Influence of UV wavelength, light intensity and humidity on photocatalytic degradation of toluene by using hybrid titania-based film

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ABSTRACT

A kind of hybrid titania-based film composed of mordenite and Degussa P-25 titania with weight ratio of 3:1 was used in the investigation. The effects of UV wavelength, UV light intensity and humidity on the degradation of dilute gas-phase toluene by using the film were experimental studied. Three conclusions were reached: (1) for the UV wavelength of 254 and 365 nm, the fractional conversion of toluene is linear with the light intensity for two light intensity regions (the slop of fractional conversion–light intensity curve for light intensity below 1.64 mW/cm^2 is obviously larger than that for light intensity above 1.64 mW/cm^2); (2) for the same light intensity, shorter wavelength leads to higher fractional conversion of toluene; (3) there exists an optimal humidity, 7600 mg/m^3, for the photo-degradation of toluene with 254 nm UV light under the test condition.

INDEX TERMS

Indoor air quality; Toluene; Photocatalytic; Titania

INTRODUCTION

With the increasing use of synthetic building material and furnishings that emit volatile organic compounds (VOCs), the VOC concentrations in indoor air tend to be higher than the level permitted by the standards**.** These may cause general symptoms, such as headache; eye, nose or throat irritations; dry cough; dizziness and nausea; difficulty in concentrating and tiredness (WHO, 1989; Kadosaki *et al.*, 1999; Meininghaus *et al.*, 1999; Kim *et al.*, 2001). Those may also do great harm to respiratory system, cardiovascular system and nervous system of human beings (Molhave, 1989).

Photocatalytic oxidation (PCO) is an innovative and promising approach to eliminate VOCs in the indoor air. Titania is a commonly used photocatalyst because of its high photoactivity, good stability and low price (Han and Zhao, 1999). Many researchers have reported that ultraviolet illuminated titania or hybrid photocatalyst (composed by titania and adsorbents) exhibited high activity to decompose many of the common VOCs in indoor air (Obee, 1996; Yoneyama and Torimoto, 2000; Maira *et al.*, 2001; Bouzaza and Laplanche, 2002). Toluene is a major indoor air contaminant which presents a significant health effect. Photooxidation of the sub-parts-per-million by volume contaminant level of toluene by titania has been investigated by some researchers (Obee, 1996; Maira *et al.*, 2001; Bouzaza and

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Laplanche, 2002). However, little research has been conducted on the photo-oxidation of toluene by the hybrid photocatalyst under low toluene concentration as far as we know.

In the present study, a kind of hybrid photocatalyst (composed of mordenite and Degussa P-25 titania with weight ratio of 3:1) was used to decompose ppmv level toluene in a glass plate reactor. The effects of UV wavelength, UV light intensity and humidity on the photo-degradation of dilute gas-phase toluene in a glass plate photoreactor were investigated.

METHODS

Film Preparation

The hybrid photocatalyst is composed of mordenite and Degussa P-25 titania with a weight ratio of 3:1. The hybrid photocatalyst film was deposited on a glass plate with dimensions of 90.0 mm length, 25.0 mm width and 4.0 mm thickness by a wash-coated process. Before the process, the glass plate was washed in 10 wt% NaOH solution and distilled water by sequence, and then dried in an oven at 70°C until the glass surface was dry. The wash-coat was 1.25 wt% of the hybrid photocatalyst in pure ethyl alcohol. The wash-coat was stirred before the glass plate was dipped in it. The film was prepared by dipping the glass plate in the wash-coat several times, and then dried in the oven at 70°C for 2 min. This process was repeated until a 0.49 mg/cm^2 film (per side) was achieved.

