

Rapid treatment of trace toluene in air with an air cleaner consisting of a continuous adsorption and desorption concentrator and photocatalytic reactor with a parallel array of nine light sources
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Rapid treatment of trace toluene in air with an air cleaner consisting of a continuous adsorption and desorption concentrator and photocatalytic reactor with a parallel array of nine light sources

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Abstract

To rapidly treat a large amount of air containing trace volatile organic compounds, we previously developed an air cleaner consisting of a continuous adsorption-and-desorption concentrator with a honeycomb rotor loading zeolite particles and a photocatalytic reactor with a parallel array of nine 6W blacklight blue fluorescent lamps. In the present work, this air cleaner was miniaturized and its performance was investigated in the treatment of 1 m³ air containing trace toluene (3.7-10.7 mg⁻³; about 1-3 ppmv). The toluene concentration in an 1 m³ room was rapidly reduced to almost a zero value in only 10 min by adsorption and then photocatalytically decomposed in a 0.022 m³ reactor box. The decomposition activity of the photocatalyst and the ability of the zeolite rotor to adsorb toluene were sufficiently stable in repeated use of the same materials.

Keywords: air cleaner, trace toluene, photocatalytic reactor, continuous concentrator, film-diffusional resistance

1. Introduction

The photocatalytic treatment of volatile organic compounds (VOCs) has several advantages that are hardly given by other treatment methods. For example, the decomposition reaction easily occurs at a room temperature without the addition of other chemical reagents and the reactants are decomposed into carbon dioxide and other minerals (Xu and Shiraishi, 1999; Maira et al., 2003). Therefore, it is reasonable to apply the photocatalytic treatment process to the indoor environment (Obee and Brown, 1995; Shiraishi et al., 2004).

It is difficult to rapidly decompose trace VOCs in a large amount of air toward a zero concentration since a film-diffusional resistance that appears in the vicinity of the photocatalyst surface decreases remarkably the rate of photocatalytic decomposition because of their low VOC concentrations (Shiraishi et al., 2005a). Such a remarkable decrease in the rate of photocatalytic decomposition occurs especially when the VOC concentration drops below 1 ppmv (parts per million in volume). We have previously developed a photocatalytic reactor with a parallel array of nine light sources (6W blacklight fluorescent lamps) individually inserted in glass tubes whose inside surfaces were coated with a transparent thin film of titanium oxide (Shiraishi et al., 2005b). This photocatalytic reactor has the following three major advantages; 1) the photocatalytic reaction takes place in the absence of diffusional resistance, 2) the UV intensity per unit surface area is high, and 3) the UV light that permeates through a glass tube acts on titanium oxide on neighbouring glass tubes. Thus, this photocatalytic reactor can decompose HCHO at a concentration level of ppbv (parts per billion in volume) toward a zero concentration.

Nevertheless, we consider that its performance is not sufficient to purify a large amount of indoor air within a reasonable time. For this reason, we recently developed an air cleaner consisting of a continuous adsorption-and-desorption concentrator and photocatalytic reactor and then treated trace formaldehyde in 10 m³ air (Shiraishi et al., 2003; Shiraishi et al., 2007). As a result, we found that this air cleaner can reduce the HCHO concentration below the environmental guideline within the first 10 minutes by the continuous adsorption-and-desorption concentrator and then decompose the concentrated HCHO toward a zero concentration by the photocatalytic reactor.

The size of the air cleaner may restrict its practical application. If the air cleaner can be miniaturized without changing the principle of the treatment, one can extend its application to the air purification in a narrow space such as cars. Also, miniaturization makes it easier to do experiment and then evaluate the performance of the air cleaner in more details.

In the present work, therefore, we miniaturized the air cleaner and evaluated its performance in the treatment of trace toluene in the air.

2. Experimental

2.1 Materials

A coating solution of titanium oxide was a product of Sundecor Co., Ltd. (Fukuoka, Japan). Toluene was purchased from Wako pure Chemical Industries, Ltd. (Tokyo, Japan). Light sources were 6W blacklight blue fluorescent lamps (210 mm long and 15.5 mm in diameter; FL6BLB-A; Toshiba Co., Ltd., Tokyo, Japan).

2.2 Experimental apparatus and procedure

The coating solution (Matsuo et al., 1990; Ohbuchi et al., 1995) was applied to the inside surfaces of Pyrex glass tubes (28 mm in inside diameter, 210 long, and 1.5 mm in thickness), followed by heating at 100 °C for one hour. The same operation was repeated five times.

