

Effects of post-manufacture board treatments on formaldehyde emission: a literature review (1960-1984)

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Abstract

This paper reviews the literature dealing with the many post-manufacture board treatments used to reduce formaldehyde emission from urea-formaldehyde bonded boards. Such treatments have almost solely used one or more of five chemical or physical principles: 1) formaldehyde reaction with NH_3 , 2) formaldehyde reaction with oxygenated sulfur compounds, 3) formaldehyde reaction with organic -NH functionality, 4) pH adjustment, and 5) physical barrier. I have categorized the available reports according to four primary board treatment methods that use the five principles in different ways. The four primary treatment methods are:

1. Application of scavengers as solids or aqueous solutions. Ammonium bicarbonate and carbonate have been used as solid powders, while the solutions involved a variety of ammonium salts, ammonium and alkali metal salts with sulfur-containing anions, and urea and other compounds having -NH functionality;

2. Exposure to scavengers as gases. NH_3 is the primary reactant here, with minor effort using SO_2 ;

3. Application of coatings. A variety of polymeric formulations have been examined as physical barriers, often supplemented by incorporating a scavenger such as urea;

4. Lamination with barrier materials. Barrier materials have included polymer films, metal films, and impregnated papers, the last also incorporating scavengers.

Firm, detailed conclusions or recommendations about the effectiveness of these methods are difficult to make due to 1) lack of information, 2) the wide variety of materials and processes employed, and 3) the importance and variety of economic and processing considerations required in selecting among options for a manufacturing process. Clearly, however, major reductions in initial formaldehyde emission can be achieved by each of the four treatment methods; most reported improvements are by at least a factor of 2 and many exceed a factor of 10. Often, the barrier properties of coatings or laminates can be supplemented by scavengers. There is a strong need for additional aging studies to verify the long-term reliability of these

treatments. At present I suggest that impregnation of boards with aqueous solutions (Method 1) is likely to be the most reliable because it should permit the use of a large scavenger excess and also allow neutralization of board acidity to reduce resin hydrolysis.

This is the fifth in a planned series of six critical reviews of the literature on different aspects of the problem of formaldehyde emission from adhesively bonded wood products. The series was initiated at the Forest Products Laboratory, Madison, Wis., in response to a need expressed by industry representatives for an independent evaluation and summation of data from diverse sources. The six aspects being reviewed concern the effects of formaldehyde-to-urea mole ratio (F/U) (48), ventilation rate and loading (49), temperature and humidity (51), separate additions to wood furnish or veneer (52), post-manufacture treatments of boards, and hydrolysis.

This paper examines the available data on effects of post-manufacture treatments of particleboard and hardwood plywood paneling on formaldehyde emission. As with the earlier reviews (48, 49, 51, 52), this analysis is based on a bibliography (50)¹ derived from several sources and, in this case, covering the period from 1960 through 1984.

¹Copies of each bibliography may be requested from the Formaldehyde Institute, 1075 Central Park Ave., Scarsdale, NY 10583, after the corresponding critique has been published. Each citation will include keywords and usually as short abstract.

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General comments

Chemical and physical principles

With few exceptions, the reported post-treatments make use of one or more of the following chemical or physical principles, details of which are available in Walker (82):

Principle A. CH_2O reaction with NH_3 . — Ammonia has been used as a formaldehyde scavenger by direct exposure of boards to ammonia gas or by treating boards with ammonium salts. It is usually stated that the ammonia-formaldehyde reaction produces stable hexamethylenetetramine (hexa), although some evidence indicates the product in acid may be methyleneimine acid salts (e.g., $\text{CH}_2\text{NH}\cdot\text{HC1}$) which convert to hexa in base. Both rate of formation and stability of hexa are greater in basic solutions. Perhaps this explains the observation by Minemura, et al. (40) that formaldehyde emission from plywood treated with a series of ammonium salt solutions decreases with increasing pH of the salt solutions.

Principle B. CH_2O reaction with oxygenated sulfur compounds. — Numerous alkali salts of oxygenated sulfur acids have been employed. Perhaps the best-known chemical principle involved here is the reaction of CH_2O with alkali sulfites or bisulfites to form the fairly stable salts of methylolsulfonic acid ($\text{HOCH}_2\text{SO}_3\text{H}$). Gaseous SO_2 exposure has also been used and presumably yields the same products in basic solution.

Principle C. CH_2O reaction with organic $-\text{NH}$ functionality. — Although some use has been made of amides and amines, by far the major application of this principle is the reaction with urea to form, at least initially, the methylolureas. Such products can, of course, readily dissociate, particularly at elevated temperatures, but they might also react with excess urea in the presence of acid to form moderately stable methylolureas. What actually occurs under the conditions of board post-treatment or thereafter is unclear.

Principle D. pH adjustment. — Hydrolytic degradation of urea-formaldehyde resins to liberate formaldehyde is minimized at near-neutral pH (16, 46). Thus, in acid-cured boards, any post-treatment that neutralizes the resin cure catalyst may have long-term benefits in addition to CH_2O scavenging activity.

Principle E. Physical barrier. — Formaldehyde emission from a board may be hindered by application of relatively impermeable coatings or laminates. In some cases this principle has been supplemented by incorporating a formaldehyde scavenger in the coating.

Board treatment methods

This review of board post-treatment could have been organized from the viewpoint of the basic principles just outlined. Instead I have chosen to emphasize the more practical viewpoints of processing and end-use application by dividing the numerous reports into five groups, based on treatment method (see Tables 1-4):

Method 1. Application of scavengers as solids or aqueous solutions;

Method 2. Exposure to scavengers as gases;

Method 3. Application of coatings;

Method 4. Lamination with barrier materials;

Method 5. Miscellaneous.

