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*Full Length Research Paper*

# The influence of nonionic surfactant Brij 30 on biodegradation of toluene in a biofilter

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**The influence of nonionic surfactant Brij 30 on toluene dissolved in the water phase and biodegradation kinetic behaviors of toluene in a polyvinyl alcohol (PVA)/peat/$KNO_3$/ (granular activated carbon) GAC composite bead biofilter were investigated. The toluene dissolved in the water phase was enhanced by the addition of surfactant into the aqueous solution and the maximum amount of toluene dissolved in the water phase occurred at the surfactant concentration of 34.92 $mgl^{-1}$. Zero-order kinetics with diffusion limitation was regarded as the most adequate biochemical reaction kinetic model. The microbial growth rate and biochemical reaction rate were inhibited at higher surfactant content and toluene inlet concentration. The degree of inhibitive effect was more pronounced at lower toluene inlet concentration. The maximum elimination capacity decreased with increasing surfactant content. The addition of nonionic surfactant Brij 30 into filter material was unfavorable for toluene degraded by the microbial.**

**Key words:** Nonionic surfactant Brij30, toluene, dissolution capacity, biodegradation, composite bead biofilter.

## INTRODUCTION

The removal of volatile organic compounds (VOCs) from polluted air stream using biological process is highly efficient and has low installation and operation/ maintenance costs. Biofiltration technology offers environmental advantages: it does not generate undesirable by-products by converting many organic and inorganic compounds into harmless oxidation products (e.g., water and carbon dioxide). Biofiltration involves the passage of a polluted air stream through a packed bed containing microorganisms immobilized within a biofilm attached to the bed-packing material. Contaminants are transferred to the interface between the gas and the biofilm and are subsequently absorbed into the biofilm. Contaminants are then used as carbon and/or energy sources for the microorganisms within the biofilm. The solid filter material provides a nutrient source and matrix for the attachment of microorganisms in the biofiltration process. Therefore, the filter material property is an important factor in obtaining an optimal pollutant removal. The optimal filter material should have the following characteristics: High moisture holding capacity, porosity, available nutrients, compression strength and pH buffer capacity (Deviney et al., 1999). A spherical polyvinyl alcohol (PVA)/peat/$KNO_3$/ granular activated carbon (GAC) composite bead was prepared and proved to be suitable as a filter material in the biofiltration process in our previous works (Chan and Lin, 2006; Chan and Peng, 2008).

Toluene is a widely used industrial chemical. It is a hydrophobic compound and one of the 189 hazardous air pollutants listed in the 1990 Clean Air Act Amendment (CAAA90) proposed by the US Environmental Protection Agency (EPA). Even at low concentrations, toluene has been found to be carcinogenic, causes damage to the liver and kidney and paralyze the central nervous system (Martin et al., 1998; Murata et al., 1999). Large volumes of toluene are released into the atmosphere during manufacturing process every year, endangering the air quality and pubic health. The use of surfactants has the potential to increase the biodegradation rate of hydrophobic organic compounds in contaminated environments by increasing

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**Abbreviations: GAC,** Granular activated carbon; **VOCs,** volatile organic compounds; **PVA,** polyvinyl alcohol; **PCP,** pentachlorophenol; **SDS,** sodium dodecyl sulfate; **CMC,** critical micelle concentration; **EBRT,** empty bed residence time; **GC,** gas chromatography.

the total aqueous solubility of these compounds (Grimberg et al., 1995; Guha and Jaffe, 1996; Meeren and Verstraete, 1996; Tiehm, 1994; Volkering et al., 1995). Nonionic surfactants are usually used in the bioavailability studies due to their relatively low toxicity compared to ionic surfactants (Yeh et al., 1998). Brij 30 was the most biodegradable surfactant among Brij 30, Tween 80 and Triton X-100 three nonionic surfactants, and the solubility of naphthalene and phenanthrene in the water phase was linearly proportional to the concentration of nonionic surfactant (Kim et al., 2001). The biodegradation rate of pentachlorophenol (PCP) was enhanced for the concentration of PCP at 140 and 220 $mgl^{-1}$ as the concentration of nonionic surfactant Tergitol NP-10 increased from 0 to 1500 $mgl^{-1}$, but it was inhibited for the concentration of PCP at 100 and 50 $mgl^{-1}$ (Cort et al., 2002). Sodium dodecyl sulfate (SDS) anionic surfactant was found to inhibit the growth of the fungus and polyoxyethylene sorbition monolaurate (Tween 20) nonionic surfactant was found to enhance inoculums development by shortening the lag period and toluene degradation in fungal vapor-phase bioreactor (Woertz and Kinney, 2004). The removal efficiency of gaseous trichloroethylene and tetrachloroethylene was enhanced as nonionic surfactant Alfonic$^{R}$ 810-60 was introduced into the activated carbon biofilter (Kim et al., 1999).