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Figure 1 shows a schematic of the air cleaner, which consists of a continuous adsorption-and-desorption concentrator system and photocatalytic reactor system. The continuous adsorption-and-desorption concentrator system contains a rotary adsorbent-loading rotor in honeycomb structure, two electric fans for adsorption and desorption, and an electric heater for desorption of VOCs adsorbed. The rotor (120 mm in diameter and 50 mm thick) consists of a roll of corrugated ceramic paper (3 mm in pitch and 1.7 mm in height) loaded with zeolite particles (6 Å in pore size). The rotor was slowly rotated at a constant rate of 1/12 r.p.m. The air containing trace toluene was continuously supplied into the 3/4 sectional area of the rotor at $0.27 \text{ m}^3 \text{ min}^{-1}$ to adsorb toluene. At the same time, the air in a loop was heated instantaneously and then supplied into the 1/4 sectional area of the rotor to desorb toluene. The desorbed toluene was quickly concentrated in the loop and then decomposed in the photocatalytic reactor fixed in the reactor box (0.022 m^3). The air temperature before entering the rotor was controlled in the range of either 90-120 °C or 130-160 °C.

The photocatalytic reactor system has a parallel array of nine UV light sources (Shiraishi et al., 2005b). The blacklight fluorescent lamp was inserted and fixed in the center of each glass tube whose inside surface was coated with a thin film of titanium oxide. These reaction tube units were arranged in parallel in a plastic transparent box (215mm in length and width and 295mm in height). The air was sucked up through the annulus of each glass tube from the bottom of the box by rotation of an electric fan (13W in electric power consumption) fixed at the upper part of the box, and then discharged from the top. Since the air was allowed to rise through the inside of each glass tube at a linear velocity of air ($>11 \text{ m s}^{-1}$), the photocatalytic reaction took place in the absence of film-diffusional resistance. Since the distance between the photocatalyst surface and light source was only 5 mm, the photocatalytic reaction occurred under a condition of high UV intensity per unit surface area, which made it possible to further lower the concentrations of trace VOCs toward a zero concentration. Moreover, the UV light that permeated a glass tube acted on the photocatalytic film on the neighbouring glass surface, so that the decomposition rate increased by about 20 %.

The air cleaner was set up in an 1 m^3 closed room where a framework of angle bars was covered with vinyl chloride sheets. An electric heating plate for volatilizing toluene and two electric fans for mixing the air were placed. All the experiments were performed according to the following procedure.

- 1) A small amount of toluene was dropped on the heating plate and the room was immediately closed.
- 2) The heating plate was switched on to instantaneously volatilize toluene.
- 3) The electric fans were switched on to sufficiently mix the air in the closed room.
- 4) After two minutes, the air cleaner was switched on to start the decomposition experiment.
- 5) At time intervals, the air sample (10 cm^3) was withdrawn from the 1 m^3 closed room and 0.022 m^3 reactor box.

2.3 Analytical method

The air withdrawn was analyzed using a gas chromatograph equipped with a FID detector (GC-8A; Shimadzu Co. Ltd., Kyoto, Japan) and the toluene concentration was determined.

3. Results and discussion

3.1 Treatment by only a continuous adsorption-and-desorption concentrator

The treatments of the air containing toluene at a concentration of 3.4, 7.8, and 10.6 mg m^{-3} were performed using only the continuous adsorption-and-desorption concentrator, where the desorption temperature was controlled in the range of 90-120 °C. **Figure 2** shows the time courses of the toluene concentrations in the closed room and reactor box in these treatments. **Figure 3** shows the result for the same treatment except for the desorption temperature that was controlled in the range of 130-160 °C. In all the treatments, the toluene concentration in the closed room is lowered to almost a zero value within 10 min. The changes are independent of the desorption temperature. On the other hand, the toluene concentration in the reactor box increases quickly and then becomes almost constant after 10 min. The toluene concentration becomes constant at a higher level when a larger amount of toluene is initially volatilized. In addition, when the adsorption temperature is higher, the toluene concentration reaches a higher level.

