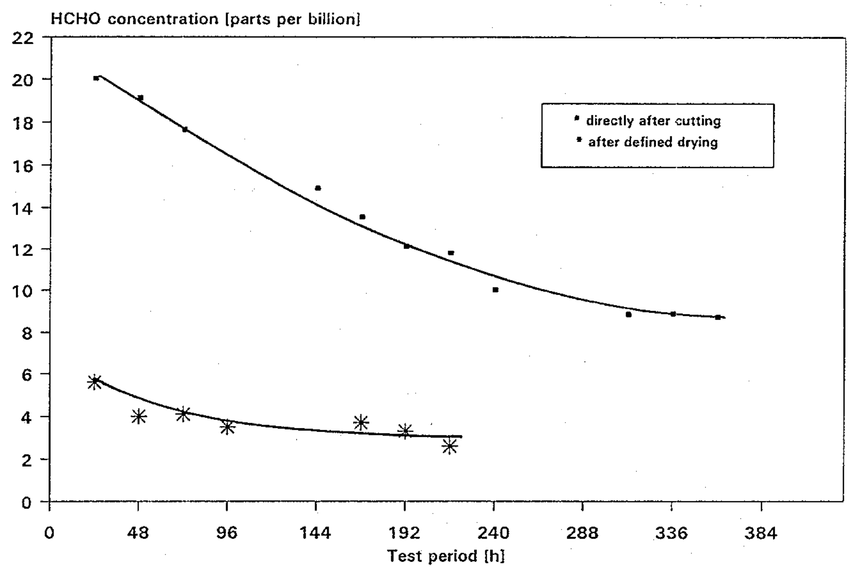

Figure 2. — Course of the formaldehyde concentration in the 1-m^3 chamber of solid oak wood.

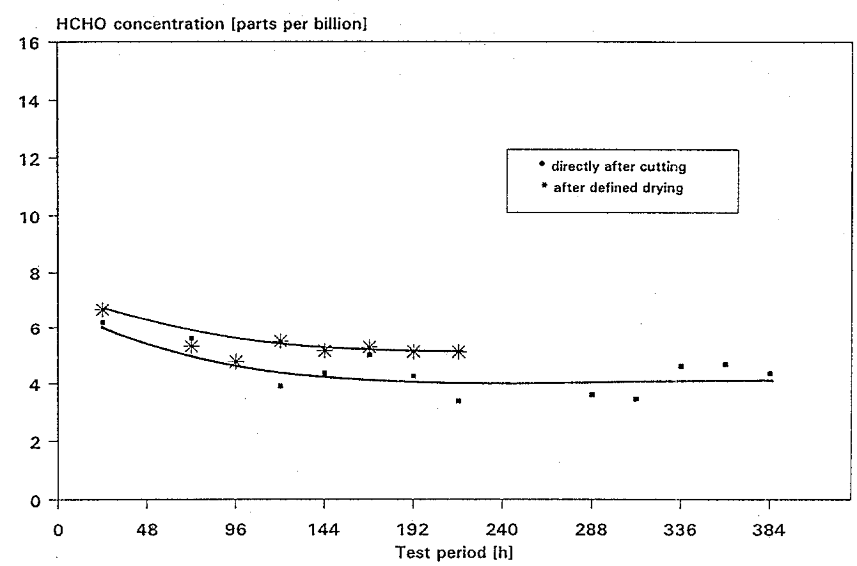

Figure 3. — Course of the formaldehyde concentration in the 1-m^3 chamber of solid Douglas-fir wood.

Calculations of a correlation (not presented here due to the relatively small testing scope) showed that solid wood behaves similarly to wood-based composite materials in relation to formaldehyde emissions. A correlation between the chamber values and values obtained from derived test methods could be established, nevertheless, partially revealing important differences between the values measured in the green and dry state. Similarities occur when comparing the parameters of the derived test methods among each other. In order to detect statistical significance, the scope of testing would have to be increased.

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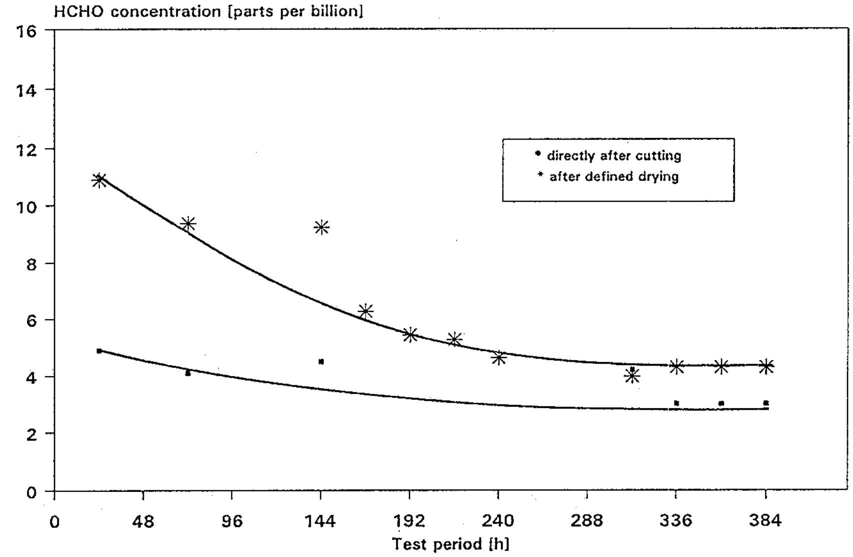

Figure 4. — Course of the formaldehyde concentration in the 1-m³ chamber of solid spruce wood.

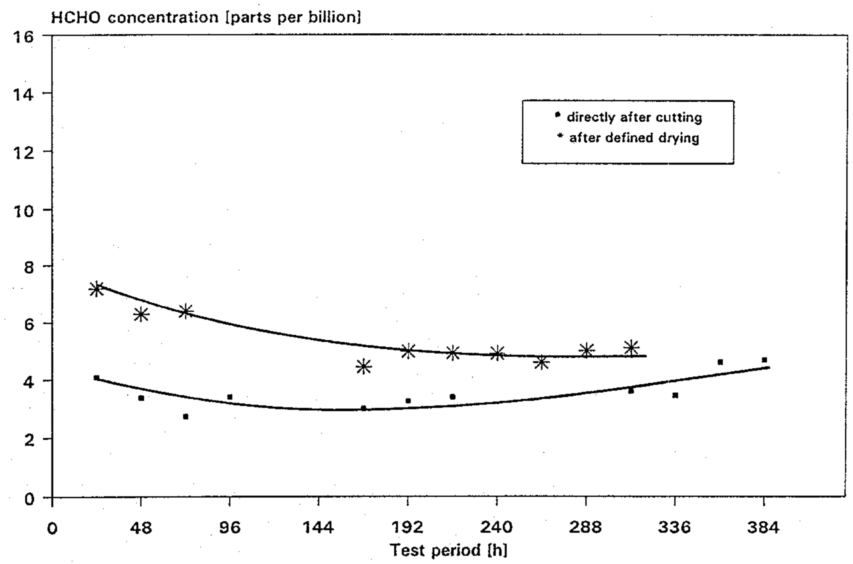

Figure 5. — Course of the formaldehyde concentration in the 1-m³ chamber of solid pine wood.