The filter material contained surfactants that would enhance the performance of biofilter treating hydrophobic contaminants. But a prepared filter material containing surfactant in the biofiltration process is unavailable in the relevant literature. We have seen that the process for degradation of VOCs in a PVA/peat/$KNO_3$/GAC composite bead biofilter could be divided into lag, exponential growth and stationary three phases, with the exponential growth and stationary phases being important for controlling the removal efficiency of the biofilter (Chan and Lin, 2006; Chan and Peng, 2008). Therefore, studies on the kinetics of exponential growth and stationary phases are very important for the operation and design of the biofiltration process. However, details of the kinetic of such biodegradation process in biofilter are scanty. This article investigates the preparation of a PVA/peat/$KNO_3$/GAC composite bead containing nonionic surfactant Brij 30 and biodegradation kinetic behaviors of hydrophobic toluene compound in this composite bead biofilter. The effect of inlet concentration and surfactant content in this biofilter on microbial growth rate and biochemical reaction rate are also studied.

## MATERIALS AND METHODS

### Materials

Peat (industrial grade from KekkilaOyj, Tuusula, Finland) was dried at 105°C before use. It has a dry density of 90 $kgm^{-3}$, a pH of 5.5, a pore volume of 96% and an organic substance content of 91%. Boric acid, sodium monobasic phosphate, sodium dibasic phosphate, potassium nitrate and toluene (extra pure grade from Union Chemical, Hsinchu, Taiwan) were used as received. PVA powder (industrial grade from Chung Chun Petrochemical, Hsinchu, Taiwan), GAC (industrial grade from Taipei Chemical, Hsinchu, Taiwan) and Brij 30 (industrial grade from Imperial Chemical Industries Limited, London, England) were also used as received. The Brij 30 has a molecular formula of $C_{12}H_{25}(OCH_2CH_2)_4OH$, a hydrophilic-lipophilic balance (HLB) of 9.7 and a critical micelle concentration (CMC) in the range of 7 to 14.52 $mgl^{-1}$ (Yeh et al., 1998; Kim et al., 2001; Laha and Luthy, 1992).

### Dissolution experiments

The experiment of toluene dissolved in the water phase was carried out by adding 3.0 g toluene into a desired surfactant concentration aqueous solution (150 ml) in a glass-stoppered Erlenmeyer 250 ml flask. The desired surfactant concentration ranged from 0 to 79.43 mg surfactant $L^{-1}$. The flasks were in a shaking isothermal water bath for about 24 h at 30°C. Then, the solution was centrifuged for 1 h at 2000 rpm to separate into water and organic phases. The amount of toluene dissolved in the water phase was measured by gas chromatography (GC) (Model GC-8A from Shimadzu, Tokyo, Japan) equipped with a FID detector and SPB$^{TM}$-5 capillary column (30 m x 0.53 mm x 1.5 μm filter thickness). The temperature of injection, oven and detector were 200, 120 and 220°C, respectively.

