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RAPID COMMUNICATION

Optimum empty bed retention time for toluene degradation in a biofilter packed with ceramic beads

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Abstract−Toluene is a toxic air pollutant and a common constituent emitted from industrial processes including chemical manufacturing operations, painting, and petroleum refining. We previously developed a new biofilter system the degradation of acetone and MEK. In this study, we evaluate the optimum EBRT to achieve an effective elimination capacity (EC) with smaller filter bed volume. In addition, we optimized the operation conditions for toluene degradation using the same biofilter system.

Key words: Biofilter, Toluene, Empty Bed Retention Time

INTRODUCTION

Most volatile organic compounds (VOCs) emitted to the atmosphere are believed to be harmful to human health as they often result in nausea, headaches, and irritation affecting the nervous and breathing systems and can lead to cancer. They also contribute to substantial damage of fauna and flora [1]. The basic concept of biofiltration is to remove contaminants by passing them through microbial layers formed in a packed bed. Any porous material capable of adsorbing gaseous compounds and supporting biological growth can possibly be used as a packing material [2]. Improvements of the biofiltration process commonly used for the removal of odorous compounds have led to better control of key parameters including temperature, microorganisms, pH, pressure drop, moisture content, filter bed, flow rate, pollutants, nutrients, and reactor models, enabling the application of biofiltration to be extended to the removal of VOCs [3]. Among these factors, the EBRT is critical for the microbial degradation performance as it is directly related to the reaction time allowed for the microbial degradation of pollutants. Previous studies of characteristic time scales of the various physical, chemical, and biological processes occurring in a biofilter have found that the diffusion rates of pollutants are slower than the microbial reaction rates. Accordingly, to improve biofilter performance, the EBRT should be greater than the time required for diffusion of the pollutants [1]. To give rise to better removal efficiency (RE) of VOC, employing a longer EBRT is required [4]. On the other hand, a longer EBRT for better RE requires larger filter bed volumes. Thus, determination of an optimum EBRT as a function of the filter bed volumes is necessary for a better performance of a biofilter system.

Toluene is a toxic air pollutant and a common constituent emitted from industrial processes including chemical manufacturing operations, painting, and petroleum refining. We previously developed a new biofilter system for the degradation of acetone and MEK [5]. While our previous work was focused on optimization of operating conditions in a newly developed biofilter system, herein we evaluated the optimum EBRT to achieve an effective elimination capacity (EC) with smaller filter bed volume. In addition, we optimized the operation conditions for toluene degradation using the same biofilter system.

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EXPERIMENTAL SECTION

1. Microorganisms and Medium

The microorganisms including activated wastewater sludge obtained from a sewage treatment plant were inoculated on the surface of the packing material. The filter bed used in this study was constructed of an inorganic porous ceramic type material (Enbion Co., Korea) to minimize the long term pressure drop. The pressure drop was measured to be 0.4 cmH_2O/1 m by a water manometer. $(NH_4)_2SO_4$ (3 g/L), $Na_2HPO_4 \cdot 12H_2O$ (9.0 g/L), $CaCl_2 \cdot 12H_2O$ (0.01 g/L), and $MgSO_4$ (0.5 g/L) were periodically provided at the top of the biofilter reactor by a nozzle during the operation period. The HPO_4^{2-} and $H_2PO_4^-$ ions act to buffer the solution to maintain the pH in the range of 6.0-8.5. The nutrient solution was inserted at a rate of 1 L/2 weeks (0.06 g N/d).

2. Experimental Conditions of the Biofilter

The overall scheme of the semi-pilot scale biofilter reactor and experimental conditions were the same as previously reported [5]. The bioreactor consisted of a tower with a height of 1.0 m and an internal diameter of 10 cm divided into three equal stages. Each column was filled with packing material, resulting in a total effective bed volume of 3.4 L. The bottom of the reactor was an arch-shape that made for easy draining of the liquid. The top of the biofilter reactor had a nozzle for supplying the nutrient solutions with a metering pump (Iwaki EHC-R220C, Japan). Air was introduced into the biofilter using a gas compressor (MCD6062Z, Fuji Electric Co. Ltd., Japan) and was controlled by a mass flow rate controller (MFC FC 770C, KNH Co., Korea). Additionally, the temperature and humidity of the air were maintained for an optimum growth of microorganism using by a humidifier with a temperature controller. Toluene was injected as a liquid with a syringe pump (KD Scientific Model

