*Sensors and Materials*, Vol. 13, No. 2 (2001) 077–085
MYU Tokyo

S & M 0431

# Removal of Indoor Formaldehyde by Hybrid Chemical Filters

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(Received October 15, 2000; accepted April 5, 2001)

***Key words***: sick house, formaldehyde, chemical filter, air cleaning, plastic

From the viewpoint of energy conservation, the number of buildings or houses which have high adiabatic characteristics or high airtight characteristics has been increasing. As a result, what we call "sick house" syndrome is also on the rise. One of the main reasons for the syndrome is formaldehyde gas evaporating from the adhesive used in block board. In this study, a high-performance chemical filter was fabricated by coating the surface of expanded plastic using polyelectrolyte thin film. The decrease in formaldehyde concentration in a closed cell was also examined for various types of chemical filters using polyelectrolytes.

## 1. Introduction

After what we call the "oil shock" of the 1970s, from the viewpoint of energy conservation, the number of buildings or houses which have high adiabatic characteristics or high airtight characteristics has been increasing. As a result, what we call "sick building" or "sick house" syndrome is also on the rise. One of the main reasons for the syndrome is formaldehyde gas evaporating from the adhesive used in block board or housing materials. People today also tend to spend 90% of the day indoors. Therefore, it is very important to develop building materials which are free from gases toxic to humans. However, it is not always easy for builders to avoid using any kind of plywood or adhesive. In fact, the use of plywood, which is veneered with thinly sliced pieces of wood, particle board, which is formed by adhering wood chips together using adhesives, and fiberboard, which is formed by adhering wood fiber together with adhesive, is unavoidable in everyday life. All these materials emit formaldehyde gas from the adhesives used in their fabrication. As a result, high-performance air cleaners or new functional materials which can adsorb and remove the formaldehyde gas emitted from the furniture or wallpaper in the room are needed.

Recently, we found that layer-by-layer self-assembly films made of polyelectrolytes have the ability to adsorb odiferous substances and smoke.[1–3] The mechanism of adsorption is not only physical adsorption but also chemical adsorption. In this study, high-performance chemical filters were fabricated by coating the surface of expanded plastic with a polyelectrolyte thin film to create a comfortable living environment which is not harmful to human health.

Decreasing the concentration of formaldehyde in a closed chamber was also evaluated for various types of chemical filters using polyelectrolytes.

## 2. Materials and Methods

In this study, the material for the filter was prepared by coating the surface of expanded plastic with a polyelectrolyte thin film. The prepared materials were inserted into a cartridge with a fan and their ability to remove gases as an air cleaner was evaluated.

### 2.1 *Substrate coating*

Polyarylamine hydrochloride (PAH) was used as the polyelectrolyte to adsorb formaldehyde. Pure water, solutions of the salt NaCl, and city water were used to dissolve the polyelectrolyte. The polyelectrolyte solutions were prepared by dissolving the polyelectrolyte powder completely by stirring for one hour. The concentration was $10^{-1}$ M.

### 2.2 *Polyelectrolyte substrate*

Filters made from glass or polypropylene are used widely in household air cleaners; however, a waste disposal problem arises after their use because they neither burn nor decompose. In this study, biodegradable expanded plastic (ECO EXCEL), which is made from cellulose acetate, was used as the substrate for the deposition of the polyelectrolyte thin film. This is a low-cost plastic which is environmentally friendly because it is easily decomposed when buried. First, the ECO EXCEL blocks were crushed into pieces to increase the surface area. Then, the polyelectrolyte was coated on the surface and inserted into a cartridge for use as an air cleaner filter.