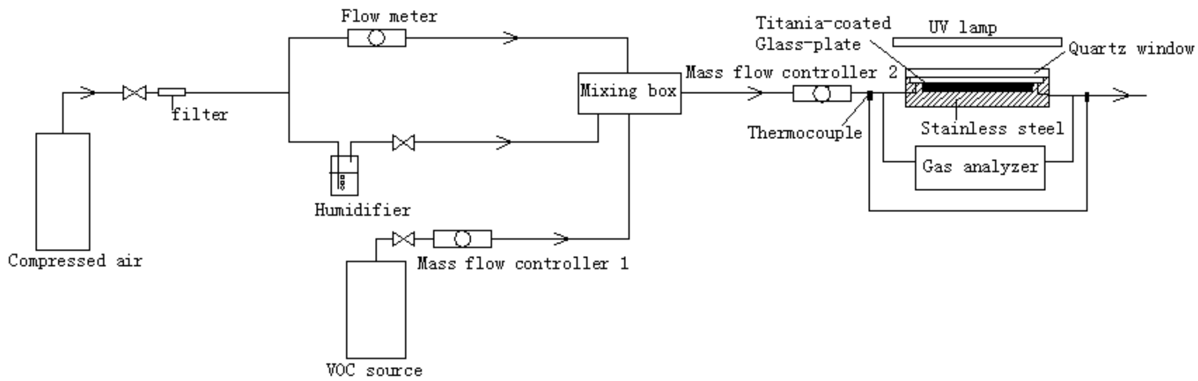

Figure 1 Schematic of the glass plate photoreactor.

Apparatus

The decomposing effect of toluene was measured in a glass plate reactor (Figure 1). Two parallel germicidal lamps (254 nm peak wavelength, 20 W) or black light lamps (365 nm peak wavelength, 20 W) provided UV irradiation for the photoreactor. Adjusting the distance between the lights and the photoreactor varied the light intensity. The light intensity at the reaction surface was measured by using UV power meters. Synthetic air (mixture of high-purity nitrogen and oxygen with volume ratio 79:21) was supplied from a compressed cylinder. The air passed through a filter and was then divided into two streams. One stream passed through a flow meter while the other passed through a humidifier to control the tested air on the desired humidity level. Toluene was mixed with the synthetic air in the mixing box and then the mixed gas was supplied through a mass flow controller to the photoreactor. Mass flow controller 2 controlled the total airflow rate. The photocatalyst-coated glass plates were placed in a well (25.0 mm wide, 380.0 mm long and 4.0 mm deep) made of stainless steel and covered with a quartz window (transmittance is 0.80 at 254 nm and 0.92 at 365 nm). The space between the photocatalyst-coated glass plate and the quartz window was the flow channel with cross-sectional area of 25.0 × 2.0 mm^2. A photoacoustic multigas monitor and a multipoint sampler and doser were used to measure the concentrations of water vapour, carbon dioxide, carbon monoxide and toluene at the inlet and outlet of the photoreactor. Two copper–constantan thermocouples were installed at the inlet and outlet of the photoreactor, respectively. A HP34970A data logger recorded the temperature.

Controls

The procedure for all the experiments were as follows: (1) airflow was started; (2) after the photoreactor inlet and outlet humidity readings were steady and equal, the toluene was introduced; (3) when the photoreactor inlet and outlet toluene concentrations were approximately equal, the UV lamps were turned on; (4) after the inlet and outlet toluene levels again reached steady state, the UV lamps were turned off and the toluene flow was stopped; (5) the photoreactor was flushed with the synthetic air for 15 min, and then airflow was stopped.

Experimental Condition

All experiments were performed at room temperature 25.0–27.0°C. The relative humidity was set to a common air conditioning level of about 47%. The inlet toluene concentration was kept on about the same level of 0.75–0.85 ppmv. The total air volumetric flow rate was 4.00 l/min, which formed a laminar flow with Reynolds number (based on the 25.0 × 2.0 mm^2 flow cross-sectional area) of 330 in the photoreactor. The UV light intensity at the glass plate surface varied from 0.62 to 4.79 mW/cm^2. Three pieces of same coated glass plates were placed in series in the photoreactor. The other glass plate of same size without photocatalyst was placed in front of the coated glass plates to make the reaction part in the fully developed laminar flow region.

Data Analysis

For the data measured by the glass plate photoreactor, the following photocatalytic fractional conversion, ε, is used (Zhang *et al.*, 2003):

$$\varepsilon = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \qquad (1)$$

where C_{in} is the average toluene concentration at the inlet under the steady state when the UV lamps are lighting and C_{out} is the corresponding value at the outlet.