### Biofilter experiments

The procedures for preparing PVA/peat/$KNO_3$/GAC composite beads and the apparatus and operation of the biofilter system were described in our previous works (Chan and Peng, 2008). Before packing, the prepared PVA/peat/$KNO_3$/GAC composite bead was immersed in a 0.384 M $KNO_3$ and a desired surfactant concentration aqueous solution to adsorb $KNO_3$ and surfactant, and to reach equilibrium (approximately 24 h). The bead moisture content was humidified to more than 1.5 g water $g^{-1}$ dry composite bead and the seeding was performed with activated sludge obtained from the wastewater treatment plant in Hsinchu Science-Based Industry Park. As the stationary phase had been maintained for more than 3 days, the biofilter operating was stopped according to the variations of toluene removal efficiency with operation time. Then, new filter material was repacked and the operation procedures described above were carried out to start another experiment with the desired inlet concentration and surfactant content. The gas flow rate was maintained at 0.03 $m^3 h^{-1}$ for all experiments and consequently, the empty bed residence time (EBRT) of biofilter column was 96.5 s. The toluene concentration in the inlet and exit air streams of each section was determined by auto-sampling and analyzed using gas chromatography (GC) (Model GC-8A from Shimadzu, Tokyo, Japan). There were no other products detected except $CO_2$ and water in the exit air stream. The toluene removal efficiency was calculated by the difference of the toluene concentration between the inlet and exit air streams. The relative standard deviation and relative error of the experimental measurements were less than 2 and 5%, respectively.

## RESULTS AND DISCUSSION

### Dissolution capacity

The variations of $S/S_0$ value with surfactant concentration are shown in Figure 1. S and $S_0$ were the amount of toluene dissolved in the water phase of the aqueous solution with and without surfactant, respectively. It was found that all the $S/S_0$ values were greater than 1.0 in the

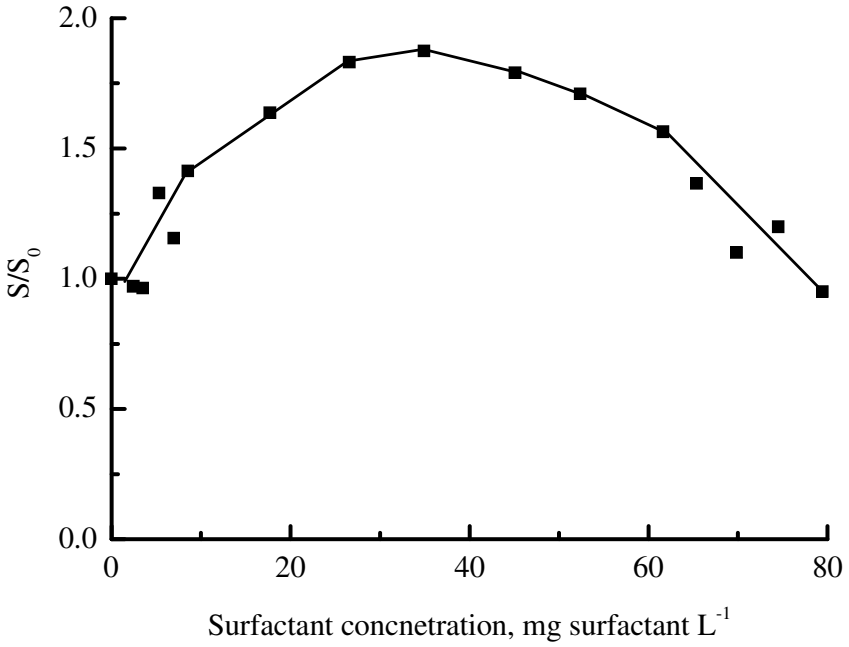


**Figure 1.** The variations of $S/S_0$ value with surfactant concentration.

surfactant concentration range of 0 to 79.43 $mgl^{-1}$. The result indicated that the addition of surfactant into aqueous solution could enhance the toluene dissolved in the water phase in this concentration range. The $S/S_0$ value increased from 1.0 to 1.87 as the surfactant concentration increased from 0 to 34.92 $mgl^{-1}$, and then, it decreased from 1.87 to 1.0 as the surfactant concentration increased from 34.92 to 79.43 $mgl^{-1}$. The result indicated that the enhancing effect increased with increasing the surfactant concentration in the former concentration range and it decreased with increasing the surfactant concentration in the latter concentration range. The optimal surfactant concentration occurred at 34.92 $mgl^{-1}$.

