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THE ABATEMENT OF INDOOR POLLUTANTS BY THE USE OF REACTIVE POLYMERIC COATINGS ON FILTERS

By Dr. H.D. Gesser

The s.p.d.f. Chemical Associates Ltd.
218 Girton Boulevard
Winnipeg, Manitoba R3P 0A7

CMHC Project Officer: Chris Ives

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ABSTRACT

Homes heated by forced air have a fan in the furnace which circulates the heated air throughout the house. A filter is usually present between the fan and the heat-exchanger. This filter can be coated with a plastic which reacts with pollutants in the air of the house and removes them as the air is circulated by the furnace fan.

We have shown that formaldehyde can be removed from the air when the fiber-glass filters are coated with the polymer polyethylenimine mixed with glycerol to keep it from hardening. By increasing the amount of polymer (plastic) applied to the filter it has been possible to extend the life of the coating to almost four months during which time the level of formaldehyde in the house is well within acceptable levels.

The same coating has previously been shown to be effective in removing acidic gases such as NO_2, SO_2. H_2S, and HX from air streams where they were present at relatively high concentrations. We now showed that for NO_2, at about 1 part per million in air, the coating is still effective but at lower concentrations as found in a home where the gas is produced by open fire such as a kitchen range or space heater, the coated filter was marginally effective.

It was also shown that in the process of air-conditioning a house or dehumidifying the air the formation of water of condensation will remove part of the water soluble pollutants, such as formaldehyde, in the air.

EXECUTIVE SUMMARY

We had previously shown that it was possible to remove formaldehyde from indoor air by treating the fiber-glass furnace filter with polyethylenimine (PEI) which reacts with formaldehyde thereby removing it from the air. No other product besides water is formed in the reaction. Initial lifetime of the coating was about 2 weeks. However when the coatings (7.5 g PEI with a density of PEI on the filter of about 23 g/in^2) were plasticized with glycerol, the lifetime was extended to about a month.

The object of this project was to extend the life of the coated filter by increasing the amount (density) of the PEI on the filter and to verify the usefullness of the reactive coating concept.

A test house was selected in which formaldehyde was introduced by heating paraformaldehyde at a constant temperature. The level of formaldehyde in the house was below the TLV of 100 ppb, namely between 50 and 80 ppb. When PEI (with glycerol) was coated onto two fiber-glass furnace filter to the extent of 30 g PEI or 50 mg/in^2 then the concentration of formaldehyde in the house was reduced to much lower levels of from 0 to about 20 ppb. After about a month, the coated filters were replaced by untreated filters and the concentration of formaldehyde increased. When the treated filters were reinserted into the furnace the concentration of formaldehyde decreased and remained low for another 3 months rising to the high levels of about 80 ppb at the end of the total exposure time of 4 months indicating the exhaustion of the coating and that the PEI had reacted completely with formaldehyde.

After 4 months of use the filters were coated with a thick layer of dust and it would be impractical to use more PEI on the filter to extend the useful life beyond the 3 1/2 months.

Another object of the project was to test the PEI coating for its removal of NO_2 at low concentrations. We had previously shown that PEI/Gly coatings were efficient in removing NO_2 from an air stream containing 250 ppm NO_2. Using an NO_2 specific ion electrode it was possible to show that the coating was effective in reducing the concentration of NO_2 in air from 1 ppm to about 0.4 ppm in a single pass.

Collaborative work with the Gas Research Institute (Chicago) using a very sensitive chemiluminescent method for the analysis of NO_2 showed that at 0.1 to 0.2 ppm of NO_2 the PEI coated filter in a house furnace was shown to be only marginally effective in the removal of NO_2. Further experiments are planned to determine if a greater effect could be realized when the coated filter is located closer to the source of the NO_2, i.e. the open flame of the kitchen gas range.

RÉSUMÉ

Nous avons déjà montré qu'il était possible de retirer le formaldéhyde de l'air intérieur en traitant le filtre en fibre de verre du générateur de chaleur au polyéthylénimine (PÉI) qui, en réagissant avec le formaldéhyde, l'élimine de l'air. Aucune autre substance que de l'eau ne se forme au cours de la réaction. Au début, le revêtement n'avait qu'une durée d'environ 2 semaines. Mais lorsque les revêtements (7,5 g de PÉI, moyennant une densité de PÉI sur le filtre d'environ 23 g/po^2) ont été plastifiés à l'aide de glycérol, la durée a été prolongée à environ un mois.

L'objet de la présente recherche consistait à prolonger la durée du filtre revêtu en augmentant (densifiant) la quantité de PÉI sur le filtre et à vérifier l'utilité du principe du revêtement réactif.