RESULTS AND DISCUSSION

Effect of UV Light Intensity

Photocatalytic reaction occurs in two regimes in association with UV light intensity: first-order regime at lower light intensity and half-order regime at higher light intensity (Kim and Hong, 2002). For the first regime, the electron–hole pairs consumption of chemical reaction is much larger than that of recombination; whereas the consumption is opposite at the second regime. In Figure 2, the influences of UV light intensity and UV wavelength on the fractional conversion of toluene are shown. For the light intensity lower than 1.64 mW/cm^2, the fractional conversion of toluene is linear with the light intensity for either the germicidal lamp or the black light lamp. The efficiency of light diminished with the increase of the light intensity above 1.64 mW/cm^2 for the germicidal lamps. The light intensity increases about two times, but the fractional conversion only increases 12% for the germicidal lamps. These results coincide with the two-regime theory of the photocatalytic reaction qualitatively. This hybrid photocatalyst shows similar property with the pure titania when the UV light intensity is varied.

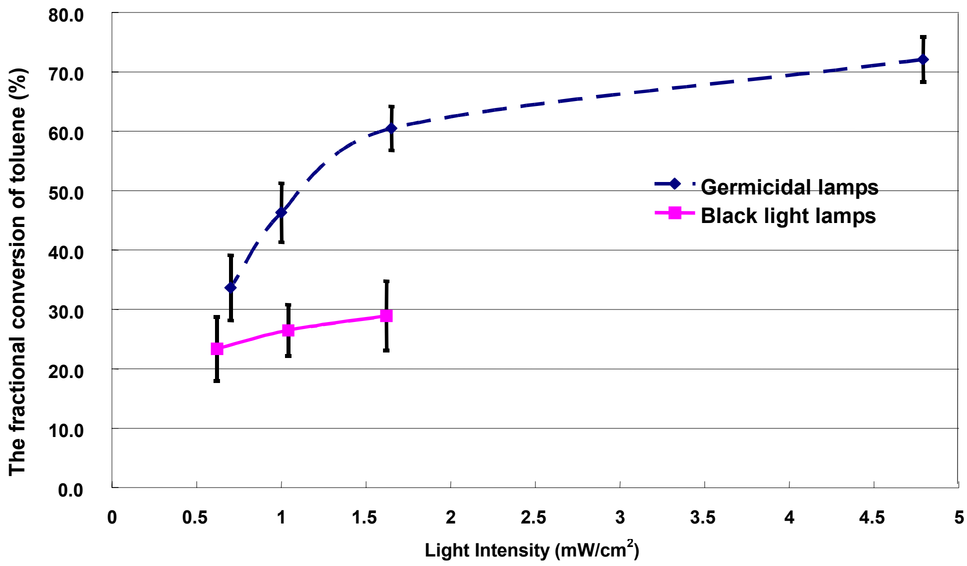

Figure 2 Effect of UV light intensity and UV wavelength on the fractional conversion of toluene using the hybrid photocatalyst: 0.75–0.85 ppmv inlet toluene; 4.00 l/min airflow rate; 47% relative humidity; 25.0–27.0°C air temperature.

Effect of UV Wavelength

Photon energy from the irradiation source is an important factor in photocatalytic reaction. UV wavelength affects the photon energy as Eqn (2) shows.

$$E = hc/\lambda \qquad (2)$$

where E is energy of a photon, J; h is Planck's constant which equals 6.626×10^{-34} J s; c is speed of light in vacuum which equals 2.998×10^{8} m/s and λ is the wavelength of photon, m. Shorter wavelength means higher photon energy. The wavelength of the germicidal lamp ranged from 200 to 300 nm with the maximum light intensity at 254 nm, and that of the black light lamp ranged from 315 to 400 nm with the maximum light intensity at 365 nm. Both lamps had sufficient energy to promote electrons from valence band to the conduction band of titania. As shown in Figure 2, it is found that shorter wavelength shows higher fractional conversion of toluene at about the same light intensity, which means higher reaction rate of decomposing toluene. For light intensity below 1.64 mW/cm^2, higher light intensity larger differences between two kinds of lamps. The effect of UV wavelength on the photo-degradation of toluene can also be seen from the product of the photoreaction. For the germicidal lamps, the increase of gas-phase carbon dioxide and carbon monoxide concentrations at the outlet could be observed obviously, but this phenomenon was not observed for the black light lamps. The photo-degradation of toluene under irradiation with shorter wavelength was more complete. This is because the photons with higher energy can promote holes with higher oxidizing ability. Furthermore, the luminous efficiency of the germicidal lamp is much higher than that of the black light lamp, because the light intensity at the peak wavelength for the germicidal lamp is larger than that of the black light lamp with the same distance from the lamps. Thus, the germicidal lamp is a better candidate of irradiation sources for photoreactors.