The CMC of Brij 30 was in the range of 7 to 14.52 $mgl^{-1}$ and the maximum amount of toluene dissolved in the water phase was at the surfactant concentration of 34.92 $mgl^{-1}$. In order to study whether the biodegradation of toluene was enhanced in the surfactant concentration less than CMC or not, we chose three surfactant concentrations as the desired surfactant concentration of aqueous solution for the composite bead immersed in and adsorbed by the surfactant for the later experiments. Three desired surfactant concentrations were 7.31 (which was in the range of CMC and lies in the $S/S_0$ value increased region), 45 and 75.67 $ml^{-1}$ (which were greater than CMC and the optimal surfactant concentration, and lie in the $S/S_0$ value decreased region). Therefore, the calculated surfactant content in the composite bead was 0, 0.021, 0.135 and 0.225 mg $g^{-1}$ composite bead for the surfactant concentration 0, 7.31, 45 and 75.67 $mgl^{-1}$, respectively.

## Microbial growth process

The variations of toluene removal efficiency with operation time at various inlet concentration and surfactant content are shown in Figure 2 (only the inlet concentration of 200 ppm and the surfactant content of 0.021 mg surfactant $g^{-1}$ composite bead are shown). It was found that the variations of toluene removal efficiency with operation time appeared in three phases: Lag phase (phase I), exponential growth phase (phase II) and stationary phase (phase III) (Buchanan, 1980; Chan and Lin, 2006; Chan and Peng, 2008; Madigan et al., 2008). Only the biochemical kinetic behaviors in the exponential growth phase and stationary phase was studied in this work. The elimination capacity of toluene was also determined.

In the exponential growth phase (phase II), the microbial growth rate increased exponentially and was represented by the following equation (Chan and Peng, 2008; Valsaraj, 1995):

$$\ln (C / C_0) = -k_g t \quad (1)$$

Where, C and $C_0$ are the concentration of VOCs in the exit and inlet air stream, respectively. A plot of $\ln (C / C_0)$ versus t should correspond to a straight line and $k_g$ can be determined. The microbial growth rate $k_g$ at various inlet and surfactant concentrations was calculated from the data in phase II and Equation 1.

The variations of the $k_g$ values with surfactant content for four inlet concentrations are shown in Figure 3. The $k_g$ value decreased with increasing surfactant content in the surfactant content range of 0 to 0.225 mg $g^{-1}$ composite

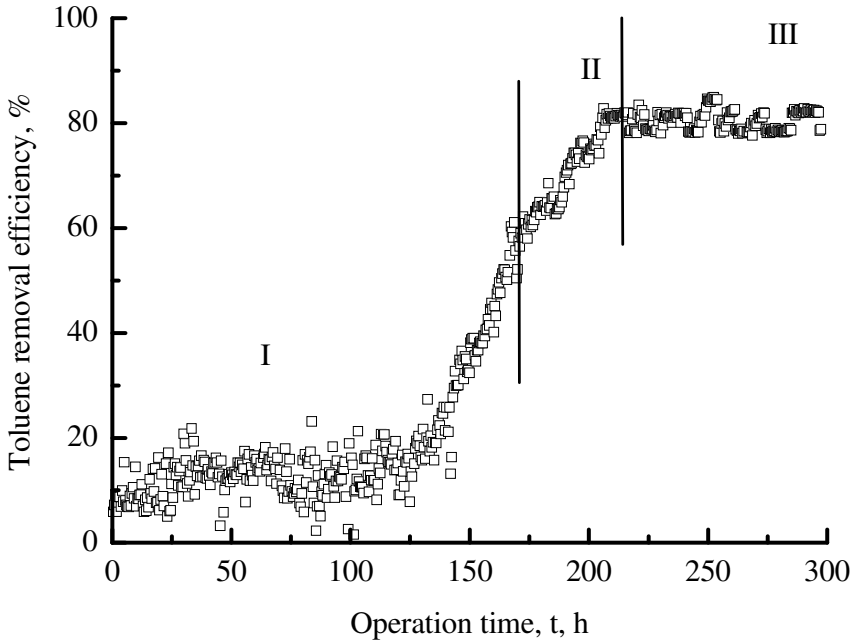


**Figure 2.** The variations of toluene removal efficiency with operation time at inlet concentration of 200 ppm and surfactant content of 0.021 mg $g^{-1}$ composite bead.