## 3. Experimental Setup

Evaluation of the chemical filter was carried out in an airtight chamber, as shown in Fig. 1. The air cleaner consists of a filter and a fan placed in the center of the chamber. The size of the chamber is $33 \times 55 \times 32$ cm$^3$. Volatilized formaldehyde gas from cotton dipped in a formaldehyde solution was introduced into the chamber and the concentration was ad-

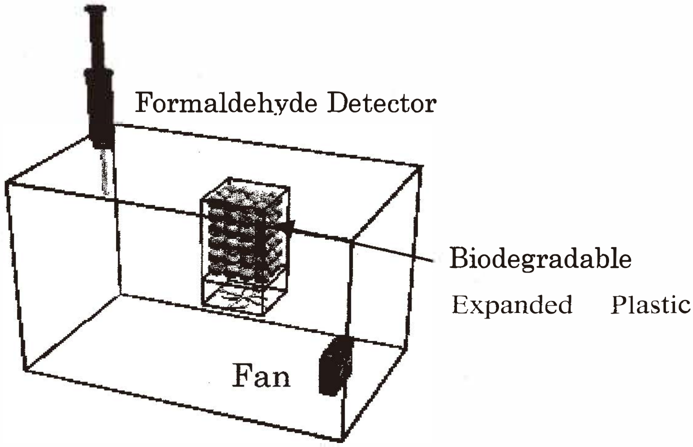


Fig. 1. Airtight chamber for the evaluation of the chemical filter.

justed to a constant value. The cotton was removed when a concentration of 10 ppm or 2 ppm was achieved. The fan located in the bottom of the chamber started to work after the formaldehyde concentration in the chamber was adjusted. The concentration of formaldehyde in the chamber was measured by gas detectors (Gas Tech, 91L, 91LL, 91PL) and a gas sensor (constant voltage decomposition method, Formtector XP308).

## 4. Measurement of Pressure Loss

The pressure loss of the filter cartridge was measured with the experimental setup shown in Fig. 2. As shown in this figure, formaldehyde gas at a constant concentration was introduced into the flow system from a permeater. The pressure loss was measured using a pressure gauge and a flow meter when the density of crushed ECO EXCEL pieces in the cartridge was changed.

## 5. Results and Discussion

Fourier transform infrared spectroscopy (FTIR) spectra of the Si substrate coated with PAH polymer film before and after exposure to formaldehyde for 24 h were measured and compared. The results are shown in Fig. 3. As shown in Fig. 3(a), there is no peak which indicates the existence of C-N-C bonding; however, in Fig. 3(b), the formation of C-N-C bonding was confirmed from the high peak at 1,510 $cm^{-1}$. The results shown in Fig. 3(b) indicate the chemical adsorption of the amino ($-NH_2$) group in the PAH film by the reaction of the –CHO group in the formaldehyde, as shown in the following equation.

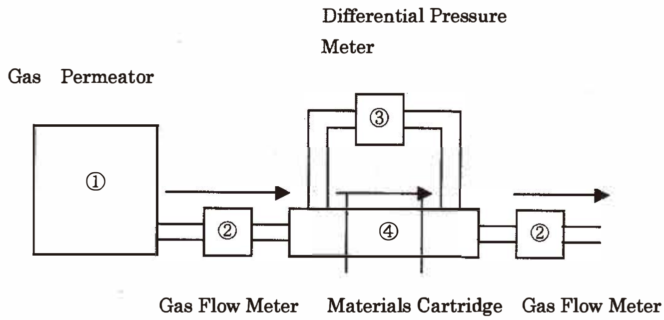


Fig. 2. Experimental setup to measure the pressure loss of the filter cartridge.

