Removal of Low-Concentration Formaldehyde by a Fiber Optic Illuminated Honeycomb Monolith Photocatalytic Reactor

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ABSTRACT

In this study, we systematically investigated the removal of low-concentration formaldehyde by the fiber optic illuminated honeycomb monolith photocatalytic reactor and the influential factors including formaldehyde concentration (0.8–2.0 ppm), relative humidity (RH30–70%), and air flow rate (800–1600 mL/min). The experimental results of various formaldehyde concentrations indicate the kinetic fits the Langmuir–Hinshelwood model, and the rate constants (k) under RH30%, 50% and 70% are 1.09, 1.37, and 1.68 μ-mole/m²/s, respectively. The Langmuir adsorption constants of formaldehyde under RH30%, 50% and 70% are 4.10 × 10⁻³, 2.98 × 10⁻³ and 2.14 × 10⁻³ ppm⁻¹, respectively. Increasing relative humidity has a positive effect on the rate constant of photocatalytic oxidation k, which is relevant to the enhancement effect of relative humidity on the formation of hydroxyl radicals. On the other hand, the formaldehyde conversion and Langmuir adsorption constant of formaldehyde decrease with the increase of relative humidity, which may be associated with the competition between formaldehyde and water molecules for the adsorption sites on the surface of TiO₂ photocatalyst. Because the air flow rate was low (≤ 1600 mL/min), the gas retention time (≥ 7.7 sec) was long enough for the reactor to achieve a high formaldehyde conversion (≥ 92%), but the breakthrough of formaldehyde might occur when air flow rate > 4200 mL/min.

Keywords: Indoor; Formaldehyde; Influential factors.

INTRODUCTION

Formaldehyde is a common indoor air pollutant (Spengler and Sexton, 1983; Tang et al., 2009; Salthammer et al., 2010). The adverse health effects of formaldehyde having been reported include: acute poisoning and mucus membrane irritation; eye conjunctivitis and irritation; irritation to upper and lower airways; degenerative, inflammatory and hyperplastic changes of the nasal mucosa in human bodies; nasal and pharyngeal diseases; pulmonary function damages; allergic asthma; neurotoxicity and probably reproductive toxicity and hematotoxicity (Tang et al., 2009; Salthammer et al., 2010). The preponderance of reliable reviews of the formaldehyde literature have suggested that a concentration below 0.3 ppm is required to protect all individuals from eye irritation. According to Golden’s research (2011), 0.1 ppm is the recommended indoor formaldehyde exposure limit for virtually everyone for odor detection and sensory irritation.

Formaldehyde has been classified as a human carcinogen that leads to nasopharyngeal cancer by the International Agency for Research on Cancer (IARC) based on many comprehensive investigations and large-scale international human studies (IARC, 2006). An evidence based assessment of an extremely large body of data concludes that the formaldehyde indoor exposure limit of 0.1 ppm should protect even the mostly susceptible persons from both irritation and any potential cancer risk (Golden, 2011). Currently, the indoor formaldehyde exposure limit of 0.1 mg/m³ (0.08 ppm) have been recommended by the World Health Organization (WHO), and several countries, including China, Japan, Taiwan (R.O.C.) and the United Kingdom, have adopted the WHO’s recommendation value. Canada, Germany and Singapore have a higher indoor exposure limit of 0.123 mg/m³ (0.1 ppm). France even has a stricter indoor air quality guideline (IAQG) [Short-term (2 hours) IAQG: 50 µg/m³; Long-term (> 1 year) IAQG: 10 µg/m³]. Even with these standards, however, the formaldehyde exposure in some fast developing countries, such as China, remains high (Tang et al., 2009; Salthammer et al., 2010). Consequently, many promising indoor air cleaning techniques for the removal of formaldehyde, including photocatalytic oxidation, have been extensively investigated (Noguchi
and Fujishima, 1998; Ao et al., 2004; Yu et al., 2006a; Zuo et al., 2006; Hodgson et al., 2007; Wang et al., 2007; Yang et al., 2007a; Shie et al., 2008; Zhang and Liu, 2008; Chen et al., 2009; Mo et al., 2009; Lu et al., 2010; Aïssa et al., 2011; Bourgeois et al., 2012; Han et al., 2012; Dozzi et al., 2013). In the photocatalytic oxidation, the ultraviolet (UV) radiation with energy above the band-gap of titanium dioxide (TiO₂) (wavelength < 385 nm) can induce the formation of electron-hole pairs and generate hydroxyl radicals further on the UV-illuminated TiO₂ surface, and then the oxidation of organic contaminants to H₂O, CO₂ and other final products is mediated by hydroxyl radicals (Anpo et al., 1991; Yu et al., 2006b; Zuo et al., 2006; Hodgson et al., 2007; Yu and Lee, 2007; Mo et al., 2009; Sun et al., 2010; Yu et al., 2010; Kandiel et al., 2013; Klementova and Zlamal, 2013; Silva et al., 2013). The key photocatalytic oxidation and reduction processes in the degradation of formaldehyde can be expressed as what follows (Noguchi and Fujishima, 1998; Ao et al., 2004):