Method of presentation

The ensuing discussion examines the reported results for each of the five board treatment methods separately, making reference to the relevant basic Principles (A-E) when appropriate. The discussion is accompanied by Tables 1 to 4 which summarize the pertinent citations. The tables contain the following information for each citation, to the extent that the information is available:

1. Investigator(s) and reference number;
2. Brief description of the treatment, including board type and nature of the treating material;
3. Amount of treating material added to the board. Unfortunately, it is frequently not clear whether the stated amounts refer to one or both sides of the board;
4. Formaldehyde emission test value, before and after treatment. The test employed is also indicated, as is time after manufacture when known. Greatest reliance should be placed on emission results from some type of chamber test (large chamber, small chamber, suction test (43), actual dwelling). Next in order of reliability are the desiccator (53) or WKI (62) tests, both of which depend upon transfer of formaldehyde through the vapor phase into a water sink. Finally, the perforator test (refluxing in toluene) (11) is the least reliable here because the temperature (110°C) may degrade (66) the complex between formaldehyde and scavenger (e.g., displace the equilibrium, $\text{NH}_2\text{CONH}_2 + \text{CH}_2\text{O} \rightleftharpoons \text{HOCH}_2\text{NHCONH}_2$) or may degrade a coating; moreover, the effect of surface coating or laminate may be masked due to the large exposed edge area in the 25- by 25-mm specimens employed. This exposed edge area effect can also unfairly penalize the observed effectiveness of coatings and laminates when nonedge-sealed specimens are used in desiccator, WKI, or small chamber methods. Where formaldehyde scavengers have been allowed to permeate throughout a board, edge-sealing of small samples is not so critical; presumably this is the situation with the Method 1 and 2 treatments.

The tables present CH_2O emission data as untreated values and as percent residual after treatment ($100 \times \text{treated/unaged, untreated}$). Where aging data are available, the tables include aged, untreated values as well as percent residuals based on the unaged, untreated board; the latter reference is used because it is the overall effectiveness of a treatment that is most important.

The percent residual emission values can be useful for comparing the relative effectiveness of systems within a given laboratory but caution should be exercised in attempting quantitative comparisons between laboratories because of limited data, differences in amounts of applied material, and/or differences in board conditioning and testing. In the ensuing discussion, therefore, I frequently refer to a semi-

quantitative improvement factor based on the following scale:

<u>100 × treated/untreated</u>	<u>Improvement factor</u>
< 10%	10 ×
~ 20%	5 ×
~ 33%	3 ×
~ 50%	2 ×

5. Data adequacy. This represents my subjective judgment about the relative reliability to be accorded a given report, on a scale of low, medium, and high. Note that in a number of cases (particularly with Japanese patents) I have not gone beyond the use of abstracts; this may result in an unfair downgrading of those findings.

I have not included board physical property measurements in the tables. In the majority of cases these were not reported. Where such data are available, however, (e.g., 18, 22, 38, 68) they indicate little or no degradation in physical properties by post-treatment, as one would certainly expect.

Board treatment results

Method 1. Application of scavengers as solids or aqueous solutions

A wide variety of chemicals and procedures have been reported (Table 1). Methods of application vary from spreading solid powder to spraying, dipping, "coating," or "impregnating" with solutions. Although details often are lacking, it appears that post-application steps also differ greatly. In a number of cases, the panels are hot-stacked after application of chemicals (e.g., entries 2c, 2d, 3a, 3b, 3e-i, 18h) or are heated for short periods (entries 1b, 1c, 16c, 18d, 21). In another procedure the panels are plastic-wrapped for 1 week after scavenger application (entry 8).

Four of the five principles are represented within Method 1:

Principle A. — NH_3 as scavenger, produced from ammonium salts (entries 1-6, 8, plus possibly 9 and 10). Of these, ammonium bicarbonate and carbonate salts apparently are employed in two Swedish commercial processes, at Swedspan (entries 3b, 3e) and at ASSI (entries 2d, 3a, 3f-i).

Principle B. — Oxygenated sulfur compounds as scavengers (entries 7, 9-17).

Principle C. — Organic NH as scavenger (entries 18-22). In some cases this has been combined with Principle A (entries 1b, 2c, 2d).

Principle D. — pH adjustment with various ammonium and alkali salts. Occasionally, base has been added deliberately to ammonium salts (entries 1a, 3d, 4a).

The following points are noted regarding the reported emission improvements using Method 1:

1. Emission reduction ranges from 2 × to 10 × whether one considers all reports or only those rated medium or even high in data adequacy (last column, Table 1).

2. Putting aside the reports judged to have low data adequacy, the results with oxygenated sulfur compounds are clearly weighted toward a 10 × improvement whereas the simple ammonium salts are clearly weighted toward a 2 × improvement. Inclusion of the low-rated reports improves the ammonium salts' performance somewhat and also shows a preponderance of 10 × reduction with the organic NH compounds. Within the sulfur compounds, the ammonium and alkali sulfites may be especially promising.

These statements about the overall Method 1 data set are generally confirmed by the systematic comparisons carried out by Minemura, et al. (40) and by Imura and Minemura (18). Table 5 lists the compounds studied by these researchers in the approximate order of decreasing effectiveness. With the exception of urea, the two listings are qualitatively similar. The apparent superiority of ammonium and alkali sulfur salts is also clear.

3. Although data are very limited, the effect of increased scavenger loading is improved performance (Table 1, entries 2c, 3f, 3g). Note that the reported scavenger loadings range from 10 to 200 g/m^2 , whereas a relatively high "free" formaldehyde level in a board, as measured by a Perforator value of 50 $\text{mg}/100 \text{ g}$, requires only about 2 g/m^2 of salt or urea for complete reaction.

4. One might expect that treatment effectiveness, as measured by percent residual or improvement factor, would be less for boards possessing initially low emission. In fact, no such correlation is obvious from the data. While Kobayashi (entry 19b), for example, reports an increase in percent residual for a tenfold decrease in untreated emissions, apparently extremely high-emitting boards were used. In contrast, Swedspan (entry 3e) reports about a threefold decrease in percent residuals for boards whose initial CH_2O chamber concentrations decrease from 0.5 to 0.1 ppm. Comparisons among the reports for the same treating compound show similar contrasting results. Finally, Roffael, et al. (entry 18h) indicate little difference between urea-treated boards prepared with resins having formaldehyde-to-urea ratios of 1.6 and 1.4.