Table 1. Operating conditions of the biofilter

Biofilter reactor	Internal diameter (cm)	10
	Height of each stage (cm)	30
	No. of stages	3
	Volume of the bed (L)	5.0
Environmental conditions	Temperature (°C)	20-27
	Relative humidity (%)	70-95
	pH of packing material	6.0-8.0
	Pressure drop (cm H_2O/m)	<0.4
Nutrients supply	Spraying interval	1/2 weeks
	Quantity sprayed	1 L
Operating conditions	EBRT (s)	15 - 60
	Upflow velocity ($cm^3cm^{-2}s^{-1}$)	1.1-4.2

1000, Boston, MA, USA) at a minimum of 0.01 mL/h. At start-up, the microorganisms were acclimated to the toluene environment as the concentration was gradually increased from 0 to 20 ppmv over 30 days. The inlet loading concentration of the supplied toluene was 32 to 64 ppmv for the entire experiment, as shown in Fig. 2. From day 67 to 103, the air flow rate was changed and monitored to determine the effect of the EBRT on the toluene RE and EC. The air flow rate was controlled from 5 to 20 L/min by a mass flow controller corresponding to an EBRT of 15-60 seconds. The operating and environmental conditions are summarized in Table 1.

3. Analytical Methods

The gas was collected from one sample port on each layer by a mini-vacuum pump (MPΣ300, S IBATA, Japan) through a sampling tube (Gerst-el TDS2/TDSA, Mülheim an der Ruhr, Germany) to control the sampling gas volume. A typical sample volume of 1 L was taken at a sampling rate ranging from 0.5-1.0 L/min. To determine the concentration of toluene, a multisorbent adsorption/thermal desorption GC/MS system was used (model Agilent 6890 GC/5975N MSD, equipped with a mass selective detector using helium as the carrier gas). Also, the filter bed was collected from the other sample port to investigate the condition of the microorganisms. The morphology of the biofilm on the packing material was observed by an environmental scanning electron microscope (ESEM, XL30 FEI, Oregon, USA) as shown in Fig. 1. The biofilter reactor was operated for about three months and the inlet/outlet concentrations of toluene were measured every two or three days.

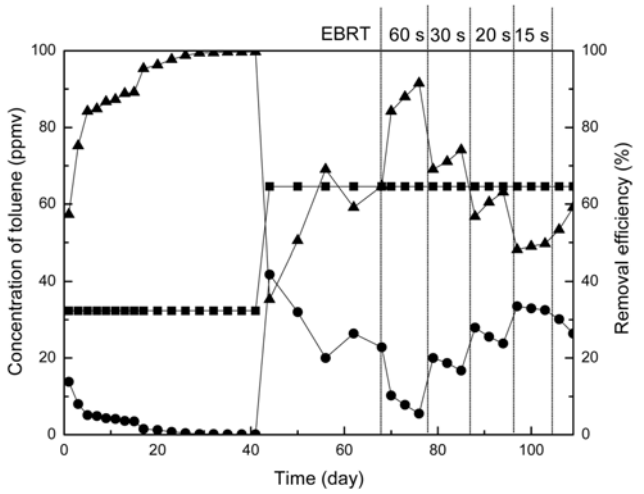

Fig. 2. Removal performance of toluene under various operating conditions (■: inlet concentration of toluene, ●: outlet concentration of toluene, ▲: RE).

The operation of the biofilter system was quantified by the following equations [6].

$$C_{ppm} = 22.4 \times 10^6 \left(\frac{\rho R_1}{MR}\right)\left(\frac{273}{T}\right)\left(\frac{P}{760}\right) \quad (1)$$

$$C_{(mg/m^3)} = \left(\frac{\rho R_1}{MR}\right)\left(\frac{273}{T}\right)\left(\frac{P}{760}\right) \times 10^6 \quad (2)$$

$$IL = \frac{Q}{V_v} C_i \quad (3)$$

$$X = \left(\frac{C_i - C_o}{C_i}\right) \times 100 \quad (4)$$

$$EC = X \times IL \quad (5)$$

RESULTS AND DISCUSSION

1. Evaluation of EC with Various EBRTs

As shown in Fig. 2, in the time period from day 40 to 55, the toluene RE changed from 95% to 35% as the toluene concentration increased by a factor of two. This result indicates that the metabolic activities of the microorganisms are very sensitive to dramatic