\[
\text{TiO}_2 \xrightarrow{\text{hv}} e^- + h^+
\]

**Oxidation**

\[
2h^+ + 2H_2O \rightarrow 2OH^- + 2H^+
\]

\[
HCHO + OH^- \rightarrow HCOO^- + H_2O
\]

\[
HCOO^- + OH^- \rightarrow HCOOH
\]

\[
HCOOH + 2h^+ \rightarrow CO_2 + 2H^+
\]

**Reduction**

\[
4e^- + O_2 + 4H^+ \rightarrow 2H_2O
\]

Accordingly, the oxidation of one formaldehyde molecule to H₂O and CO₂ requires four electron-hole pairs stoichiometrically and the participation of H₂O molecules.

A variety of photocatalytic reactors have been designed for indoor air purification, such as plate (Yu et al., 2006b; Zuo et al., 2006; Yang et al., 2007b; Yu and Lee, 2007; Chen et al., 2009; Yu et al., 2010), honeycomb monolith (Hodgson et al., 2007; Wu et al., 2013; Yu et al., 2014), annular (Yang et al., 2007a; Shie et al., 2008), packed-bed (Han et al., 2012), and optical fiber (Bourgeois et al., 2012). Among these reactors, the performance of plate [once-flow removal efficiency (ORE): 11–19.6% (Yang et al., 2007b); 50% (Chen et al., 2009)] and annular [ORE: 8–14% (Yang et al., 2007a)] reactors on formaldehyde decomposition were lower than those of the packed-bed [ORE: 40–90% (Han et al., 2012)], optical fiber [ORE: 60% (Bourgeois et al., 2012)] and the honeycomb monolith type [ORE: 35–86% (Hodgson et al., 2007; Mo et al., 2009)]. In our previous study, we had successfully improved the removal efficiency of α-pinene to > 90% by using a fiber optic illuminated honeycomb monolith photocatalytic reactor (Yu et al., 2014). In this study, we focused on the removal of low-concentration formaldehyde with the same reactor. We also conducted a series of experiments to investigate the impacts of relative humidity, formaldehyde concentration, and air flow rate, because these factors could significantly affect the performance of photocatalytic reactor.

**MATERIALS AND METHODS**

The experimental system (Fig. 1) of this study includes a compressed air cylinder (21% O₂, 79% N₂, Air Products San Fu Co., Ltd., Taiwan), a humidity control system, a formaldehyde vapor generating system, a fiber optic illuminated honeycomb monolith photocatalytic reactor, and a gas sampling and analysis system. The air from the compressed air cylinder was pretreated by a homemade diffusion dryer, a homemade active carbon cartridge and a High Efficiency Particulate-Free Air (HEPA) capsule (PN 12144, Pall Cooperation, USA) to ensure that the air was clean. The humidity control system consisted of a homemade aeration tank and two mass flow controllers (Brooks Model 5850E, Brooks Instrument, USA) which controlled the flow rates of saturated and dry air. The relative humidity of the experimental system was being monitored by a Hydrometer (A2 & 100A, ROTRONIC AG, Switzerland). The formaldehyde vapor generating system was composed by a Gastight® syringe (Hamilton Company, USA), a syringe pump (KDS-100 Infusion Pump, KD Scientific, USA), a temperature control device (FGH051-020, OMEGALUX, USA), a heater, and a homemade gas mixing chamber. The formaldehyde solution (37% formaldehyde) in the Gastight® syringe was introduced stably into the heater with the syringe pump and then evaporating into vapor. The vapor of formaldehyde was flushed into the gas mixing chamber and then mixed with other gas streams of the experimental system.