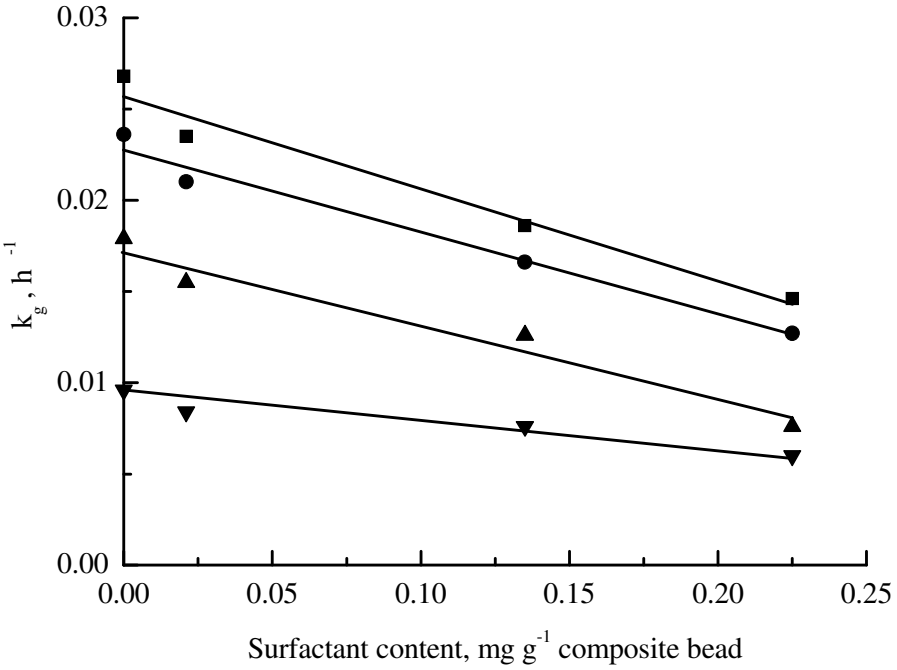


**Figure 3.** The variations of the $k_g$ values with surfactant content for four inlet concentrations: (■) 50 ppm, (●) 100 ppm, (▲) 200 ppm and (▼) 300 ppm.

bead. The result indicated that the microbial growth rate was inhibited at higher surfactant content and the addition of surfactant into composite bead was unfavorable for microbial growth. The sequestration of a hydrophobic compound within surfactant micelles might decrease its biodegradation rate by decreasing its concentration in the water phase (Roch and Alexander, 1995). In addition, surfactants might produce toxicity and decrease the activity

of the microbial (Volkering et al., 1995). Therefore, the greater amounts of toluene dissolved into micelles in the biofilm or the greater toxicity in microbial at higher surfactant content can lead to greater inhibition of the biodegradation of toluene. The slope of the linear profiles in this surfactant content were 50, 100, 200 and 300 ppm, inlet concentration was $1.50 \times 10^{-4}$, $1.36 \times 10^{-4}$, $1.03 \times 10^{-4}$ and $4.14 \times 10^{-5}$ g composite bead $h^{-1}$ $mg^{-1}$, respectively. The result indicated that the degree of inhibitive effect was more pronounced at lower inlet concentration.

The $k_g$ value decreased with increasing inlet concentration in the inlet concentration range of 50 to 300 ppm for four surfactant contents. An increase in the inlet concentration generally would enhance the transfer rate of VOCs from the gas phase to the biofilm (enhancing effect). This phenomenon could explain the fact that more microorganisms were caused to participate in the biodegradation. However, high concentrations of some recalcitrant VOCs may produce inhibitive effects on the metabolic activity of the microbial population (Leson and Winer, 1991). Therefore, the result indicated that the inhibitive effect predominated and the microbial growth rate was inhibited at higher inlet concentration. The result corresponded to the result reported that higher concentration of toluene and xylene caused inhibition of microbial growth (Leon et al., 1999; Shim et al., 2005).

**Biochemical reaction process**

In the stationary phase, the population of viable cells was at a relatively constant value. The earliest and commonly used biofiltration model under steady state condition was proposed by Ottengraf. Three basic situations of Ottengraf model was first-order kinetics, zero-order kinetics with reaction limitation and zero-order kinetics with diffusion limitation (Ottengraf, 1986). The corresponding equations expressed the rates of biochemical reaction for each situation as follows:

First-order kinetic:

$$\ln (C / C_0) = -k_1 \theta \quad (2)$$

Zero-order kinetic with reaction limitation:

$$C_0 - C = k_0 \theta \quad (3)$$

Zero-order kinetic with diffusion limitation:

$$1 - (C / C_0)^{1/2} = k_d \theta \quad (4)$$

Where, $k_1$, $k_0$ and $k_d$ are the rate coefficient of first-order kinetic, zero-order kinetic with reaction limitation and zero-order kinetic with diffusion limitation, respectively (Yang and Allen, 1994).