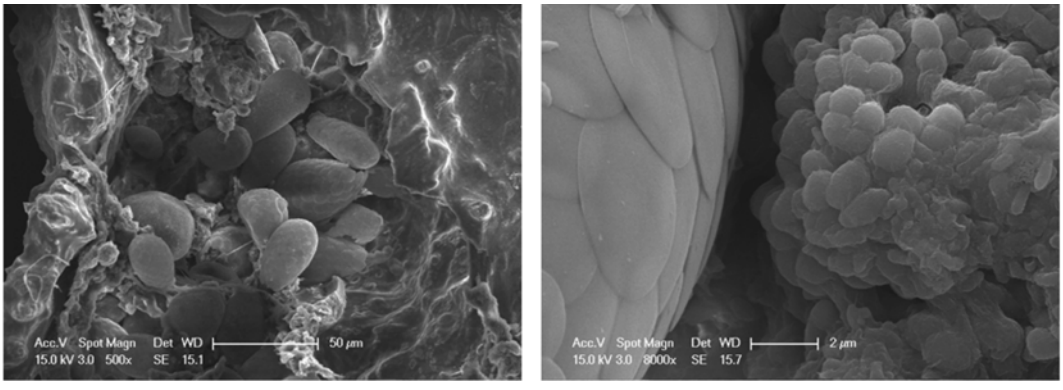

Fig. 1. SEM images of biofilm on the media surface in biofilter for the removal of toluene: 500× (left), 8,000× (right).

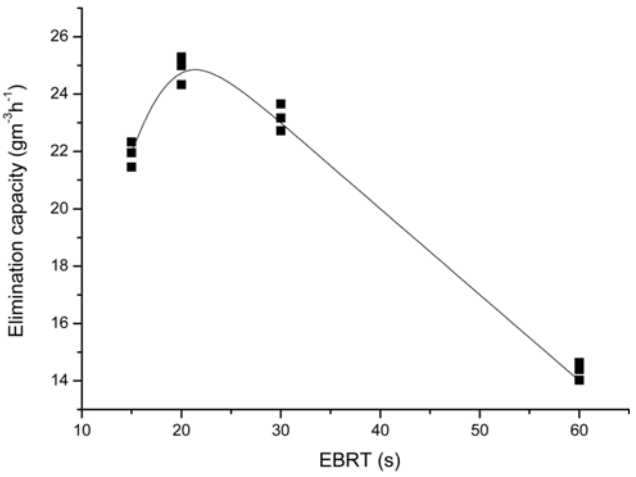

Fig. 3. Variation of EC under various EBRTs in biofilter performance.

changes of inlet loading concentration, indicating that the microorganisms have a weakness to dramatic changes of atmospheric conditions. In the time period from day 67 to 103, in order to determine the effect of the EBRT on the toluene RE and EC, each EBRT experiment lasted for nine days. All data were collected at pseudo-steady state after two days acclimation. This is because biomass accumulation at the high EBRT values and high loadings could lead to change of biofilter performance [7]. To evaluate the RE and maximum EC at various EBRTs, the air flow rate was changed to 5, 10, 15, and 20 L/min. At the same time, the inlet loading rates were changed to 3.7 $gm^{-3}h^{-1}$-62.5 $gm^{-3}h^{-1}$ in order to maintain the same inlet concentration of toluene of 64 ppmv. The effect of the EBRT on the RE and EC was evaluated, as shown in Fig. 3, at the same inlet concentration. The toluene RE improved as the EBRT increased because the longer EBRT values correspond to long contact times between the toluene gas and filter bed materials, which increases the biodegradation time. The highest elimination capacity was obtained at an EBRT of 20 s where the EC was 25.2 $gm^{-3}h^{-1}$. The EC value of our results was similar to the results obtained using other types of biofilters reported by Suidan et al. as 22.6 to 26.7 $gm^{-3}h^{-1}$ of toluene [8]. Our research indicated when EBRT was less than 20 s, the diffusion rate was lower than the reaction rate, while when EBRT was over 20 s, the diffusion rate was higher than the reaction rate. Accordingly, the highest value of EC was obtained when EBRT was optimized at 20 s.

Higher inlet loadings could lead to higher biofilm thickness, but thicker biofilm could retard the diffusion rates of exterior to interior of the biofilm. Biofilm-specific electron transport system (ETS) activity is directly related to degradation efficiency [9]. A previous study reported that substrate inlet loading rates were not directly related to biofilm-specific electron transport system activity in a pseudo steady state (after 5 days) [9]. Under the condition of a specific concentration of toluene (64 ppmv), we conclude that the optimum EBRT is 20 s for our biofilter system, when an optimum RE of toluene with a minimum filter bed volume is achieved.

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NOMENCLATURE

ρ : density [g/ml]
R_l : flow rate of liquid toluene [ml/h]
M : molecular weight [g/mol]
R : flow rate of supplied air [L/h]
IL : inlet loading rate [$gm^{-3}h^{-1}$]
Q : gas flow rate [m^3/h]
V_V : void volume [m^3]
X : conversion rate [%]
C_i : toluene inlet concentration [g/m^3]
C_o : toluene outlet concentration [g/m^3]
EC : elimination capacity [$gm^{-3}h^{-1}$]

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