The honeycomb monolith photocatalytic reactor (Wu et al., 2013) contained a honeycomb ceramic support, fiber optics, and three UV lamps (Heraeus, Germany), and was maintained at room temperature in an incubator (Low Temperature Thermostatic B.O.D. incubator, LE-509, YIHER Inc. Taiwan). The honeycomb monolith was coated with Evonik (Degussa) P25 TiO₂ photocatalyst and the coating area was 0.65 m². P25 TiO₂ has an average particle size of 30 nm, a surface area of 47 m²/g, and a mixed-crystal structure of 70% of anatase and 30% of rutile. The Gas sampling and analysis system consisted of a Gas Sampling Bulk (1000 mL, SUPELCO Inc., USA) and a formaldehyde concentration detector (Formaldermertm htV-M, PPM Technology Ltd., Wales, UK). The Formaldermertm has a detection range of 0.01 to 10 ppm and a resolution of 0.001 ppm. This instrument was calibrated by the manufacture routinely. After the calibration, 94% of all the Formaldermertm readings meet the NIOSH criteria for an acceptable method (Range studied: 0.025 to 2.45 mg/m³ (22-L samples); overall precision (Sr): 0.057; accuracy: ±19.0%) (Tucker, 2002) when measuring 0.3 ppm of formaldehyde over a relative humidity range of 25–70%.

The formaldehyde concentrations at the outlet of the photoreactor were measured when the system achieved a steady state to evaluate the formaldehyde conversion as what follows:
Formaldehyde conversion (%) = \frac{C_{\text{off}} - C_{\text{on}}}{C_{\text{off}}} \times 100\% \quad (1)

where \(C_{\text{off}}\) and \(C_{\text{on}}\) are the formaldehyde concentrations when the UV lamp was off and on (after the UV lamp was turned on for 30 minutes), respectively. Fig. 2 shows the variation of formaldehyde concentration during one of the experiments (RH = 50%, Formaldehyde = 0.8 ppm, air flow rate = 1400 mL/min). When the UV lamp was off, the formaldehyde concentration was 0.801 ± 0.010 ppm. When the UV lamp was on, the formaldehyde concentration reduced to 0.048 ± 0.009 ppm. The photocatalytic oxidation rate \(r\) (μ-mole/m²/s) of the formaldehyde was calculated by the following equation:

\[
r = \frac{(C_{\text{off}} - C_{\text{on}}) \times Q}{R \times Ac}
\]

in which \(Q\) (m³/s) is the air flow rate of the photoreactor; \(Ac\) (0.65 m²) is the TiO₂ coating area; \(R\) is the unit conversion factor (24500 ppm-m³/mol, which is derived from the fact that 1 ppm = 4.09 × 10⁻³ mol/m³).

RESULTS AND DISCUSSION

Effect of Relative Humidity

In general, humidity (water molecule) has both positive and negative effects on the photocatalytic reaction. First, increasing humidity is beneficial to the generation of highly oxidative hydroxyl radicals (Yu et al., 2006b; Yu and Lee, 2007; Sun et al., 2010; Yu et al., 2014), and may increase the concentration of adsorbed hydroxyl groups, which may scavenge adsorbed organic chemicals, on the surface of TiO₂ (Ameen and Raupp, 1999). On the other hand, water molecules may compete with the formaldehyde molecules for the adsorption sites on the surface of TiO₂, and thus result
Fig. 2. The variation of formaldehyde concentration during the experiment (RH = 50 ± 3%; Formaldehyde = 0.8 ppm; air flow rate = 1400 mL/min). When the UV lamp was off, the formaldehyde concentration was 0.801 ± 0.010 ppm. After the UV lamp was on, the formaldehyde concentration would drop to 0.048 ± 0.009 ppm.

in the decrease of photocatalytic oxidation rate. Attaching water onto TiO₂ would lead to the structural changes in the surface band bending that enhanced the recombination of photocatalytic-induced electron-hole pairs, and therefore reduced the photocatalytic efficiency (Anpo et al., 1991).