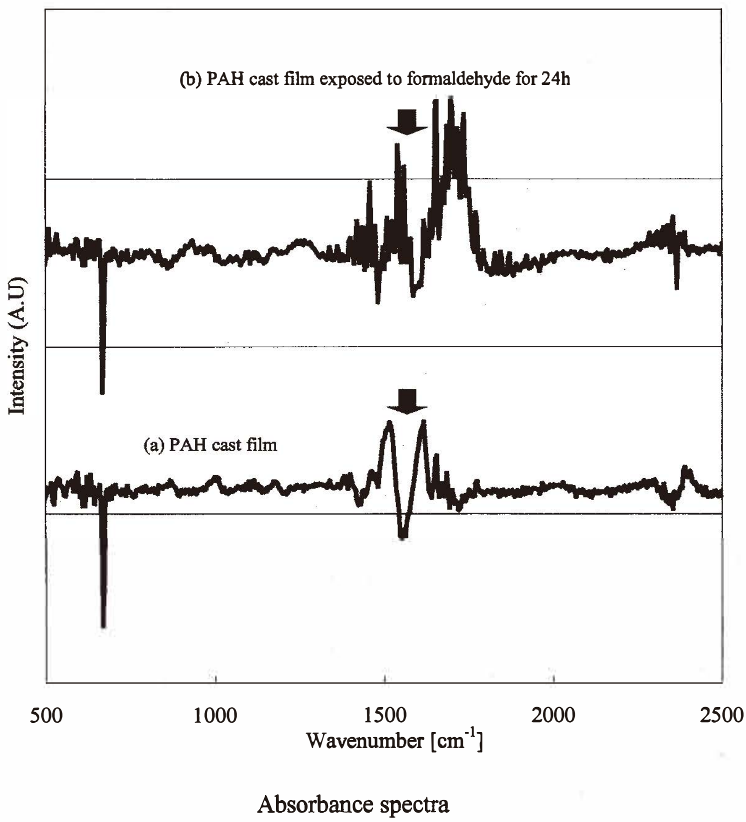


Fig. 3. FTIR spectra of the Si substrate coated with PAH polymer film before and after exposure to formaldehyde for 24 h.

$$\begin{array}{l} 2H-CHO + CH_3CH_2-NH_2 \qquad \overset{OH}{|} \\ \quad \rightarrow CH_3CH_2-\underset{\underset{CH_3}{|}}{NH} + H-\underset{\underset{O}{\|}}{C} \end{array}$$

Details of the adsorption mechanism are currently under investigation. However, this finding agrees well with the observation that, when the filter pieces were dried, the adsorption characteristics were much higher than when the filter pieces contain moisture. We consider that the chemical equilibrium proceeds to the right by removing the water to form the coated film.

Using the chemical adsorption effect of polyelectrolytes, the differences in the gas adsorption of ECO EXCEL pieces with and without the PAH coating were examined. The transient characteristics of the formaldehyde concentration in the chamber are shown in Fig. 4. For each experiment, the concentration of formaldehyde was adjusted to 10 ppm as an initial value. After adjusting the value, the fan under the filter cartridge was turned on. When the filter pieces were not put in the chamber, the concentration of formaldehyde was constant for more than 3 h while the fan continued to work. When the bare ECO EXCEL pieces were placed into the chamber, the concentration decreased to 8 ppm in 10 min and to 5 ppm in 150 min. This was considered to be a result of the physical adsorption of formaldehyde gas by the porous ECO EXCEL pieces. On the other hand, when PAH film was coated on the surface of ECO EXCEL pieces, the concentration of formaldehyde gas decreased to half the initial value within 2 min and to 1 ppm after 25 min. By adjusting the salt concentration in the PAH film coating on the surface of the ECO EXCEL pieces, the adsorption characteristics were markedly improved. The formaldehyde concentration decreased to half the initial value in 1 min, to 1 ppm in 3 min, and to 0.5 ppm in 10 min.

To investigate the mechanism of the improvement in the adsorption characteristics, the surface roughness profiles for each filter material was measured with a profilometer. The surface roughness profiles for each filter are shown in Fig. 5. As shown in these profiles, the surface roughness of the filter material was largest when the salt concentration was 0.07 M. In our experiments, the adsorption by the filter was the highest at this concentration. Therefore, we consider that this improvement is due to the increase in the film thickness and with surface area. The porous ECO EXCEL coated with polyelectrolyte has excellent formaldehyde gas adsorption characteristics.

It has been reported that the typical formaldehyde concentration in a newly built house is approximately 0.5 ppm. The recommended value for formaldehyde concentration from the Japan Ministry of Welfare and the World Health Organization (WHO) is less than 0.08 ppm. To examine the practicality of the newly developed filter, the initial value of the formaldehyde concentration was adjusted to 2 ppm using a permeater (Gas Tech, PD-1B) for the next experiment. The experimental setup is shown in Fig. 2. The amount of flow was 1.5 l/min, and the flow velocity was 1.58 cm/s. The experimental results show that the concentration of formaldehyde downstream was 0.06 ppm and remained constant for more than 10 h. In this case, the density of the ECO EXCEL pieces was 0.112 g/cm$^3$.