5. The limited aging data refer solely to ammonium salt-treated boards and indicate no loss in effectiveness for up to 11 months at ambient conditions (entries 1b, 3a, 3b, 3e). No accelerated aging data are available nor are data regarding retention of scavenger effectiveness when measurements are made at elevated temperature or humidity. The apparent long-term ammonium salt effectiveness contrasts with that reported for boards exposed to ammonia gas (see Method 2 discussion). Perhaps the difference is due to the customary use of a large excess of NH_3 in the form of a moderately stable salt and to the tendency of the salt to neutralize the resin's acid cure catalyst.

6. Salt treatment combined with coatings (Method 3) can result in very low emission. All entries in Table 1 for the work of Imura and Minemura (18), for example, refer to boards that were coated with polyurethane after the salt treatments, and the percent residuals are based on a desiccator value of 0.50 $\mu\text{g}/\text{mL}$ reported for a coated

TABLE 1. — Formaldehyde scavengers applied as solids or solutions (Method 1).

Investigator (Ref.)	Material	Treatment		CH ₂ O emission		Data adequacy ^e
		Process ^a	Amount ^b	Untreated ^c	Residual ^d	
1a. Ohno & Furudera (58)	Ammonium acetate	PLY. Spray 1M aqueous + 1M NaOH			(%) "odor-free"	L
1b. Otsuka (59)	Ammonium acetate	PLY. Coat 3% aqueous + 10% ethylene urea (EU), dry 30 min. 60°C	100(?)	D? 2.7 (EU coated initial)	6 (initial)	L
1e. Sasaki (69)	Ammonium acetate	PLY. Dip 5% aqueous, press 60 sec. 140°C		2.4 (EU coated 3 mo.)	4 (3 mo.)	L
2a. Imura & Minemura (18)	Ammonium bicarbonate	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C. coat polyurethane	30(?)	D. 0.50 (urethane coated)	64	H
2b. Minemura et al. (40)	Ammonium bicarbonate	PLY. "Impregnate" aqueous		D. ~50	8	H
2c. Neumann (54)	Ammonium bicarbonate	PB. F/U 1.4. Powder on one side. Stack.	96, 192	P. 18	50, 22	M-H
2d. Westling (84)	Ammonium bicarbonate	PB. Spray two sides 18% aqueous + 0.5-1.0% urea. Stack 24-48 hr. 60°C	28-35 (total)	C. ~1	40-37	H
3a. Anonymous (3)	Ammonium carbonate and/or bicarbonate?	PB. ASSI process. Spray aqueous. Stack.		P. 30	33 (initial & 5 mo.)	M
3b. Johansson (22)	Ammonium carbonate (+ other NH ₄ compounds?)	PB. Swedspan process. Spray aqueous. Stack.		C. ~1	<1-15 (initial & 5 mo.)	M
3c. Minemura et al. (40)	Ammonium carbonate	PLY. "Impregnate" aqueous		D. ~50	40	H
3d. Ohno & Furudera (58)	Ammonium carbonate	PLY. Spray 1M aqueous + 1M Ca(OH) ₂			"odor-free"	L
3e. Swedspan (79)	Ammonium carbonate	PB. Spray aqueous. Stack.		C. 0.1-0.5 (initial)	13-41 (initial)	M
3f. Westling (84)	Ammonium carbonate	PB. Powder on one side. Stack 48 hr. 40-60°C	10-20	P. 31	9-32 (8 mo.)	H
3g. Westling (84)	Ammonium carbonate	PB. Spray 20% aqueous. Stack 48 hr. 60°C	10-20	P. 31	50-34	H
3h. Westling (84)	Ammonium carbonate	PB. Spray 20% aqueous. Stack 24 hr. 60°C	20	C. 1	39	H
3i. Westling (84)	Ammonium carbonate	PB. Spray 16% aqueous + 4% ammonium acetate. Stack 48 hr. 60°C	20	P. 31	43	H
4a. Ohno & Furudera (58)	Ammonium chloride	PLY. Spray 10% aqueous + NaHCO ₃			"odor-free"	L
4b. Ohno & Furudera (58)	Ammonium chloride	PLY. Spray 1M aqueous + 1M Na ₂ SO ₃			"odor-free"	L
5. Snajder & Hosek (74)	Ammonium copper complex	PB. Spray aqueous		C. (or D?) 1-3	<10 (>1 yr.)	L
6. Minemura, et al. (40)	Ammonium phosphate	PLY. "Impregnate" aqueous		D. ~50	32	H
7. Minemura, et al. (40)	Ammonium sulfamate	PLY. "Impregnate" aqueous		D. ~50	17	H
8. Goto (14)	Ammonium sulfate	PLY. Coat one side polyurethane, coat second side 20% aqueous, wrap plastic, store 1 wk.		D. 27	2	L
9a. Imura & Minemura (18)	Ammonium sulfite	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C, coat polyurethane	~30(?)	D. 0.50 (urethane coated)	30	H
9b. Minemura, et al. (40)	Ammonium sulfite	PLY. "Impregnate" aqueous		D. ~50	3	H
10a. Imura & Minemura (18)	Ammonium thiosulfate	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C. coat polyurethane	~30(?)	D. 0.50 (urethane coated)	2	H
10b. Minemura, et al. (40)	Ammonium thiosulfate	PLY. "Impregnate" aqueous		D. ~50	3	H
11. Minemura, et al. (40)	Potassium pyrosulfite.	PLY. "Impregnate" aqueous		D. ~50	15	H
12. Minemura, et al. (40)	Potassium sulfite	PLY. "Impregnate" aqueous		D. ~50	6	H
13a. Imura & Minemura (18)	Sodium bisulfite	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C, coat polyurethane	~30(?)	D. 0.50 (urethane coated)	50	H
13b. Minemura, et al. (40)	Sodium bisulfite	PLY. "Impregnate" aqueous		D. ~50	20	H
14a. Imura & Minemura (18)	Sodium hydrosulfite.	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C, coat polyurethane	~30(?)	D. 0.50 (urethane coated)	88	H
14b. Minemura, et al. (40)	Sodium hydrosulfite.	PLY. "Impregnate" aqueous		D. ~50	21	H
15a. Imura & Minemura (18)	Sodium pyrosulfite	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C, coat polyurethane	~30(?)	D. 0.50 (urethane coated)	52	H
15b. Minemura, et al. (40)	Sodium pyrosulfite	PLY. "Impregnate" aqueous		D. ~50	9	H
16a. Imura & Minemura (18)	Sodium sulfite	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C, coat polyurethane	~30(?)	D. 0.50 (urethane coated)	<2	H
16b. Minemura, et al. (40)	Sodium sulfite	PLY. "Impregnate" aqueous		D. ~50	3	H
16c. Miwa (41)	Sodium sulfite	PLY. Coat one side 10% aqueous + 10% urea, dry 15 min. 60°C	40(?)	D. 18	2	L
17. Minemura, et al. (40)	Sodium thiosulfate	PLY. "Impregnate" aqueous		D. ~50	34	H
18a. Hojo (17)	Urea	PLY. Spray 40% aqueous	50	D. 20	5	L
18b. Imura & Minemura (18)	Urea	PLY. "Impregnate" 10% aqueous, dry 5 min. 105°C coat polyurethane	~30(?)	D. 0.50 (urethane coated)	14	H
18c. Kawahara & Takashima (24)	Urea	PLY. F/U 1.4. Spray 20% aqueous both sides	8		"reduced"	L
18d. Kitakado (28)	Urea	PLY. Coat both sides 30% aqueous, press and heat 20 min. in 60-150°C air	100-200		"odorless"	L
18e. Kitakado (29)	Urea	PLY. Coat 30% aqueous + two coats aminoalkyd resin	10	D. 10 (resin coated control)	5	L
18f. Minemura, et al. (40)	Urea	PLY. "Impregnate" aqueous		D. ~50	23	H
18g. Otsuka (60)	Urea	PLY. Coat 30% aqueous + 10% hydrazide polymer + 5% hydrazine derivative	28(?)	D. 12	<1	L
18h. Roffael, et al. (64-66)	Urea	PB. Spray both sides 10% aqueous. Hot-stack 1 wk.	40	W. F/U 1.6 = 110 F/U 1.4 = 52	72 64	H
19a. Fahlberg-List (12)	Amide	PLY. 2.5% aqueous dicyandiamide, 10% NH ₃ , 1.2% N ₂ H ₄ , 1.5% casein	20(?)		"remove odor"	L
19b. Kobayashi (30, 31)	Amide	PLY. "Treat" 30% aqueous while warm		D. 180, 16	<1, 1	L