3.2 Treatment by a combination of continuous adsorption-and-desorption concentrator and photocatalytic reactor

The treatments of the air containing toluene at a concentration of 3.4, 7.2, and 9.5 mg m^{-3} were performed by a combination of continuous adsorption-and-desorption concentrator and photocatalytic reactor, *i.e.*, the air cleaner, where the desorption temperature was controlled in the range of 90-120 °C. **Figure 4** shows the time courses of the toluene concentrations in the closed room and reactor box in these treatments. **Figure 3** shows the result for the same treatment except for the desorption temperature that was controlled in the range of 130-160 °C. Again, in all the treatments, toluene in the closed room is lowered to almost a zero concentration within 10 min. Also, it is clear that these changes are independent of the adsorption temperature. The rates of treatment for the 1 m^3 air in the closed room are almost the same as those in Fig.2. Therefore, it is obvious that the rapid decrease in the toluene concentration within 10 min is mainly owing to the adsorption of toluene onto the zeolite rotor. When the photocatalytic reactor is switched on, however, toluene after 10 min is decreased at a faster rate and reaches a zero concentration in a shorter time.

The toluene concentration in the reactor box increases quickly, taking a maximum at 10-15 min and decreasing toward a zero value. Obviously, this decrease is due to the photocatalytic decomposition of toluene. The maximum concentration of toluene is higher when a larger amount of toluene is initially volatilized in the closed room. Thus, it takes a longer time until toluene becomes a zero concentration. Moreover, the higher the adsorption temperature, the higher the maximum toluene concentration. However, it takes shorter time until toluene reaches a zero concentration. This is because when a larger amount of toluene is desorbed at a higher temperature, its

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concentration in the reactor box becomes higher, so that toluene on the photocatalyst surface becomes a higher concentration and is then decomposed at a higher rate.

3.3 Change of air temperature in an 1 m³ closed room

In spite of different desorption temperatures, the air cleaner consumed electricity of about 360 W when the concentrator and photocatalytic reactor were simultaneously operated. More than half of the electric power was consumed by the heater, which more or less caused an increase in the air temperature. **Figure 6** shows the time courses of the air temperatures in the closed room when toluene was decomposed by the air cleaner. When the adsorption temperatures were controlled in the range of 90-120 and 130-160 °C, the room temperature at 27 °C increased gradually to 31.5 and 34.5 °C, respectively. Although this increase would be so serious when the air cleaner is used for the air purification in cars because the air volume is usually in the range of 3 to 5 m³, it is important from the standpoint of saving electricity to further investigate an operational method to switch off the heater after adsorbing and then desorbing toluene (Shiraishi et al., 2007).

3.4 Effect of the number of reaction tube units

To further save the electricity energy, we investigated the performance of the air cleaner when the number of reaction tube units was reduced to four, as shown in **Fig.7**. The initial toluene concentration was set at 3.4 mg m⁻³. The experimental result is shown in **Fig.8**, where the time courses of the toluene concentrations in the reactor box are compared between the air cleaners equipped with the photocatalytic reactor having nine and four reaction tube units. Since the time courses of the toluene concentrations in the 1m³ room were almost the same in both the air cleaners, the experimental data is not shown. In the case of nine reaction tube units, the maximum concentration of toluene is lower than that in the case of four reaction tube units because the rate of decomposition is so large that the decomposition is completed in a shorter time. In the case of four reaction tube units, on the other hand, the toluene concentration decreases more slowly because the rate of decomposition is lower. However, it should be noted that toluene is certainly decomposed to a zero concentration, because the photocatalytic decomposition was performed under the condition of negligible film-diffusional resistance and high UV intensity per unit photocatalyst surface (Shiraishi et al., 2005b). Thus, it would be reasonable to use the four reaction tube units according to circumstances.

3.5 Comparison of toluene decomposition using an air cleaner with that using only a photocatalytic reactor

The photocatalytic reactor was placed in the 1m³ room and operated to directly decompose toluene at various initial concentration. **Figure 9** shows the time courses of the toluene concentrations in this decomposition experiment. Toluene is decomposed to a zero concentration in a relatively short time, suggesting that using only the photocatalytic reactor may be sufficient if the amount of the air to be treated is small, for example, almost equal to or less than 1 m³.

