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Formaldehyde Removal from Air by a Biodegradation System

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Abstract A biodegradation system was used for the treatment of formaldehyde-polluted air. Air pressure dropped 12 mm water in the trickling biofilter during the experiment of about 4 months. In the range 20–300 mg m^{-3} influent formaldehyde, this biodegradation system obtained 4.0–40.0 mg h^{-1} degradation capacity, with 100%–66.7% degradation efficiency. The amount of formaldehyde degraded by the trickling biofilter was more than that by the activated sludge bioreactor below 200 mg m^{-3} influent gaseous formaldehyde while the amount by the trickling biofilter was less than that by the activated sludge bioreactor over 200 mg m^{-3} influent gaseous formaldehyde.

Keywords Formaldehyde · Trickling biofilter · Activated sludge bioreactor

Formaldehyde (HCHO), a ubiquitous air pollutant, is often released from industrial products or processes, the enzymatic oxidation of methylated compounds, and the photooxidation of atmospheric hydrocarbons (Barber and Donohue 1998). It can be also produced by secondary reactions of hydrocarbons with O_3 (Kondo et al. 1995). Formaldehyde can cause eye irritation, respiratory tract irritation, dizziness, fatigue and neurotoxicity (Shinohara et al. 2004). Moreover, formaldehyde has been classified as a genotoxicity, mutagen and suspected carcinogen (Ballarin et al. 1992; Naya and Nakanishi 2005). To date, physical adsorption with porous materials, chemical reaction with potassium permanganate and/or organic amines, and catalytic oxidation of HCHO into CO_2 and H_2O with supported noble metals were found to be effective for eliminating atmospheric formaldehyde (Tang et al. 2006). However, the physical adsorption and chemical reaction cannot completely transfer HCHO into harmless compounds. The catalytic oxidation is not widely used due to the high cost of noble metals.

Formaldehyde is a key intermediate in the metabolism of several C_1 compounds in methylotrophic microorganisms (Marx et al. 2003). Formaldehyde is a highly reactive compound that has a toxic effect on all organisms through its nonspecific reactivity with proteins and nucleic acids (Ferdman 1973). To avoid this problem, methylotrophs detoxify formaldehyde by dissimilatory and assimilatory pathways (Vorholt 2002). Some methylotrophs exhibit high levels of ability to degrade formaldehyde. For instance, Vorholt et al. (2000) reported that the removal capacity of cytoplasmic formaldehyde in the methylotrophs was up to 0.1 mol $L^{-1}min^{-1}$. Recently, formaldehyde degradation by microorganisms has attracted the scientific attention on how to develop a technique to utilize the biodegradation (Oliveira et al. 2004; Eiroa et al. 2005). The purpose of this work was to incorporate a trickling biofilter (TF) into an activated sludge bioreactor (AS) to remove low concentration of gaseous formaldehyde.

Materials and Methods

The bioreactor, schematically given in Fig. 1, consisted of three major components: a TF, an AS and a gaseous formaldehyde generation system.

One of the two cubical open-top chambers with 30 cm in size, made of polymethyl methacrylate, was used as the

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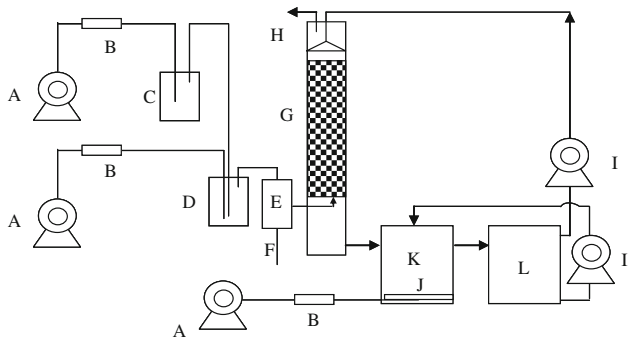

Fig. 1 Scheme of the bioreactor. A- air pump, B- flowmeter, C- formaldehyde solution (37%) vessel, D- mixing vessel, E- buffer vessel, F- gas sampling port, G- trickling biofilter, H- gas outlet, I- peristaltic pump, J- aerator, K- activated sludge bioreactor, L- precipitation pool

AS, the other as precipitation pool. A cylindrical polymethyl methacrylate chamber with 100 cm long and 12 cm in diameter was used as the TF. Plenty of ceramic rings with 5-mm in diameter were used as biomass immobilization support in the TF, resulting in about 60% bed porosity. The 50 cm long packed bed was supported at the bottom and covered on the top by a fine perforated polyethylene sieve plate to get a homogeneous distribution of the airflow and liquid drainage through the biofilter packing. All tubes for gas flow were made of Teflon.

The TF was operated counter-currently. The air was supplied to the TF by two air pumps. The air stream with high-concentration formaldehyde was obtained by bubbling a low-flow-rate air stream through a formaldehyde solution vessel. Then the air containing formaldehyde vapor was adequately mixed with a high-flow-rate air stream. The influent polluted air stream with the desired concentration of formaldehyde was obtained by regulating both the low-flow-rate and the high-flow-rate air stream with two flowmeters. In this study, 20, 50, 80, 110, 150, 200, 250, 300 mg m^{-3} influent formaldehyde-polluted air were investigated at about 200 L h^{-1} flowrate.