Continued on next page.

TABLE 1. — Continued.

Investigator (Ref.)	Material	Treatment		CH ₂ O emission		Data adequacy ^e
		Process ^a	Amount ^b	Untreated ^c	Residual ^d	
20. Nishino & Shikata (55)	Aminoguanidine-HCl	B. Bd. Spray aqueous			10	L
21. Otsuka (60)	Azodicarbonamide	PB. Coat 25% aqueous + 10% ZnCl ₂ Heat 20 min. 160°C	100(?) ?	19 ppm	< 1	L
22. Noda (56)	Ethylamine	PLY. Coat 4% aqueous + 15% H ₃ BO ₃		D. (?) 19	24	L
23. Kotani & Kageyama (32)	Methylacetoacetate	PLY. Coat both sides	50(?)	D. 30	7	L

^aPB = particleboard; PLY = hardwood plywood; B. Bd. = building board.

^bmg of solid chemical per m² per side unless stated otherwise. (?) indicates reference unclear whether one or two sides.

^cC = chamber test result in ppm. D = Japanese desiccator test result (19) in µg/mL. W = WKI test result (62) in mg/100 g.

^d100 × treated/unaged, untreated.

^eL = low; M = medium; H = high. Subjective judgment of reliability of results based on amount of detail in available report. L ratings result from availability of abstract only.

TABLE 2. — Exposure to scavengers as gases (Method 2).

Investigator (Ref.)	Treatment ^a	CH ₂ O emission		Data adequacy ^b
		Untreated ^c	Residual ^d	
1. Andersen, et al. (1)	PB. SO ₂ . 50 ppm in air, ~30% RH, 22°C 80 hr.	C. 0.6	55 (60 hr.)	H
2. Eidai (10)	PB. NH ₃ . Exposed 10 sec., 20°C.	D. 4.2	5	L
3. Goto (13)	PLY. NH ₃ . "Pass through NH ₃ atm." Wrap plastic 1 wk.	D. 3	3	L
4. Jewell (21)	Mobile homes. NH ₃ . 3.8L 28% NH ₄ OH per 194 m ³ , in shallow pans 12 hr., 27°C.	R. Mean of 12 = 0.74	Mean of 12 = 26 (initial) Mean of 4 = 32 (13-20 wk.)	H
5. Johansson (22)	PB. NH ₃ . RYAB process. NH ₃ one side, partial vacuum on other.	C. ~1.3 (initial) ~0.8 (5 mo.)	8 to 25 (initial) 12 to 37 (5 mo.)	H
6. Liiri, et al. (33)	NH ₃ . RYAB process. NH ₃ one side, partial vacuum other side. PB1. PB1. Plus latex coat. PB2. PB3.	C. 1.8 (initial) 1.6 (7 mo. RH cycle) 0.25 (initial coated) 0.29 (7 mo. RH cycle, coated) 2.7 (initial) 2.3 (7 mo. RH cycle) 3.5 (initial) 2.7 (7 mo. RH cycle)	~0 (initial) ~0 (7 mo. RH cycle) 20 (initial) 44 (7 mo. RH cycle) 13 (initial) 17 (7 mo. RH cycle) 6 (initial) 15 (7 mo. RH cycle)	H
7. Maderthauer (35) Charles (9) Simon (72)	PB. NH ₃ . Verkor FD-EX process. NH ₃ exposure (1-2 min.) + formic acid exposure.	P. 174 32	3 (initial); ~4 (2 yr.) 14 (initial); ~20 (3 yr.)	M
8. Mikhailova, et al. (39)	"Wood-filled plastic panels." F/U 1.3. NH ₃ .		"Suitable for use in homes."	L
9. Myers (46)	PLY. NH ₃ . Exposure over 0.5M. NH ₄ OH, 16 hr.	C. ~3 (30 d. 35°C) ~0.3 (30 d. 75°C + 35 d. 35°C)	<1 (30 d. 35°C); 2 (30 d. 75°C + 12 d. 35°C)	H
10. Roffael, et al. (66)	PB. NH ₃ . Exposure over 25% aqueous NH ₃ .	W. ~20 to 30	14 (initial); ~40 (3 mo.)	H
11. Shvartaman, et al. (71)	PB. NH ₃ . Exposure in hot air.		"Decreased"	L
12. Verbestel (81)	PB. NH ₃ . Exposure 2.5 min. Air stream 2 min. Formic acid 2.5 min.	P. 30	23	M
13. Werner (83)	Homes. NH ₃ . Fumigation.	R. 1	30 to 40	L
14. Yamakichi (85)	PLY. NH ₃ . Vapors from (NH ₄) ₂ CO ₃ in closed chamber.		5	L
15. Yoshimitsu, et al. (86)	PLY. NH ₃ . 0.5 to 2% in air, 7 to 30 min., 80°C		"Deodorized"	L