The substrate utilization rate by the microbial was generally expressed by the Michaeilis-Menten relationship. Under the steady state of microbial population, three possible situations may be encountered in a biochemical reaction system (Yang and Allen, 1994): Situation (1) if the substrate concentration was very low ($K_s >> C_0$), the reaction rate expression could be simplified to first-order kinetic; situation (2) if the substrate concentration was very high ($K_s << C_0$), the reaction rate expression could be simplified to zero-order kinetic; situation (3) if the substrate concentration $C_0$ was comparable with $K_s$, the reaction rate expression can not be simplified and the Ottengraf diffusion limiting model is found to be the most approximate expression.

In order to verify the biochemical reaction kinetic model, assume that there was a plug air flow in the biofilter column and the following equation was derived from the Michaelis-Menten equation (Valsaraj, 1995):

$$(C_0 - C) / \ln (C_0 / C) = V_m (\theta / \ln(C_0 / C)) - K_s \quad (5)$$

Where, θ is EBRT, $K_s$ is half-saturation constant and $V_m$ is maximum reaction rate. A plot of $(C_0 - C)/ \ln (C_0 / C)$ versus $\theta / \ln (C_0 / C)$ should correspond to a straight line, and $K_s$ and $V_m$ can be determined. The plot of $(C_0 - C) / \ln (C_0 / C)$ versus $\theta / \ln (C_0 / C)$ for four surfactant contents is shown in Figure 4 (only the surfactant content of 0.021 mg $g^{-1}$ composite bead is shown). The calculated $K_s$ for the surfactant content of 0, 0.021, 0.135 and 0.225 mg $g^{-1}$ composite bead were 38.33, 37.27, 15.27 and 32.43 ppm, respectively. The calculated $V_m$ for the surfactant content of 0, 0.021, 0.135 and 0.225 mg $g^{-1}$ composite bead were 2.04, 1.97, 1.31 and 1.90 ppm $s^{-1}$, respectively. The ratio of $C_0 / K_s$ for the surfactant content of 0, 0.021, 0.135 and 0.225 mg $g^{-1}$ composite bead were 1.31 - 7.83, 1.34 - 8.05, 3.27 - 19.65 and 1.54 - 9.25, respectively. The results indicated that the relationship of $C_0$ and $K_s$ does not correspond to situation 1 or situation 2, but to situation 3. Therefore, zero-order kinetic with diffusion limitation was regarded as the most adequate biochemical reaction kinetic model because the concentration $C_0$ was comparable with $K_s$ for four surfactant contents in this study. Thus, the $k_d$ value at various inlet concentrations and surfactant content was calculated from the data in phase III and Equation 4

The variations of the $k_d$ values with surfactant content for four inlet concentrations are shown in Figure 5. The $k_d$ value decreased with increasing surfactant content in the surfactant content range of 0 to 0.225 mg $g^{-1}$ composite bead. The result indicated that the biochemical reaction rate was also inhibited at higher surfactant content and the addition of surfactant into composite bead was unfavorable for VOCs degraded by the microbial. The result corresponded to the report that the higher the concentration of nonionic surfactant Brij30 or Brij 35, the slower the biodegradation rate of phenanthrene in the aqueous phase (Yuan et al., 2000). The slope of the linear profiles in the surfactant content range of 0 to 0.225 mg $g^{-1}$ composite bead for 50, 100, 200 and 300 ppm inlet concentration was $7.04 \times 10^{-5}$, $6.38 \times 10^{-5}$, $3.61 \times 10^{-5}$ and $1.69 \times 10^{-5}$ g composite bead $s^{-1}$ $mg^{-1}$, respectively.