Fig. 3 shows the effect of relative humidity on the formaldehyde conversion of photocatalytic oxidation. The formaldehyde conversion decreased with the increase of RH in the RH region examined, indicating that raising relative humidity had a negative effect on the photocatalytic oxidation of formaldehyde. According to the in Situ DRIFTS study conducted by Sun et al. (2010), formaldehyde molecules are adsorbed on the hydroxyl groups on the surface of TiO₂ photocatalyst via hydrogen bonding. Water molecules are competitors for the same adsorption sites, and thus raising relative humidity is unfavorable to the formaldehyde adsorption, which leads to the decrease of photocatalytic oxidation rate and formaldehyde. However, the water vapor also plays an important role in the photocatalytic reaction. In the dry TiO₂ surface, the superoxide radical anions (O₂⁻•), generated by adsorbed oxygen molecules reacting with electrons, is believed to be the major oxidizer in the photocatalytic oxidation of formaldehyde. However, the production rate of the final products CO₂ and H₂O is pretty low in this condition. The introduction of water causes an increase of the formation of hydroxyl radical OH•, an extremely powerful oxidant with higher redox potential than O₂⁻•, on the TiO₂ surface. Consequently, the production rates of both the intermediate formate and the final products CO₂ and H₂O increased considerably (Sun et al., 2010).

Effect of Formaldehyde Concentration

As demonstrated in Fig. 4, the formaldehyde photocatalytic oxidation rate increased with the increase of formaldehyde concentration. However, the variation of formaldehyde conversion was not very obvious in the formaldehyde concentration region investigated. In order to study the kinetics of the formaldehyde photocatalytic oxidation, we

Fig. 3. Effect of relative humidity on the formaldehyde conversion (temperature = 25 ± 2°C; UV intensity = 26 μW/cm²).
assumed that each channel of the honeycomb monolith reactor is a plug-flow reactor and the surface reaction rate of photocatalytic oxidation is associated with the surface coverage \( \theta \) (Langmuir isotherm), and then we have (Yu and Lee, 2007):

\[
r = k\theta = k \frac{KC}{1 + KC} = \frac{dF}{dAc} = \frac{Q \times dC}{W \times dL}
\]

(3)

where \( k \) (\( \mu \)-mole/m\(^2\)/s) is the rate constant of Langmuir-Hinselwood model; \( K \) (ppm \(^{-1}\)) is the Langmuir adsorption constant of formaldehyde; \( F \) (ppm m\(^2\)/s) is the flow rate of formaldehyde; \( Ac \) (m\(^2\)) is the TiO\(_2\) coating area; \( Q \) (m\(^3\)/s) is the gas flow rate; \( W \) (m) is the circuit of all the channels of honeycomb monolith; \( L \) (m) is the length of each channel; \( C \) (ppm) is the formaldehyde concentration in the reactor. Integrating both sides of Eq. (3), we get the following equation:

\[
\frac{A/Q}{(C_{in} - C_{out})} = \frac{1}{kK} \ln \left( \frac{C_{in}}{C_{out}} \right) + \frac{1}{k} \int_0^T \frac{dF}{dAc}
\]

(4)

According to Eq. (4), the plot of \( \ln(C_{in}/C_{out})/(C_{in} - C_{out}) \) vs. \( A/Q((C_{in} - C_{out}) \) are linear with a slope of \( 1/kK \) and an intercept of \( 1/k \) as shown in Fig. 5. The kinetic parameters \( k \) and \( K \) under RH 30\%, 50\% and 70\% are also listed in Fig. 5. The Langmuir adsorption constant of formaldehyde \( K \) reveals a negative correlation with relative humidity, implying that there is a competition between formaldehyde and water molecules for the adsorption site on the TiO\(_2\) surface. The rate constant of photocatalytic oxidation \( k \) is positively correlated with relative humidity, which may be relevant to the enhancement effect of relative humidity on the formation of hydroxyl radicals. Because the hydroxyl radicals act as a major oxidizer in the photocatalytic oxidation of organic chemicals, there may be a correlation between the rate constant of photocatalytic oxidation \( k \) and the rate constants for the gas-phase reaction with hydroxyl radicals \( k_{OH} \) (Atkinson, 1989; Gill and Hites, 2002), and Fig. 6 presents the positive correlation between \( k \) and \( k_{OH} \) (Obee and Hay, 1997; Yu et al., 2006b; Yang et al., 2007b; Huang, 2008; Lai, 2009).