^aPB = particleboard, PLY = hardwood plywood.

^bL = low; M = medium; H = high. Subjective judgment of reliability of results based on amount of detail in available report. L ratings result from availability of abstract only.

^cC = chamber test result in ppm; D = Japanese desiccator test result (19) in µg/mL. W = WKI test result (62) in mg/100 g.

^d100 × treated/unaged, untreated.

control. However, the uncoated control board had a desiccator value of 13.2 µg/mL; therefore percent residuals for the total treatment are actually 0.04 times those given in the table entries for those researchers.

To summarize Method 1, significant reductions in CH₂O emission (2 × to 10 ×) are achievable by post-treatments of boards with a variety of solid or aqueous formaldehyde scavengers. The variety in materials and processes, plus lack of detailed information, make it difficult to recommend a specific procedure, however,

especially when decisions must also depend on cost and processing factors. Nevertheless, the existence of two commercial processes based on Method 1 is indicative of its practicality. Among the salts studied, several ammonium and alkali salts of sulfur acids appear to provide the greatest reduction in emission. Combining this type of board treatment with coatings can be very effective. Additional information is needed however, on the effectiveness after long-term or accelerated aging and during elevated temperature-humidity exposure.

TABLE 3. — Application of coatings (Method 3).

Investigator (Ref.)	Material ^a	Amount ^b	CH ₂ O emission		Data adequacy ^c
			Untreated ^d	Residual ^e (%)	
1. Anderson (2)	PB. Waterbase. One coat. Edge and face.	2-3 mils wet	C. 0.76	20	M
2. Anonymous (4)	PB. One fill coat, two waterbase, one print, one urea-alkyd. One side, edges.		D. 0.72	100	M-H
3. Barghoorn (5)	Two sides, edges. PB. Veneered with UF. Nitrocellulose or urethane acrylic. Edges sealed.	~200	P. 52 (unveneered)	25 <10	L
4. Boatwright (6)	PB. Chemical Products CP-216 coating. Two coats on exposed side.		C ₁ 1.1 (initial) 0.8 (~30 d. T/RH cycle)	19 (initial) 14 (~30 d. T/RH cycle)	H
5. Casco (7, 8)	PB. Acrylic dispersion, containing NH ₃ . One surface.	100-150		~50 (1 coat) ~30 (2 coat)	L
6. Imura & Minemura (18)	PLY. Nitrocellulose lacquer. Surface and edges. One coat. PLY. Nitrocellulose lacquer. Surface and edges. Two coats. PLY. Nitrocellulose lacquer. Surface and edges. Two coats. PLY. Polyurethane. Surface and edges. Two coats. PB. Polyurethane. Surfaces and edges. Two coats. PB. Nitrocellulose lacquer. Surface and edges. Two coats.	100 -200	D. 40 40 ~75 ~75 ~35	12 2 ~1 <1 ~25 ~33	H
7. Jernstrom, et al. (20)	PB. Poly(vinylacetate) latex paint Poly(vinylacetate) latex paint + 3% urea Poly(vinylacetate) latex paint + 2% ethyleneurea Poly(vinylacetate) latex paint + 4% ethyleneurea	350 350 175 175	C ₁ 1.2	17 11 5 1	H
8. Kettenis (27)	PB. Solvent based vinyl copolymer paint. Latex wall paint, aqueous. Alkyd dispersion, aqueous. Vinyl dispersion, aqueous, + CH ₂ O scavenger.	580(?) 620(?) 825(?) 505(?)	C _s 0.24	7 7 9 15	M-H
9. Liiri, et al. (33)	PB. Alkyd base + latex surface. Alkyd base + alkyd surface. Latex base + alkyd surface. Latex base + latex surface. Finnish reactive paint base #1 + latex surface. Finnish reactive paint base #2 + latex surface.	138 + 163(?) 133 + 138(?) 175 + 133(?) 175 + 158(?) 213 + 158(?) 208 + 163(?)	C _s ~1 (initial, 5 mo. RH cycle)	13,7 (initial, 5 mo. RH cycle) 6,1 (initial, 5 mo. RH cycle) 15,1 (initial, 5 mo. RH cycle) 17,21 (initial, 5 mo. RH cycle) 3,1 (initial, 5 mo. RH cycle) <1 (initial, 5 mo. RH cycle)	H
10. Liiri, et al. (34)	PB. Finnish reactive paint base + latex surface on apartment wall panels.		R	<0.12	H
11. McVey (37)	PB. Prime coat Base coat fill Conventional fill or UV fill Base coat + conventional or UV fill UV fill + acrylic top coat		D.	70 80 50 25 10	L
12. Minemura, et al. (40)	PLY. Lacquer Urethane Epoxy		D. 1.3-22.3	~54 ~45 5-21	M
13. Mohl (43)	5 PB's. Standard retail paint, 1 side. Standard retail paint, 2 sides. Hide glue paint, 1 side. Hide glue paint + 4% urea, 1 side.		S.	24-52 9-52 32-54 (32 d.) 17-30 (32 d.)	M
14. Molhave (44)	PB. Falima 271 with CH ₂ O scavenger, 2 sides.	68	C _s 0.46	58	H
15. Myera (46)	PLY. Valspar 50100 with urea. All surfaces, edges.	63 total	C _s 3 (26 d., 35°C) 0.3 (30 d., 75°C + 35 d., 35°C)	10 (12 d., 35°C) 1 (30 d., 75°C + 21 d., 35°C)	H
16. Noda (57)	PLY. Polyamide derivative.	10(?)	D(?)19	10	L
17. Roffael (63)	PB. Falima F (paint with NH ₃ , casein, N ₂ H ₄)	200(?)	W. 64	5	M-H
18. Rundle (67)	PB. Wax-based hot melt.	88(?)	C _s	1	H
19. Seymour (70)	PLY. Acrylic emulsion + 5% urea. Back and edges. PB. Acrylic emulsion + 5% urea. All surfaces.	1.5 mil 1.5 mil	C _s 23 C _s 31 C _s 0.57	9 4 0 0 9	M
20. Spedding, et al. (75)	PB. Water-base primer-sealer. One coat. Alkyd paint. Two coats. Interior acrylic paint. Two coats				M
21. Sundin (78)	PB. Polyester lacquer	>250(?)	C ₁ >1.0 ppm, i.e., Class E ₂ or E ₃	0.1 ppm, i.e., Class E1	M-H
Skiest (73)	Urethane lacquer Acid-curing lacquers on base coat.	>250(?) >100 ea. (?)	C ₁ >1.0 ppm, i.e., Class E ₂ or E ₃	0.1 ppm, i.e., Class E1	