Effect of Humidity

In order to investigate the effect of humidity on the photoactivity of the hybrid photocatalyst, the water vapor concentration varied from 0 to 14900 mg/m^3. Figure 3 shows the experiment results for water vapour concentration from 4000 to 14900 mg/m^3. The results of 0 and 2000 mg/m^3 water vapour concentration are not shown because there a deactivation of the hybrid photocatalyst is observed. The outlet concentration of toluene increased gradually until it was equal to the inlet concentration. The time course of the deactivation for the lower humidity was shorter. The catalytic activity of the hybrid photocatalyst recovered after flowing humid air over the UV-irradiated photocatalyst film for 1 h. This showed that the water stabilizes the hybrid photocatalyst during the PCO of toluene. It is shown in Figure 3 that there is an optimal water vapour concentration of 7600 mg/m^3 for the photo-degradation of toluene for the cases studied. This may be the result of competitive adsorption of toluene and water vapour at the surface of the photocatalyst.

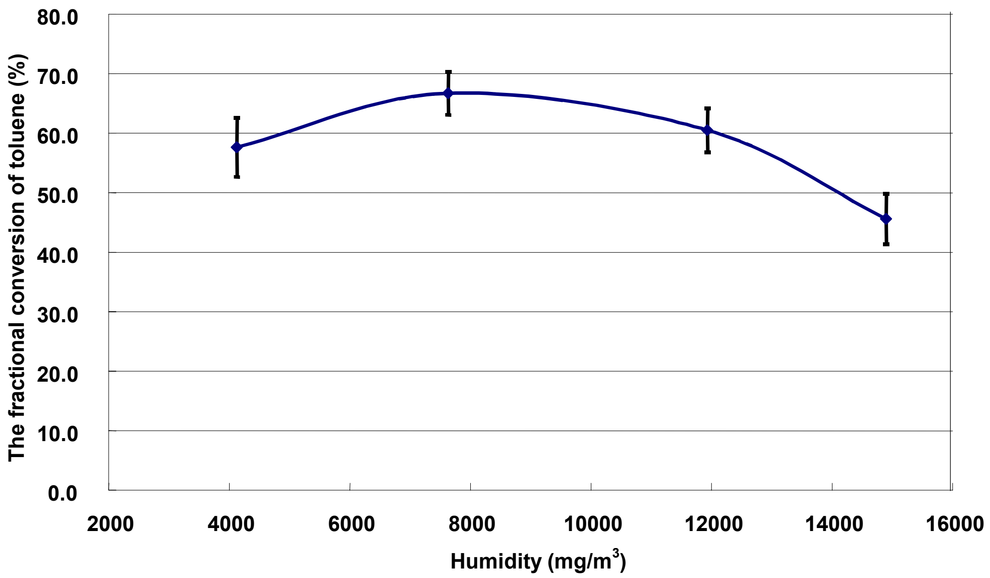

Figure 3 Effect of water vapour concentration on the fractional conversion of toluene: 0.75–0.85 ppmv inlet toluene; 4.00 l/min airflow rate; 1.64 mW/cm^2, 254 nm UV light intensity; 25.0–27.0°C air temperature.

CONCLUSIONS

From the experimental results for the hybrid photocatalyst composed by modernite and P25 with weight ratio of 3:1 decomposing dilute gas-phase toluene in a glass plate reactor, three conclusions were drawn:

1. For the UV wavelengths of 254 and 365 nm, the fractional conversion of toluene is linear with the light intensity for two light intensity regions (the slop of fractional conversion–light intensity curve for light intensity below 1.64 mW/cm^2 is obviously larger than that for light intensity above 1.64 mW/cm^2).
2. For given light intensity, shorter wavelength leads to higher fractional conversion of toluene.
3. There exists an optimal humidity, 7600 mg/m^3, for the photo-degradation of toluene with 254 nm UV light under the test conditions.

ACKNOWLEDGEMENTS

This work is supported by National Nature Science Foundation of China (Grant No. 50276033) and by Tsinghua University (Grant No. 200007005).

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