On a choisi une maison d'essai dans laquelle du formaldéhyde a été introduit en chauffant du paraformaldéhyde à une température constante. Le niveau de formaldéhyde de la maison était en deçà de la valeur limite d'exposition (TLV) de 100 ppb, soit entre 50 et 80 ppb. Lorsque du PÉI (avec glycérol) a été enduit sur le filtre en fibre de verre du générateur de chaleur à raison de 30 g de PÉI ou de 50 mg/po^2, alors la concentration de formaldéhyde de la maison a été réduite à des niveaux beaucoup plus faibles variant entre 0 et 20 ppb. Après environ un mois, les filtres revêtus ont été remplacés par des filtres non traités et la concentration de formaldéhyde s'est mise à augmenter. Lorsque les filtres revêtus ont été remis en place, la concentration de formaldéhyde s'est atténuée et est demeurée faible pendant encore trois mois, atteignant le niveau élevé d'environ 80 ppb à la fin de la période d'exposition totale de 4 mois, indiquant l'épuisement du revêtement et la réaction complète du PÉI avec le formaldéhyde.

Après 4 mois d'utilisation, les filtres étaient couverts d'une épaisse couche de poussière de sorte qu'il était impossible d'enduire davantage le filtre de PÉI pour prolonger sa durée utile au-delà des 3 mois et demi.

La recherche visait également à tester l'efficacité du revêtement de PÉI à retirer le NO_2 à de faibles concentrations. Nous avons déjà montré que les revêtements de PÉI et de glycérol étaient efficaces pour retirer le NO_2 de l'air enregistrant une teneur de 250 ppb de NO_2. En utilisant un électrode à ions propre au NO_2, il a été possible de montrer que le revêtement réussissait efficacement à réduire la concentration de NO_2 de l'air, en faisant passer sa teneur de 1 ppm à environ 0,4 ppm d'un seul coup.

Les travaux entrepris en collaboration avec le Gas Research Institute (Chicago) à l'aide d'une méthode par chimiluminescence très sensible pour l'analyse de NO_2 ont révélé qu'à une concentration de 0,1 à 0,2 ppm de NO_2, le filtre revêtu de PÉI du générateur de la maison parvenait peu à soustraire le NO_2 de l'air. D'autres expériences sont prévues dans le but de déterminer la possibilité d'obtenir de meilleurs résultats en situant le filtre revêtu plus près de la source de NO_2, par exemple, près de la flamme nue d'une cuisinière à gaz.

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THE ABATEMENT OF INDOOR POLLUTANTS BY THE USE OF REACTIVE POLYMERIC COATINGS ON FILTERS

A Final Report

by Dr. H. D. Gesser

The object of the project was to test the polyethylenimine coatings on filters for the removal of NO_x at lower and more realistic levels than previously studied (1) and to establish the durability (lifetime) of the coatings for formaldehyde while confirming its efficiency.

Part A: The Removal of NO_2 Gas by Coated Filters.

A nitrate specific ion electrode was ordered but it was not received until the beginning of August. Hence it was necessary to develop and test an alternate method of analysis. The one chosen was a colorimetric method of analysis (2) for nitrite (NO_2^-) and nitrate (NO_3^-) because the reaction of NO_2 with water or dilute alkaline solution is as follows:

$$2\ NO_2 + H_2O \cdots\cdots\blacktriangleright HNO_3 + HNO_2 \quad \text{or} \qquad (1)$$

$$2\ NO_2 + 2\ OH^- \cdots\cdots\blacktriangleright NO_3^- + NO_2^- + H_2O \qquad (2)$$

Calibration curves were prepared and used to evaluate a suitable method of generating NO_2. It was first attempted to produce NO_2 by heating lead nitrate, $Pb(NO_3)_2$, by the reaction

$$Pb(NO_3)_2 \cdots\cdots\blacktriangleright PbO_2 + 2\ NO_2 \qquad (3)$$

This required passing air over the heated salt at a fixed rate and keeping the salt at a constant temperature. It was noted however that fluctuations occurred in the level of NO_2 produced and that these could not be readily eliminated.

the accepted TLV is 100 ppb. Since low and inconsistent results were obtained it was decided to generate formaldehyde in my own house where better control could be maintained.

The formaldehyde was generated by heating paraformaldehyde in a weighable vial (at a constant temperature) placed near the air intake located in a basement room. The loss of paraformaldehyde could be determined daily and compared with the concentration in the house. After several trials it was possible to adjust the rate of loss of paraformaldehyde to be about 0.06 g/hour. This rate of weight loss was found to depend on the mass of paraformaldehyde in the vial, which was kept at a constant level.

The concentration of formaldehyde in the house was found to fluctuate and was very low when the input of formaldehyde was taken into account. Based on the volume of the house and assuming 5 air changes/hour it was estimated that the level of HCHO in the house should stabilize at about 70 ppb. One explanation for the lower values is that the exchange rate in the house is higher than estimated, i.e. the air changes per hour is more than the 5 ACH assumed. This was not reasonable because the house is air-conditioned and we keep the house reasonably air-tight. It was concluded that another loss was occurring and this was verified when the water condensed by the air-conditioner was tested and found to contain considerable quantities of formaldehyde. The results are summarized in Appendix A as a draft of a note which will be submitted for publication

It is obvious that consistent results are only possible when the air-conditioner is not in service. These results are given in Table 3.