Continued on next page.

TABLE 3. — Continued.

Investigator (Ref.)	Material ^a	Amount ^b	CH ₂ O emission		Data adequacy ^c
			Untreated ^d	Residual ^e	
22. Sundin (77)	PB. Acrylate paint, both sides, sealed edges.	200(?)	C ₁ 3.2	56	H
	CH ₂ O absorbing paint, both sides, sealed edges.	100(?)	1.2	20	
23. Valspar (80)	PB & PLY. Acrylic coating with CH ₂ O scavenger.	1.5 mil		~5	

^aMaterials are described as in original reports. PB = particleboard; PLY = hardwood plywood.

^bg/m² of each surface unless stated otherwise. (?) indicates area covered was not specified.

^cL = low; M = medium; H = high. Subjective judgment of reliability of results based on amount of detail in available report. L ratings result from availability of abstract only.

^dC = chamber test value in ppm. Subscripts s and l refer to small or effectively large chamber. D = Japanese desiccator test value (19) for Japanese reports or U.S. desiccator test value (53) for U.S. reports, in µg/mL. P = perforator test value (11) in mg/100 g. S = suction test value (43) in mg/m². R = dwelling value in ppm. W = 48 hr. WKI test value (62) in mg/100 g.

^e100 × treated/unaged, untreated.

Method 2. Exposure to scavengers as gases

Only one study reports the use of SO₂ gas, following Principle B (Table 2, entry 1). After 80 hours' exposure to SO₂, a 2 × improvement factor was observed. All other reports in Method 2 make use of Principle A by exposing products to gaseous NH₃. Conditions of exposure to NH₃ vary widely: seconds (entry 2) to hours (entries 4,9); 20°C (entry 2) to 80°C (entry 14). There are also differences in NH₃ concentration. Two reports involve dwelling "fumigation" (entries 4, 12) while the remainder involve boards. Two proposed commercial processes exist: 1) the RYAB (entries 5, 6) in which boards pass through a chamber wherein NH₃ is present on one side and partial vacuum on the other and 2) the Verkor FD-EX (entry 7) in which boards pass through one chamber containing NH₃ and a second containing formic acid gas which neutralizes excess NH₃.

For the most part, reported improvements are in the 5 × to 10 × range shortly after the exposure. In some cases where aging data are available, some loss in effectiveness occurs after aging. Percent residuals increase, for example, at least twofold after ambient aging for 3 months (entry 10) or after humidity cycling for 5 months (entry 6). This decreased effectiveness may be due to the loss of excess NH₃, and subsequent liberation of formaldehyde via hydrolysis of the hexamethylenetetramine (or methyleneimine) formed by the original NH₃ exposure (66).

NH₃ sorption by boards would be expected to increase the resin pH in an acid-cured board, thereby leading to decreased resin hydrolysis and reduced formaldehyde emission (Principle D). But, the board pH apparently does not rise above neutral after NH₃ sorption (66) so that conditions for highly stable hexamethylenetetramine (or methyleneimine) may not be achieved. It should be noted, too, that the product of reaction between formaldehyde and SO₂ (methylsulfonic acid) is also reportedly unstable in acid but forms stable salts in basic solution (81).

Thus, significant reductions (2 × to 10 ×) in emission can be achieved by exposure to NH₃ gas under laboratory or manufacturing plant conditions. As with Method 1, the existence of two commercial processes indicates the practicality of Method 2. Moreover, combined with coating application (Method 3), NH₃ ex-

posure can produce extremely low CH₂O emission (entry 6). Here too, however, additional study would be very desirable to clarify the importance of reported losses in effectiveness during aging and/or to reduce those losses.

Method 3. Application of coatings

The use of coatings (Table 3) to reduce formaldehyde emission obviously involves Principle E (physical barrier), although this is often supplemented by incorporating a formaldehyde scavenger. Unfortunately, missing details complicate a definitive evaluation of Method 3; detailed identification of the coating is usually absent, and often it is not clear whether the material is applied as single or multiple coats or on one or both surfaces and on edges.

Double coats can result in marked improvement over a single coat (entries 6, 11), as can coating both surfaces instead of one (entries 2, 13, 19). As noted earlier, using small specimens with uncoated edges can lead to gross underrating of a coating material – e.g., from desiccator tests, a residual CH₂O emission of 49 percent with unsealed edges and 25 percent with sealed edges, relative to controls that had unsealed and sealed edges, respectively (4). In the light of reported uncoated surface and edge effects, I have assumed in the following that a report of strong emission reduction values (e.g., improvement factor of ~ 5 × to 10 ×) implies that such effects were absent or minimized.