The activated sludge, collected from Beijing Beixiaohe sewage plant, was firstly incubated feeding 10 mg L^{-1} formaldehyde in the nutrients solution for 50 days. Then the incubated activated sludge was regulated to 4,200 mg L^{-1} (MLSS) used to inoculate the TF and the AS. The nutrient solution for the activated sludge incubation and the experiment run was composed of KH_2PO_4 (800 mg L^{-1}), NH_4Cl (600 mg L^{-1}), $MgSO_4$ (36.6 mg L^{-1}), $ZnSO_4$ (1.4 mg L^{-1}), $MnSO_4$ (1.787 mg L^{-1}), $CaCl_2$ (10 mg L^{-1}) $FeSO_4$ (1.367 mg L^{-1}).

The aqueous phase was continuously recirculated at 15 L h^{-1} to the top of the bioreactor by a peristaltic pump, and sprinkled on the surface of the biofilter packing bed. The trickling aqueous phase was collected in the AS, where pH was controlled at about 7.0 by addition of a 1-mol L^{-1} NaOH solution. A 0.5-L fresh nutrient solution was continuously applied into the AS every day to make up the nutrient and water consumption. The sludge in the AS was aerated by an air pump at the rate of 2 L min^{-1}. After aeration the suspension was transferred into the precipitation pool for separating sludge. The precipitate was transferred into the AS and the supernatant was used to recirculate.

The inlet and outlet gas stream from the TF were collected using a gas collection system and analyzed by the acetylacetone spectrophotometric method (Chinese National Standard 1995). The detection limit of 0.5 mg m^{-3} and the fortified recovery of 95.3%–104.2% were obtained in this method. A 30-ml sludge sample was collected from the inlet and outlet of the AS, respectively. The supernatant was collected for formaldehyde determination by the acetylacetone spectrophotometric method after the suspension centrifugation (2,000 rpm). Formaldehyde volatilized from the AS was determined by formaldehyde analyzer (4160, Interscan Co. USA) after closing the top for 2 min. A manometer was used to measure the air pressure drop of the TF.

Firstly, the TF has run at 20 mg m^{-3} rate of influent formaldehyde for 45 days to acclimatize the microorganism to the new process conditions. Then the system has run for 10 days at each projected influent formaldehyde concentration, respectively. All data were recorded once everyday.

Results and Discussion

Formaldehyde degradation capacity of the TF was very low during the early days after the microorganism inoculation (Fig. 2). The degradation capacity gradually increased with

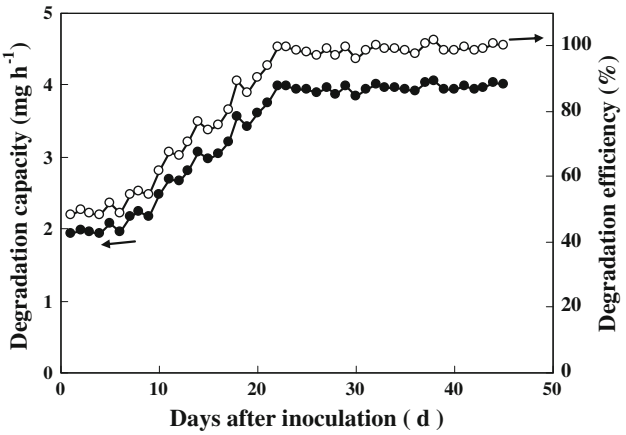

Fig. 2 Change in formaldehyde degradation in the trickling biofilter with incubation days. *Solid circles* and *open circles* represent degradation capacity and efficiency, respectively

time and tended to stabilize about 3 weeks after the inoculation. Figure 2 shows that the degradation efficiency took the same tendency as the capacity. This can be explained as the microbial biomass in the packed bed kept increasing during the period.

During the experiment of about 4 months, air pressure dropped 12 mm water in the TF. Prado et al. (2004) also found a negligible pressure drop in the TF throughout their experiment, which was attributed to a high trickling flowrate (9.0 L h^{-1}). In this study, high flowrate of trickling water (15 L h^{-1}) and high bed porosity (60%, before inoculation) may jointly contribute to the low pressure drop. Thus, no backwashing was performed during the experiment.

Figure 3 shows the changes in formaldehyde degradation in the bioreactor with influent gaseous formaldehyde concentration. Elevating influent formaldehyde concentration from 20 to 150 mg m^{-3} significantly increased degradation capacity from 4.0 to 29.8 mg h^{-1}, corresponding to more than 99% degradation efficiency. Further enhancing influent concentration led to a slight increase in degradation capacity and a sharp decrease in degradation efficiency. With influent formaldehyde concentration elevating from 200 to 300 mg m^{-3}, the degradation capacity increased from 37.3 to 40.0 mg h^{-1}, corresponding to a decrease in the degradation efficiency from 93.3% to 66.7%. As can be concluded, at this superficial air velocity, degradation capacity reached a maximum value of about 40 mg h^{-1}.