**Effect of Gas Flow Rate**

In general, when the gas flow rate of the experimental system is slow, the flow pattern in the channel of the honeycomb monolith photocatalytic reactor is close to laminar flow, and the mass transfer of gaseous pollutants to the catalyst surface is restricted. In this condition, the rate determining step of the photocatalytic reaction is the gas-phase mass transfer. Increasing the gas flow rate can increase the mass transfer by increasing the turbulent diffusion and reducing the boundary layer in the gas-surface interface, and thus enhancing the photocatalytic reaction. However, when the gas-phase mass transfer rate is faster than the surface reaction rate of catalyst, the rate determining step of the photocatalytic reaction becomes the surface reaction. In this situation, increasing the gas flow rate has no significant effect on the photocatalytic reaction rate (Yu et al., 2006a, b; Yu and Lee, 2007; Yu et al., 2010).

The effect of gas-phase mass transfer can be proven by the kinetic parameters obtained from the experiments. When the photocatalytic reaction achieved a steady state, the gas-phase mass transfer rate equalled to the surface reaction rate. Therefore, we have:

\[
N_{\text{formaldehyde}} = k_f (C - C_{\text{interface}}) = k - \frac{KC}{1 + KC}
\]

(5)

where \( N_{\text{formaldehyde}} \) (ppm m/s) is the gas-phase mass transfer rate of formaldehyde from bulk phase to the interface between the gas phase and the surface of photocatalyst; \( k_f \) (m/s) is the mass transfer coefficient of formaldehyde; \( C \) (ppm) is the formaldehyde concentration in the bulk phase; \( C_{\text{interface}} \) (ppm) is the formaldehyde concentration in the interface. The comparison between the formaldehyde concentrations in the bulk phase and the interface can be used to determine the rate limit step of the photocatalytic reaction. Nevertheless, \( C_{\text{interface}} \) is hard to be measured directly. To estimate \( C_{\text{interface}} \), we rearrange the equation above, and then we have:

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**Fig. 4.** Effect of formaldehyde concentration on the formaldehyde conversion and photocatalytic oxidation rate (temperature = 25 ± 2°C; UV intensity = 26 μW/cm\(^2\)).
Fig. 5. The linear correlation between $A/Q(C_{in} - C_{out})$ and $\ln(C_{in}/C_{out})/(C_{in} - C_{out})$.

Fig. 6. The correlation between the rate constants for the gas-phase reactions with hydroxyl radical ($k_{OH}$) (Atkinson, 1989; Gill and Hites, 2002) and the rate constants of photocatalytic oxidation ($k$). (relative humidity = 50%; room temperature; UV intensity = 26–83 $\mu$W/cm$^2$).

$$C_{\text{interface}} = C \left[ 1 - \frac{kK}{k_g (1 + KC)} \right]$$  \hspace{1cm} (6)

In the photocatalytic reaction, gas-phase-mass-transfer limit happens as $C_{\text{interface}} \ll C$ and thus $kK >> k_g (1 + KC)$. Conversely, the surface-reaction limit occurs when $C_{\text{interface}} \approx C$ and hence $kK \ll k_g (1 + KC)$. The mass transfer coefficient of formaldehyde $k_g$ can be estimated by the following correlation (Uberoi and Pereira, 1996):

$$Sh = 2.696 \left[ 1 + 0.139 Sc \times Re \right]^{0.81} = \frac{k_g \times d}{L}$$  \hspace{1cm} (7)

in which $Sh$ is the Sherwood number; $d$ is the hydraulic diameter defined as 4 times the channel open area divided by the wetted perimeter (0.14 cm); $D$ is the diffusivity of formaldehyde (0.18 cm$^2$/sec); $L$ is the monolith length (5 cm); $Sc$ is the Schmidt number ($\equiv \mu/D$; $\mu$: viscosity of air $= 1.81 \times 10^{-5}$ Pa·sec; $D$: density of air $= 1.2$ kg/m$^3$); $Re$ is the Reynolds number ($\equiv v d / \mu$, $v$ = velocity in monolith channel; in this experimental setting, $Re = 0.3$–0.6, which represents a laminar flow pattern). The mass transfer coefficient of formaldehyde $k_g$ is around 0.0347 m/s and does not vary significantly in the air flow rate range 800–1600 mL/min as shown in Fig. 7. Therefore, $kK \ll k_g (1 + KC)$ and the rate determining step was the surface reaction.