It is useful to compare the experimental data in **Fig.9** with the time course of the toluene concentration in the reactor box when the photocatalytic reactor was used

together with the continuous adsorption and desorption concentrator, *i.e.*, when the air was treated using the air cleaner (Figs.4 and 5). At the desorption temperature of 90-120 °C, the toluene concentration in the reactor box when toluene was decomposed using the air cleaner decreases more slowly than does that in the 1m³ room when toluene was directly decomposed using only the photocatalytic reactor. This is due to the fact that the photocatalytic decomposition occurred slowly at almost the same rate as in the direct decomposition of toluene in the 1 m³ room using only the photocatalytic reactor, because the toluene concentration in the reactor box was at almost the same level as that in the 1 m³ room when only the photocatalytic reactor was used. The main cause of why no toluene was satisfactorily concentrated despite the use of the continuous adsorption-and-desorption concentrator is probably no rapid desorption of toluene from the rotor even when the toluene concentration in the reactor box was reduced because of low desorption temperature. At the desorption temperature of 130-160 °C, on the other hand, the toluene concentration in the reactor box decreases slightly faster than that in the 1 m³ room when toluene was directly decomposed using only the photocatalytic reactor; compare the runs where the concentrations of toluene just after volatilized into the 1 m³ room are almost the same.

From the standpoint of the time courses of the concentrations of toluene substantially remaining in the 1 m³ space, it is a certain that there is no remarkable difference between the experimental data in the decompositions of toluene using only the photocatalytic reactor and using the air cleaner. From the stand point of practical application, on the other hand, it is meaningful to compare the time courses of the toluene concentrations in the 1 m³ space. In this respect, it is clear that the air treatment using the air cleaner is superior to that using only the photocatalytic reactor because the air cleaner can drop the toluene concentration below its environmental guideline (0.26 mg m⁻³) in only 10 min. In general, if the amount of air is larger, the treatment time using the air cleaner certainly becomes shorter than that using only the photocatalytic reactor (Shiraishi et al., 2003; Shiraishi et al., 2005b; Shiraishi et al., 2007).

3.6 Durability of the air cleaner

All the decomposition experiments were performed using the same zeolite rotor and photocatalytic reactor over six month. The total number of the runs was more than one hundred. Nevertheless, no distinct decrease was observed in the decomposition activity of the photocatalyst and the ability of the zeolite rotor to adsorb toluene, indicating that these materials can stably be used over a long term.

4. Conclusion

The experimental results clearly show that the miniaturized air cleaner has a remarkably high performance. The toluene concentration in the 1 m³ room can be decreased very quickly to almost a zero value in the first 10 min, which is owing to the adsorption by the continuous adsorption-and-desorption concentrator. The toluene concentration in the reactor box is initially increased quickly but certainly decreased to a zero value, which is owing to the photocatalytic decomposition. The

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decomposition activity of the photocatalyst and the ability of the zeolite rotor to adsorb toluene are sufficiently stable in repeated use of the same materials.

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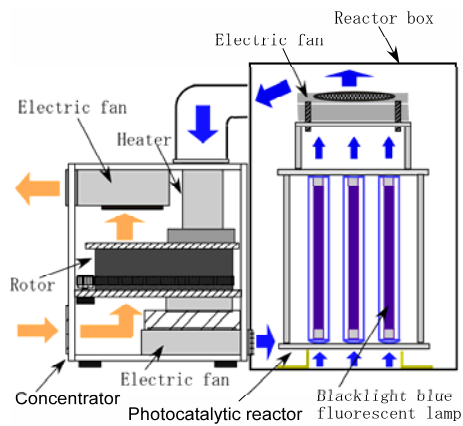


Fig.1 A schematic diagram of air cleaner

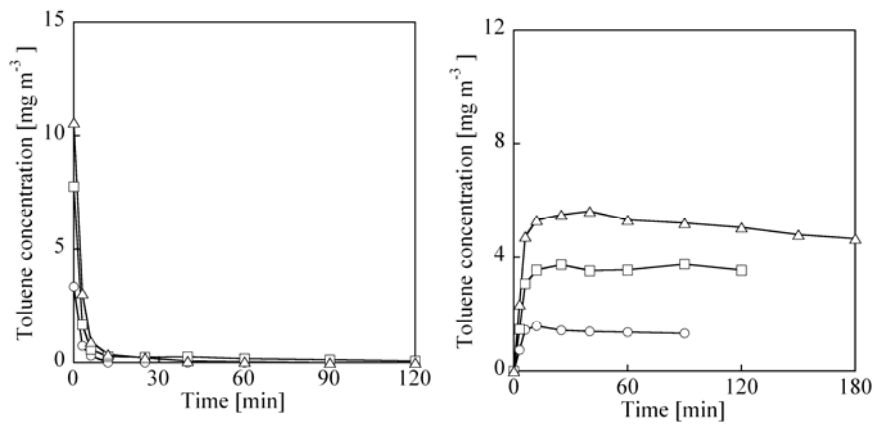


Fig.2 Time courses of toluene concentrations in an 1 m³ room (left) and reactor box (right) in the continuous adsorption and desorption treatment of air containing a small amount of toluene at different initial concentrations. Toluene was desorbed from the rotor at 90-120 °C