In order to evaluate the contributions of the TF and the AS to formaldehyde degradation, data on the degradation by the TF and the AS are given in Fig. 4. In this experiment, no formaldehyde was detected in effluent water from the AS, indicating that all formaldehyde form influent water was completely degraded by the microorganisms in

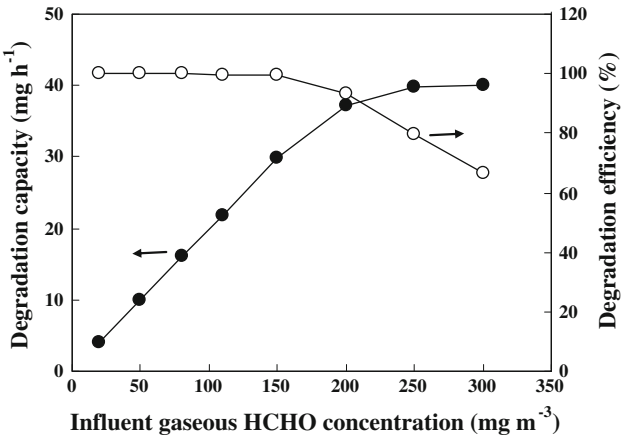

Fig. 3 Changes in formaldehyde degradation in the bioreactor with influent gaseous formaldehyde concentration. *Solid circles* and *open circles* represent degradation capacity and efficiency, respectively

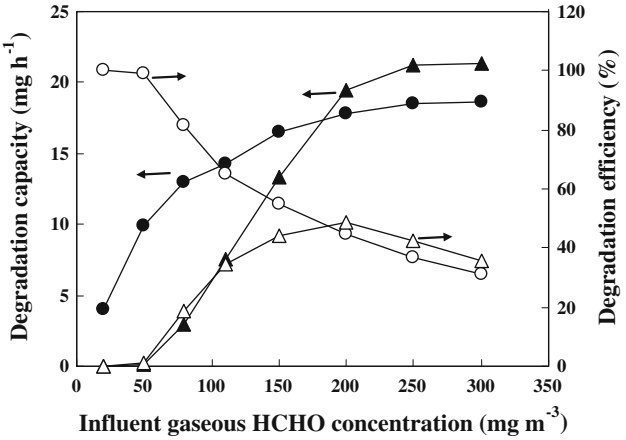

Fig. 4 Changes in formaldehyde degradation in the trickling biofilter (TF) and activated sludge bioreactor (AS) with influent gaseous formaldehyde concentration. *Solid circles* and *open circles* represent TF degradation capacity and efficiency, respectively. *Solid triangles* and *open triangles* represent AS degradation capacity and efficiency, respectively

the AS. Thus, the degradation efficiency of the AS is defined as formaldehyde degraded by the AS accounting for the loaded influent gaseous formaldehyde. A much steeper gradient of degradation capacity along influent formaldehyde concentration appeared in the range 20–50 mg m^{-3} compared with more than 50 mg m^{-3} for the TF. Correspondingly, the degradation efficiency of the TF attained 100% in the range 20–50 mg m^{-3} and sharply dropped down with further elevating influent concentration. This result indicates that all formaldehyde from waste gas was degraded by the TF in 20–50 mg m^{-3} influent concentrations. Higher influent concentration resulted in excess formaldehyde out of the TF. One part of the excess formaldehyde was carried by waste gas out of the TF as it could not be absorbed by trickling water. The other part of the excess formaldehyde was carried into the AS and sequentially removed, creating an increase in degradation capacity, especially significant in the range 50–200 mg m^{-3} influent concentrations. Over 200 mg m^{-3} influent concentration, the degradation capacities of the AS was higher than that of the TF. The degradation efficiency of the AS reached a maximum at 200 mg m^{-3} influent concentration. These results demonstrate that a TF coupled with an AS could significantly intensify formaldehyde degradation over 50 mg m^{-3} influent concentration compared with a single TF.

Figure 4 shows that a single TF can entirely remove low concentration formaldehyde (less than 50 mg m^{-3}) from waste gas. This treated formaldehyde concentration is higher than that reported by Fei et al. (2005), who found a trickling biofilter could remove 100% formaldehyde from less than 15 mg m^{-3} waste gas. Low degradation efficiency at high concentration of formaldehyde in Fei et al.

(2005) may be attributed to no nutrient addition into the trickling water. The degradation capacity of the TF for formaldehyde in this study is similar to that in Prado et al. (2004). Eiroa et al. (2005) reported that an activated sludge bioreactor could remove 99.5% formaldehyde from wastewater even in the range 26–3,168 mg L^{-1}. Our previous study also showed that the activated sludge in a sequencing batch reactor degraded more than 98% formaldehyde from wastewater below 60 mg L^{-1} (Xu et al. 2008). In this study, influent soluble formaldehyde into the AS was less than 2 mg L^{-1} and no formaldehyde was observed from effluent water out of the AS. This indicates that some formaldehyde escaped from the biodegradation system is due to the excess formaldehyde out of absorption by the trickling water. Thus, further elevating the volume of trickling water may increase the degradation efficiency for high concentration of formaldehyde (Fei et al. 2005).

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