McVey (37) comments that virtually any coating is likely to yield some reduction in formaldehyde emission, and the residual values in Table 3 (second last column) attest to a very wide range in effectiveness (<1% to 80% residual). Among those results where edge or single surface emissions are apparently not a problem, effectiveness predominates in the 5 × to 10 × improvement factor range. It is certainly noteworthy that the German standard specifies certain coating materials (entry 20) to be acceptable for converting Class E2 or E3 boards (>1 ppm in chamber test) to Class E1 boards (0.1 ppm in chamber test).

From data obtained within a given laboratory, a limited comparison of coating types is possible (> means more effective):

1. Polyurethane ≥ nitrocellulose (entry 6);

TABLE 4. — Laminates and miscellaneous (Methods 4 and 5).

Investigator (Ref.)	Treatment ^a	CH ₂ O emission		Data adequacy ^b
		Untreated ^c	Residual ^d (%)	
LAMINATES				
1. Anonymous (4)	MDF. 1 face 2 mil polyester, film, 1 face phenolic kraft. Unsealed edges. Sealed edges.	D. 5.5 4.7	94 15	M-H
2. Groah, et al. (15)	PLY. 2 mil vinyl film one side, edges sealed. Only unfinished backs exposed. Both surfaces exposed. Vinyl surface only exposed.	D. 3.0	70 4	H
3. Kawahara & Takashima (23)	PLY. Poly(vinylacetate) + urea as adhesive for decorative paper sheets. Both sides.	D. 30	50	GM
4. Kawashima, et al. (25)	PLY. Decorative paper impregnated with NH ₄ C1, urea, oxamide and bonded with UF.		"odor free"	L
5. Matsushita (36)	PLY. Paper impregnated with urea and bonded to PLY back with polypropylene.		"odor free"	L
6. Minemura, et al. (40)	PLY. Polyvinylchloride paper. Cellulose paper.	D. 0.9-22 D. 0.9-22	69-100 75-89	M
7. Mohl (43)	PB. Variety of boards covered with various melamine resin papers.	S.	2-42	L-M
8. Mozisek (45)	PB. Al sheet bonded with polyethylene or vinyl.		"reduced"	L
9. Rapoport, et al. (61)	PB. Decorative paper.		~85	L
10. Spedding, et al. (75)	PB. Printed cellulose wallpaper. Vinyl wallpaper.	C _s 0.57	100 0	M
11. Sundin (78); Skiest (73)	PB. Melamine-impregnated paper.	C ₁ > 1.0 ppm, i.e., Class E2 and E3	0.1 ppm, i.e., Class E1	M-H
12. Sundin (77)	PB. Polyethylene foil, both sides, edges sealed. Vinyl paper, both sides, edges sealed. Melamine-formaldehyde impregnated paper, edges sealed, both sides.	C ₁ 3.2 3.2 3.2	34 25 50	H
MISCELLANEOUS				
13. Meyer & Carlson (38)	PB. Exposure to 4 MHZ radio-frequency. 0.2 - 4 KV/cm, ~2 min.	D. ~1.3	~6	H
14. Miwa & Tomokawa (42)	PLY. Paper impregnated with 20% aqueous ammonium sulfamate, dried, used to wrap PLY for 10 days.	D. 30	12	L
15. Santar, et al. (68)	PB. γ irradiation 0.5 to 100 kGy.		80-85	L

^aDescriptive terms used as in original reports. MDF = medium-density fiberboard, PLY = hardwood plywood; PB = particleboard.

^bSubjective judgment as to adequacy of details supplied. L = low (usually only abstract available); M = medium; H = high.

^cD = U.S. desiccator test (53) for U.S. reports and Japanese desiccator test (19) for Japanese reports. C₁ refers to large chamber and C_s to small chamber. S = sucking method (43).

^d100 × treated/unaged, untreated.

2. Alkyd dispersion \approx latex wall paint \approx solvent based vinyl (entry 8);

3. Epoxy > lacquer or nitrocellulose (entry 12);

4. Alkyd paint > interior acrylic paint (entry 20);

5. Latex base/alkyd surface \approx alkyd base/alkyd surface > alkyd base/latex surface > latex base/latex surface (entry 9).

The last comparison indicates the existence of interaction between coating combinations.

Kazekevicz and his colleagues have measured the formaldehyde permeabilities of various coating materials in the free film form and estimated the potential of such materials as barriers to formaldehyde emission (26, 76). For a 100- μ m-thick polyurethane film (\approx 100 g/m² coverage) their data predict greater than 100-fold reduction in formaldehyde emission rate, i.e., more effective barrier properties than seems to be observed on wood panels (Table 3). On a relative basis, their data further predict that barrier properties should be in the order: alkyd paint > polyurethane >> vinyl-based paint.

One might expect that some coating materials (e.g., polyurethane) would possess formaldehyde-scavenging capability in addition to their barrier properties. The

available data do not provide strong evidence that such is the case. However, several investigators have taken the logical step of including a formaldehyde scavenger in a coating (entries 5, 7-10, 13-15, 17, 19, 22, 23). With a few exceptions these combination systems yield 5 × to 10 × reductions in CH₂O emission compared to un-

TABLE 5. — Effect of impregnating compound (Method 1) on plywood emission.

Salt	Residual emission ^a	
	Minemura, et al. (40)	Imura & Minemura (18)
Sodium sulfite	3	<2
Ammonium thiosulfate	3	2
Ammonium sulfite	3	30
Potassium sulfite	6	
Sodium pyrosulfite	9	52
Ammonium bicarbonate	8	64
Potassium pyrosulfite	15	
Sodium bisulfite	20	50
Sodium hydrosulfite	21	68
Urea	23	14
Ammonium phosphate	32	
Ammonium carbonate	40	

^aDesiccator test.

TABLE 6. — Treatment summary.