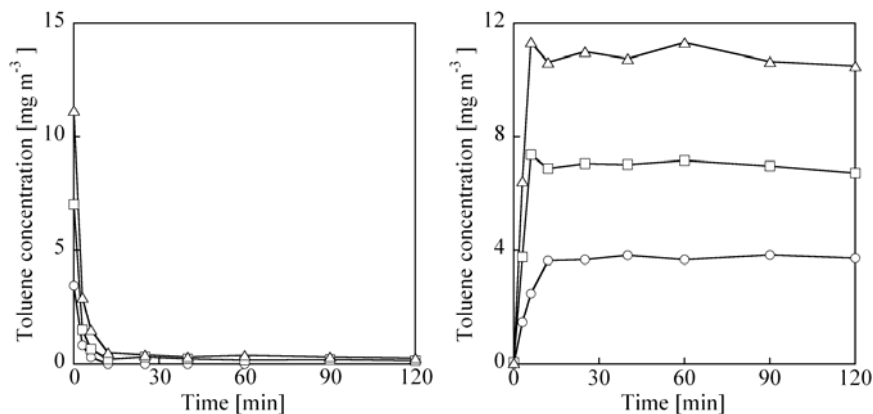


Fig.3 Time courses of toluene concentrations in an 1 m³ room (left) and reactor box (right) in the continuous adsorption and desorption treatment of air containing a small amount of toluene at different initial concentrations. Toluene was desorbed from the rotor at 130-160 °C.

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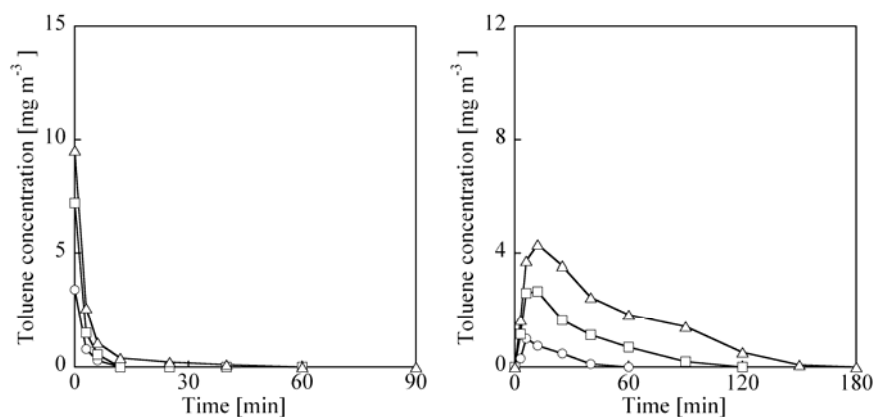


Fig.4 Time courses of toluene concentrations in an 1 m³ room (left) and reactor box (right) in the air cleaner treatment of air containing a small amount of toluene at different initial concentrations. Toluene was desorbed from the rotor at 90-120 °C.

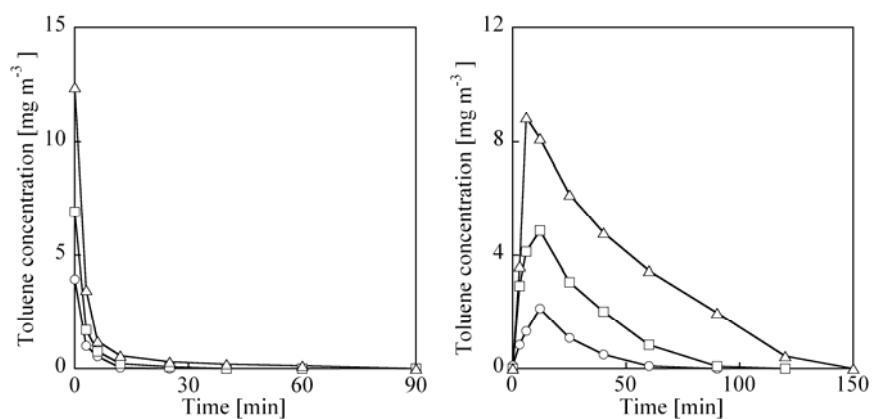


Fig.5 Time courses of toluene concentrations in an 1 m³ room (left) and reactor box (right) in the air cleaner treatment of air containing a small amount of toluene at different initial concentrations. Toluene was desorbed from the rotor at 130-160 °C.