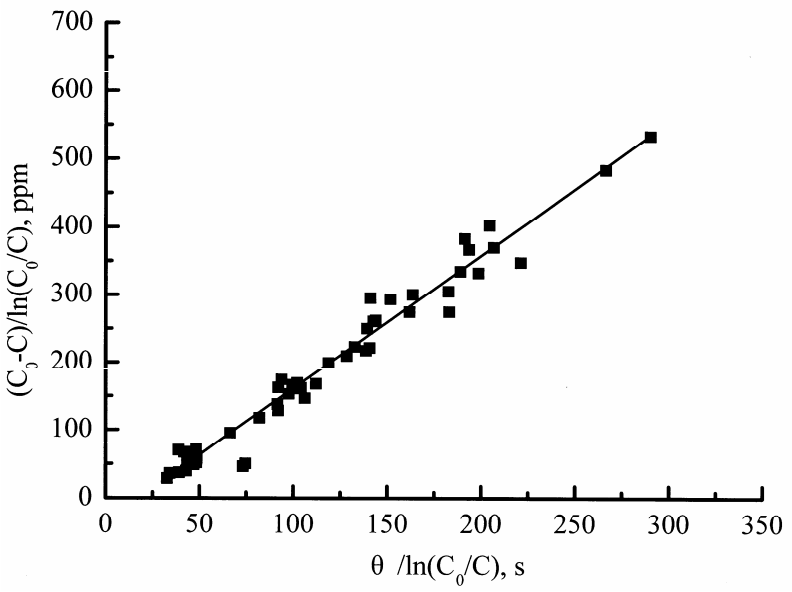


**Figure 4.** Plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ at surfactant content of 0.021 mg $g^{-1}$ composite bead.

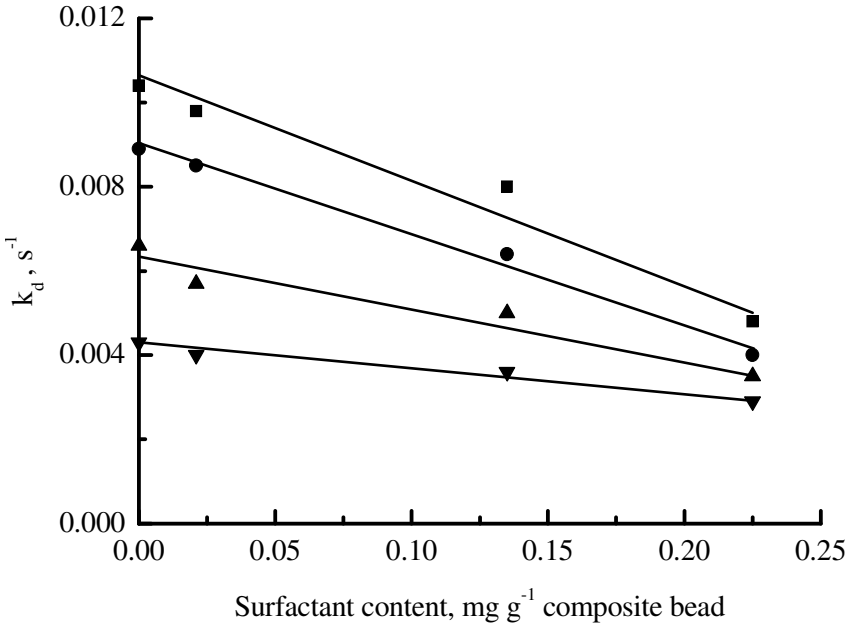


**Figure 5.** The variations of the $k_d$ values with surfactant content for four inlet concentrations: (■) 50 ppm, (●) 100 ppm, (▲) 200 ppm and (▼) 300 ppm.

The result indicated that the degree of inhibitive effect was more pronounced at lower inlet concentration.

The $k_d$ value decreased with increasing inlet concentration in the inlet concentration range of 50 to 300 ppm for four surfactant contents. The result indicated that the biochemical reaction rate was also inhibited at higher inlet concentration. The result is similar to the report that the pseudo first-order biodegradation rate constant decreased as the toluene inlet concentration increased in an acclimated mixed-culture biofilter (Park et al., 2004).