(1) Treatment method	(2) Emission improvement ^a		(4) Product range ^b	(5) Long-term reliability ^b	(6) Processing changes required ^b	(7) Estimated relative cost ^b
	Initial	Aged				
1. Solids or aqueous solutions	2× -10× (~1/3 >10×)	Little data. Expect some loss in effectiveness for some treatments. Data needed.	1	1 Can use excess and adjust pH.	1-2 Spray, brush, dip, dry, some hot-stacking.	1-2 Vary with material, heating, etc.
2. Gas exposure	2× -10× (~1/3 >10×)	Evidence for some loss in effectiveness. Data needed.	1	3 Aging a problem?	3 Exposure chambers, gas handling.	2-3
3. Coating	2× -10× (~1/3 > 10×) aided by scavenger	Data needed.	2	2 Film integrity critical.	2 Spray, brush, dry.	2
4. Laminate	2× -10× (few > 10×)	Data needed.	4	2 Film integrity critical.	3-4 Bonding step.	3-4

^aSee text, General Comments, for definition.

^bSubjective judgments. Lowest numbers preferred.

coated boards. The actual contribution of the scavenger in the coating can be estimated in only three cases, the improvement being approximately fivefold in one case (entry 9) and twofold in another (entry 13). In the third case (entry 7), Jernstrom, et al. report that urea is moderately effective in reducing CH₂O emission (-30% reduction at 3% addition) but tends to produce surface defects in the paint film; in contrast, they find ethyleneurea to be a much more effective scavenger (~threefold reduction at 2% addition) that does not degrade the coating film (20).

Thus, where coatings can be used, they offer the potential for excellent protection against formaldehyde emission, although edge coating can be critical with small panels. Commercial application of the Method 3 principle is extensive (though perhaps sometimes serendipitous) in the form of decorative coatings on plywood, particleboard, and fiberboard panels. The addition of scavengers to the board or coating can be highly beneficial. Clear recommendations as to coating type cannot be made on the basis of available information. Aging studies may be especially desirable here because a coated board retains much of its emission potential, and loss of film integrity will allow the retained formaldehyde to escape.

Methods 4 and 5.

Lamination with barrier materials and miscellaneous

Data for laminates (Table 4) are much less extensive than for coatings. Where edge and backside effects clearly do not influence test values (entries 1, 2, 11, 12), effectiveness is in the 2 × to 10 × improvement factor range (col. 4). A variety of laminating materials have been used – polymer films (entries 1, 2, 10, 12), metal (entry 8), and impregnated papers (entries 3, 4, 5, 7, 10, 11). The impregnated papers appear to gain their effectiveness by virtue of formaldehyde scavengers (Principle C) such as urea (entries 3, 5) or possibly melamine (entries 7, 11, 12) and/or by virtue of the barrier effect (Principle E) arising from cured melamine resins (entries 7, 11, 12).

Kazekevicz and colleagues have also reported formaldehyde permeabilities of several polymer films

(26, 76). The data indicate that formaldehyde barrier properties should be in the order: poly(ethyleneterephthalate) > poly(propylene) ≈ poly(styrene) > poly(ethylene vinyl acetate) > poly(ethylene) > poly(vinyl chloride) > Nylon 6 > cellophane, with the best being three orders of magnitude better than the worst.

Thus, laminates also can be an effective means of reducing formaldehyde emission from boards. Obviously, however, their use is limited to rather special applications, because of cost and/or product properties.

The miscellaneous category of post-treatment methods includes three reports (Table 4). One of these involves wrapping plywood with ammonium sulfamate-impregnated paper (entry 14). Since this procedure probably owes its effectiveness to ammonia vaporization and sorption, it might well have been included in Table 2 as an example of a less direct (and more expensive?) exposure to a gaseous scavenger. Entries 13 and 15 of Table 4 both involve board irradiation; the former indicates effectiveness in the 10× range, apparently as a result of resin postcure.

Conclusions

My conclusions about each of the four primary post-treatment methods are summarized at the end of each Method discussion. Table 6 attempts to summarize in general terms the advantages and disadvantages of these four treatment methods. The relative ratings in columns 4 to 7 are my own highly subjective judgments and are intended only as indicators for commercial post-treatment process selection. My overall conclusions are:

1. Major reductions in initial formaldehyde emission are achievable by each of the four general post-treatment methods. Most reported improvements are at least by twofold and many exceed tenfold. In many applications the barrier properties of coatings (Method 3) or laminates (Method 4) can be enhanced by using scavengers (Methods 1 and 2). It appears, moreover, that commercial applications of all four general methods exist.

2. There is a strong need for additional aging studies to verify the long-term reliability of these post-

treatments. My expectation is that impregnation with aqueous scavengers offers the potential for greatest long-term reliability (Table 6, col. 4) because one can, in principle, use a large excess of scavenger and also control the board pH in order to minimize formaldehyde emission arising from hydrolysis processes.

3. The choice of method will depend upon 1) the extent and long-term reliability of emission reduction needed, 2) the particular end use for the product, and 3) economic and processing considerations. My subjective ratings in columns 4 to 7, Table 6, indicate that a Method 1 (solid or solution) treatment should be used most often and a Method 4 (laminated) least often. Where a Method 3 (coating) or a Method 4 (laminated) treatment is preferred – or necessary – a supplemental Method 1 (solid or solution) treatment could be useful to ensure low initial and long-term CH₂O emission. For Method 3 (coating) and for some laminates in Method 4, an alternative supplement is incorporation of a scavenger in the coating or laminate.

4. Firm recommendations on specific variations within each of the four methods are difficult to make because of lack of detailed information and economic and processing considerations. In Method 1 (solid or solution) the data indicate greatest emission reductions with ammonium and alkali metal salts of sulfur anions (e.g., sulfites and thiosulfate); the benefit of achieving a neutral board pH after treatment should also be considered. In Method 2 (gas exposure), NH₃ is the only gas given serious study to date. In Method 3 (coating) and Method 4 (laminated) a variety of materials seem satisfactory, but some differences in CH₂O emission reduction effectiveness definitely exist.

5. Clearly, many of these post-manufacture treatments could, in principle, be used to supplement other formaldehyde emission reduction steps that are currently being taken – e.g., use of low mole ratio resins (48) and/or separate additions of scavenger systems to furnish (52).

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