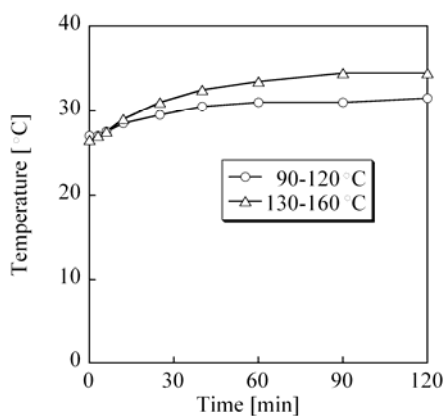


Fig.6 Changes in the temperature in an 1 m³ room when toluene was desorbed from the rotor at 90-120 and 130-160 °C.

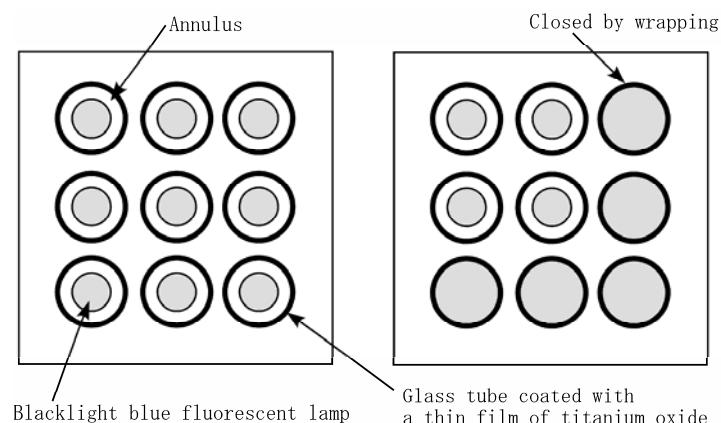


Fig.7 Arrangement of reaction tube units in a photocatalytic reactor with a parallel array of nine UV-light sources in the use of nine reaction tube units (left) and four reaction tube units (right). The open circle with a thick line represents reaction tube units where the flow channel is open and the UV light is switched on, while the closed circle with a thick line represents the reaction tube units where the flow channel is closed and the UV light is switched off.

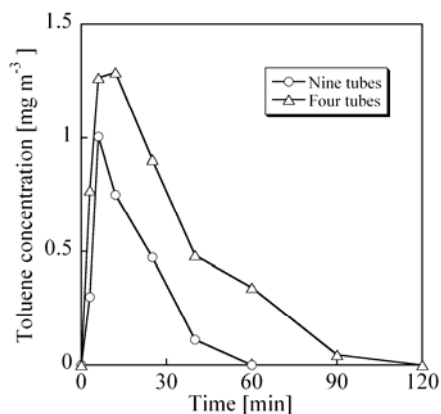


Fig.8 Time courses of toluene concentrations in a reactor box in the air-cleaner treatment of air containing toluene at 3.4 mg m^{-3} at different initial concentrations. Toluene was desorbed from the rotor at $90\text{--}120 \text{ }^\circ\text{C}$ and the photocatalytic reactor had nine and four reaction tube units.

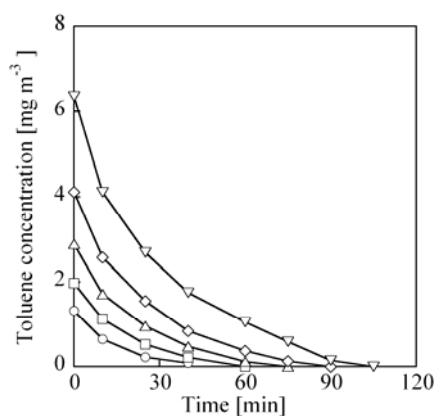


Fig.9 Time courses of toluene concentrations in an 1 m^3 room in the photocatalytic treatments of air containing a small amount of toluene using a photocatalytic reactor with a parallel array of nine light sources.