We had previously shown (5) that a fiber-glass furnace filter (16" x 20") coated with PEI/Glycerol (7.5 g) at a PEI density of about 23 mg/in^2 was effective in reducing the formaldehyde level for about one month in a UFFI house. To extend the lifetime of the filter it was necessary to increase the mass of the PEI coating. This was accomplished by spraying the fiber-glass furnace filters in the house (20" x 25" and 25" x 16") with a total of

46 g of PEI/Gly 2:1 or a density of 50 mg/in^2. The lifetime of these treated filters in removing formaldehyde and the higher capacity is shown in Table 3. The results are plotted in Figure 1 and clearly show that the heavy coating of PEI can extend the life of the coated filter from the initial period of one month to about 4 months. The excessive build-up of dust on the filter makes it impractical to keep the filter for longer times and in fact 3 months of use is believed to be an appropriate limit unless the dust is first removed by an electrostatic filter.

A color indicator is in the process of development. This would change colors when the filter became exhausted or excessive formaldehyde or acidic gases or vapors were present.

For basic pollutants such as ammonia, amines, or pyridine, it is possible to use an acidic polymer such as polyacrylic acid plasticized with glycerol. This is presently being tested in a factory where ammonia is released during production.

There is also some indication (4) that the PEI may reactively destroy ozone and it is planned to investigate this shortly.

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TABLE 1. The Concentration of NO_2 in the Experimental Flair House

Date	$[NO_2]$ ppm
June 13	1.22
14 am	0.82
14 pm	0.35
15	0.40
17	8.15
18	0.16
Coated filter installed in furnace.	
June 19 am	0.082
19 pm	0.59
20 am	0.48
20 pm	1.96
21 am	2.20
21 pm	1.63
22 am	0.57
22 pm	2.45
24	4.08
25 am	2.04
25 pm	2.45
26	2.85
Coated filter replaced by untreated filter	
June 27 am	4.65
27 pm	1.22
30	4.08
July 2	2.45

Table 3 The Change in Formaldehyde Levels in Test House due to PEI, Coated Filter A in, B out

Date		[H_2CO] ppb	Date		[H_2CO] ppb	Date		[H_2CO] ppb
	B							
Oct	1	64		A		Dec	16	10
	2	52	Nov	2	20		17	9
	3	67		3	6		19	18
	4	58		4	0		22	21
	5	62		5	3		23	16
	A			6	12		24	0
	6	32		8	23		26	7
	7	2		10	16	Jan	8	27
	8	3		12	35		9	25
	9	12		14	23		10	3
	10	58		16	17		12	7
	12	17		18	1		14	13
	14	0		20	4		17	21
	17	4		22	20		18	19
	18	0		24	0		19	32
	19	0		26	14		20	29
	20	2		28	3		21	45
	21	15		30	7		22	84
	22	13	Dec	1	2		23	76
	23	0		2	0		25	108
	B			3	2		27	60
	24	56		4	24		28	61
	25	71		6	17		B	
	28	89		7	7	Feb	1	53
	29	70		8	16		2	35
	29	65		9	0		3	62
	30	81		10	6		4	71
	31	73		12	7		5	86
Nov	1	50		14	0			

TABLE 2: The Concentration of Formaldehyde in New Display Houses

	Date		[HCHO] ppb
House #1	June	15	5.5
			2.9
		20	5.5
			9.8
House #2	July	12	30
			14
		19	82
			28
		27	6
			10

Appendix A

A SIMPLE PROCESS FOR THE REMOVAL OF INDOOR WATER SOLUBLE POLLUTANTS

H. D. Gesser*, P. Wong and I. E. Gesser

Department of Chemistry, University of Manitoba, Winnipeg, MB. Canada, R3POA7

ABSTRACT: The removal of indoor water vapour by an air conditioner has been shown to remove a large fraction of the formaldehyde entering the house.

Many indoor pollutants, such as NOx, formaldehyde and hydrogen halides are water soluble and could be removed by solubilizing them in water. This has been verified during the course of some experiments designed to test the efficiency and durability of filters coated with reactive polymers to remove formaldehyde from indoor air (1,2).

A bungalow (162 m^2 plus a basement of equal area) with a forced air heating system was selected for testing. Formaldehyde was introduced into the house by heating para-formaldehyde (in a weighing bottle) at a constant temperature. The weight loss was approximately 0.06 g/h which based on an estimated 5 ACH should give approximately 0.07 ppm formaldehyde in the house. Values obtained ranged from 0.02 ppm to 0.06 ppm and seemed to depend on the time of testing. The TLV for formaldehyde in homes is generally accepted at 0.100 ppm in indoor air.

The house had central air conditioning and a test of the water condensed by the cooling coils showed it to contain from 0.4 to 1.2 ppm formaldehyde. The volume of water collected ranged from an average of 50 to 300 mL/h and depended on the use of the dishwasher (which vented indoors) and the frequency of showering that occurred as well as the outside temperature.

The results clearly show that the circulation of indoor air over the cool water coated coils of the air conditioner can remove water soluble pollutants. The possibility of

similtaneous humidifying and dehumidifying the indoor air makes it possible to remove water soluble pollutants on a continuous basis.

Further testing uunder controlled conditions is in progress.

Financial assistance from Canada Mortgage and Housing Corporation is gratefully acknowledged.

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