Since the chemical filter to remove the formaldehyde consisted of crushed expanded plastic pieces, the removal is expected to be improved as the density of the pieces is increased. In fact, when the volume of the cartridge is constant, the removal may decrease

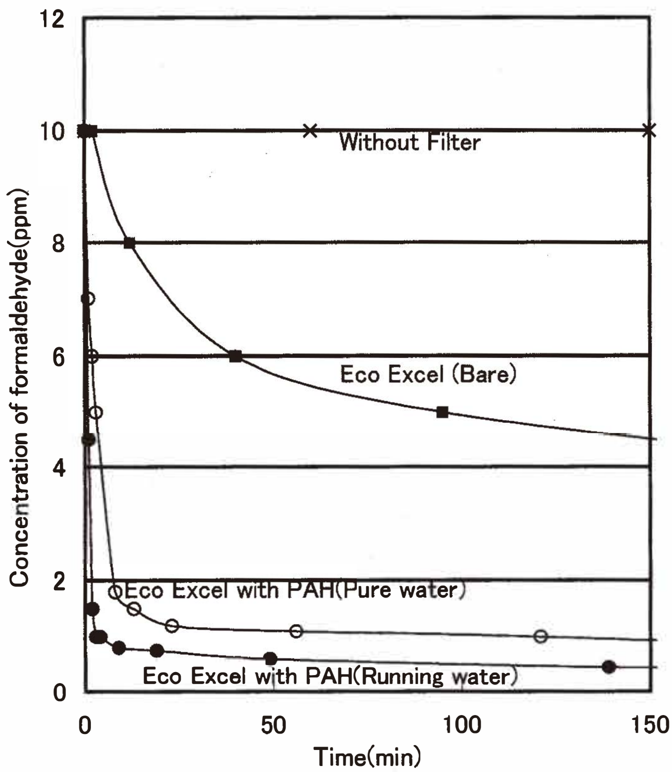


Fig. 4. Transient characteristics of the formaldehyde concentration in the chamber.

because of the increase in the pressure loss. Initially, we predicted that the pressure loss would be increased by coating the surface using polyelectrolyte films. The experimental setup shown in Fig. 2 was used to measure the pressure. The results are shown in Fig. 6. In this figure, the pressure loss of the ECO EXCEL filters formed with and without PAH were compared. Unexpectedly, the results clearly show that the pressure loss of the filter coated with polyelectrolyte was significantly lower than the filter that was not coated. We consider that the porosity of the ECO EXCEL filter was increased by annealing during the coating process of the polyelectrolyte film. These experimental results demonstrate the advantage of the newly developed chemical filter formed by coating the surface of expanded plastic using polyelectrolyte.