## Elimination capacity

Elimination capacity (EC) and load were calculated according to the equation presented below (Deviney et

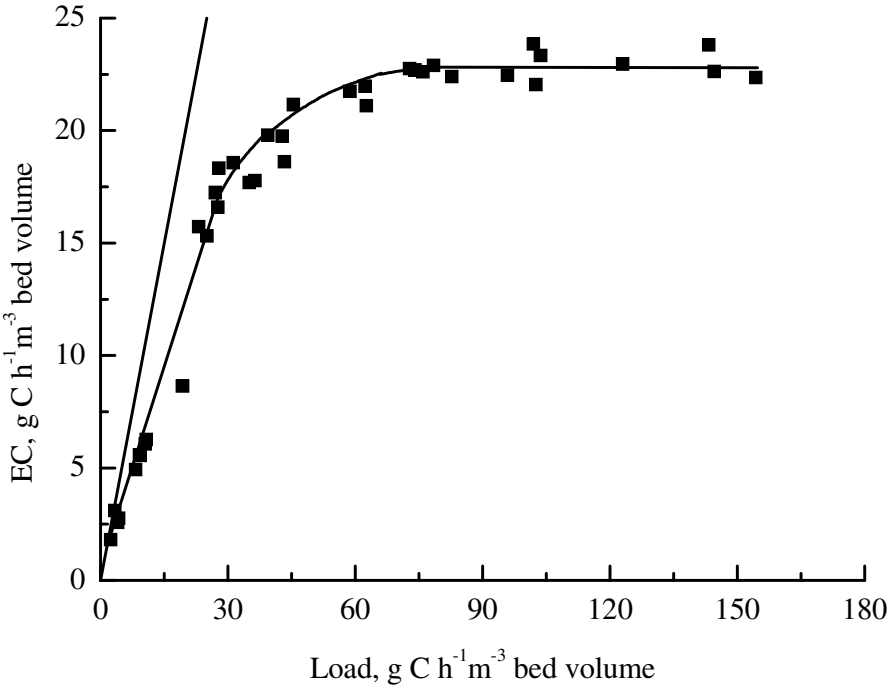


**Figure 6.** The variations of elimination capacity (EC) with load at the surfactant content of 0.021 mg $g^{-1}$ composite bead: (■) 100% removal.

al., 1999):

$$EC = Q (C_0 - C) / V \quad (6)$$

$$Load = Q C_0 / V \quad (7)$$

Where, Q is the flow rate of inlet air steam and V is the bed volume of filter material as packed. Under low load conditions, the elimination capacity essentially equals the load, and the system is calculated to have 100% removal efficiency. By increasing the load on a system, a point will be reached where the overall load will exceed the overall elimination capacity, generating removal efficiencies less than 100%. This point is typically called the critical elimination capacity. As the load continues to increase, a maximum overall elimination capacity will eventually be reached. This maximum overall elimination capacity is independent of the contaminant concentration and residence time within a reasonable range of operating conditions (Deviney et al., 1999). The relationship of elimination capacity of biofilter versus load for four surfactant content is shown in Figure 6 (only the surfactant content of 0.021 mg $g^{-1}$ composite bead is shown). It was found that elimination capacity increased and tended towards a constant value with increasing inlet load. The result was closely related to the result reported in previous works (Chan and Peng, 2008). The maximum elimination capacity for the surfactant content of 0, 0.021, 0.135 and 0.225 mg $g^{-1}$ composite bead was 24.37, 22.98, 21.59 and 16.71 g C $h^{-1}$ $m^{-3}$ bed volume, respectively. The result indicated that the maximum elimination capacity decreased with increasing surfactant content in the composite bead. Therefore, the addition of surfactant into composite bead caused the inhibition of microbial growth and biochemical reaction, as discussed in the earlier section of this paper.

## Conclusions

This work was to test the dissolution capacity of toluene in the water phase and the biodegradation of toluene in a composite bead biofilter as the nonionic surfactant was added. The toluene dissolved in the water phase was enhanced by the addition of surfactant into the aqueous solution. The addition of surfactant into the composite bead caused the inhibition of microbial growth and biochemical reaction. The microbial growth rate and the biochemical reaction rate were inhibited at higher surfactant content and toluene inlet concentration. The degree of inhibitive effect was more pronounced at lower toluene inlet concentration. The maximum elimination capacity decreased with increasing surfactant content and it was in the range of 16.71 - 24.37 g C $h^{-1}$ $m^{-3}$ bed volume.

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