Fig. 7 shows that when the gas flow rate gradually
increased from 800 mL/min to 1,600 mL/min, the gas retention time of the photocatalytic reactor would reduce from 181 to 91 seconds and thus the formaldehyde conversion gradually decreased from 95% to 92% and then exhibited a stable state. Meanwhile, the photocatalytic oxidation rate increased from $0.80 \times 10^{-3}$ to $1.54 \times 10^{-3}$ μ-mole/m²/s, indicating that the air flow rate significantly affected the “apparent photocatalytic oxidation rate” [Eq. (2)]. The “apparent photocatalytic oxidation rate” calculated by Eq. (2) is the average reaction rate of the reactor. However, the surface reaction rate of the photocatalytic oxidation should be $4.41 \times 10^{-3}$ μ-mole/m²/s [Eq. (3)], and almost all the formaldehyde was oxidized near the inlet of the reactor when the air flow rate was low. The formaldehyde can penetrate more deeply under higher air flow rate, and the “apparent photocatalytic oxidation rate” would be higher. The breakthrough of formaldehyde may occur when the “apparent photocatalytic oxidation rate” equals to the surface reaction rate, and it happens when the air flow rate is higher than 4200 mL/min, according to the correlation between the “apparent photocatalytic oxidation rate” and the air flow rate.

**Limitation**

- Lack of the data of mineralization rate:
  
  In fact, we did monitor the time variations of CO and CO₂ in the inlet and outlet streams by using an Indoor air quality monitor (Q-Trak 7565-X, TSI Inc.) as we did in our previous studies (Yu et al., 2006b; Yu and Lee, 2007; Yu et al., 2010; Huang, 2008; Yu et al., 2014). However, the sensitivity of the instrument is not good enough to detect the very small variations of CO and CO₂. Even the formaldehyde was oxidized completely; it generated only 2 ppm CO or 2 ppm CO₂ (maximum), which is almost equal to or below the detection limit of the Q-Trak. In addition, it is impossible to monitor the very small variation of H₂O (< 2 ppm) under such a high background humidity (20% relative humidity = 6070 ppm H₂O) in our experimental system. Due to the limitation of our experimental setting, we cited other research conclusions to support our discussion.

**CONCLUSIONS**

In this study, we systematically investigated the removal of formaldehyde by the fiber optic illuminated honeycomb monolith photocatalytic reactor and the influential factors including formaldehyde concentration (0.8–2.0 ppm), relative humidity (30–70%), and air flow rate (800–1600 mL/min). The experimental results of various formaldehyde concentrations indicate that the kinetic fits the Langmuir-Hinshelwood model. Increasing relative humidity has a negative effect on the formaldehyde conversion and Langmuir adsorption constant of formaldehyde, which is relevant to the competition between formaldehyde and water molecules for the adsorption sites on the surface of TiO₂ photocatalyst. On the other hand, the rate constant of photocatalytic oxidation $k$ increased with the increase of relative humidity, which may be caused by the enhancement effect of relative humidity on the formation of hydroxyl radicals.

In our experimental setting, because the air flow rate was low ($\leq 1600$ mL/min), the gas retention time ($\geq 7.7$ sec) in the reactor was long enough for the reactor to achieve a high formaldehyde conversion ($\geq 92$%), and most of the formaldehyde was oxidized near the inlet of the reactor. However, the formaldehyde conversion may decrease gradually with the increase of air flow rate and the breakthrough of formaldehyde may occur when air flow rate $> 4200$ mL/min (face velocity $> 1.7$ cm/sec; gas retention time $< 3$ sec). Consequently, a gas retention time longer than 3 seconds was required for this photocatalytic reactor to attain an acceptable performance on the removal of indoor formaldehyde.
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REFERENCES


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