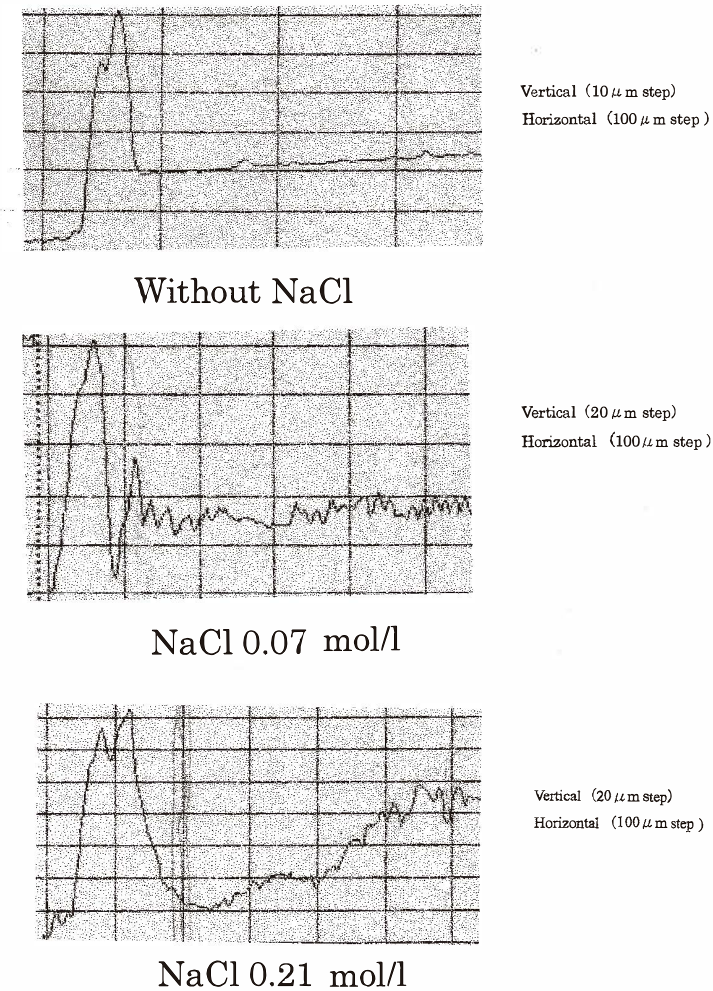


Fig. 5. Comparison of the roughness of the filter surface measured with a profilometer.

## 6. Conclusions

Simple but high-performance chemical filters for air cleaners to remove formaldehyde were successfully developed by coating the surface of ECO EXCEL with polyelectrolyte. The advantages of the newly developed filters are the rapid adsorption of formaldehyde gas and the low pressure loss. An air cleaner using the chemical filter developed herein would

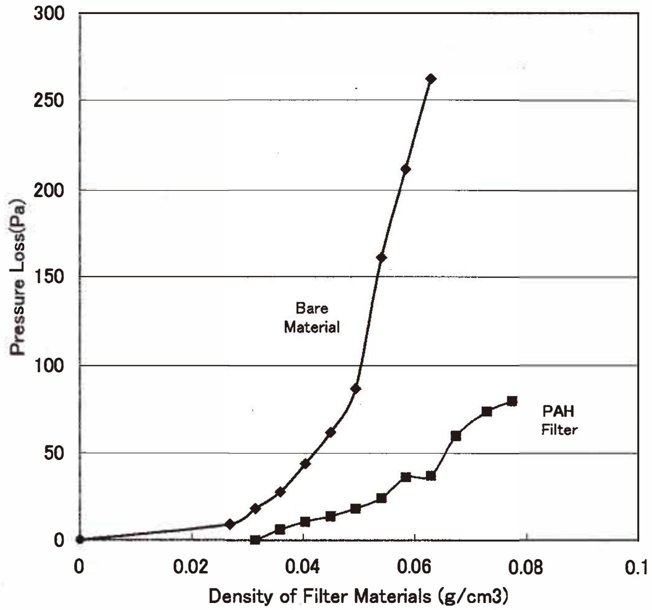


Fig. 6. Pressure loss of the ECO EXCEL filter.

be suitable for newly built house furnishings to prevent sick house syndrome. After the adsorption characteristics decreased due to saturation, the filter can be easily disposed of as it is composed of combustible materials with a low combustion calorific value. By forming the layer-by-layer adsorption film, which is an advantage of polyelectrolytes, not only formaldehyde, but various other toxic gases may be effectively adsorbed into the chemical filter.

## Acknowledgements

Part of this work was supported by financial aid from Plust Corp. The biodegradable expanded plastic was supplied by the company. We thank Mr. Hitoshi Hayashibara, President of Plust Corp. We also thank Professor Keisuke Shimizu and Mr. Satosi Doi of the Intellectual Property Center of Keio University for important advice on this study.

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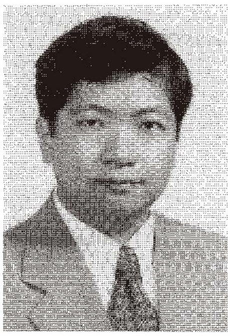

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