

## Degradation of Benzene and Toluene by a Fluidized Bed Bioreactor Including Microbial Consortium

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**Abstract**—A fluidized bed bioreactor including microbial consortium was used to remove benzene and toluene simultaneously. The microbial consortium was obtained from sewage treatment plant, and showed maximum benzene degradation rate of 45.2 mg/l·h·mg cell in 30 °C and pH 7.0, and maximum toluene degradation rate of 44.4 mg/l·h·mg cell in 30 °C and pH 8.0. The optimum operating condition of the fluidized bed bioreactor was 30 °C, pH 7.0 and 150 cm of liquid bed height. The average removal efficiency of benzene was 94% for inlet concentration of 53(±5) ppm benzene and that of toluene was 96% for an inlet concentration 48(±5) ppm toluene at 600 l/h of gas volumetric flow rate. The maximum removal capacity in the experimental condition was 22.3 g/m<sup>3</sup>·h for benzene and 16.3 g/m<sup>3</sup>·h for toluene.

Key words: Benzene, Toluene, Fluidized Bed Bioreactor, Microbial Consortium, Removal Efficiency

### INTRODUCTION

Benzene and toluene, the typical VOC's, have been produced in large amounts at petrochemical plants, leather processing plants, etc. [You et al., 2001]. Benzene and toluene have long been known as hazardous to human health [US EPA, 1977]. The facilities to remove the air pollution chemicals in plants are absolutely needed to maintain good air quality. The technology to remove VOC's has been focused on the biological treatment since it has reduced the operating costs and the secondary contamination [Devinny et al., 1999]. The microorganisms to degrade benzene and toluene aerobically include *Pseudomonas putida*, *Rhodococcus rhodochrous*, and *Mycobacterium vaccae* [Deeb and Alvarez-Cohen, 1999; Lee et al., 1993], and the activated sludge with various VOC degrading microbes has been widely used in recent researches [Webster et al., 1996].

A biofilter has typically been used in the biological treatment of VOC; however, clogging of overgrown cells and limitation of oxygen transport to cells can be serious problem when loading rates of VOC are increased [Devinny et al., 1999]. In a fluidized bed bioreactor, the inlet gas dissolved in solution of the bioreactor fluidized carriers and solution vigorously, then reduced cell growth on the carrier and increased oxygen supply [Oh et al., 1998]. As a result, the fluidized bed bioreactor was suitable for the treatment of high loading rate of VOC.

In this research, benzene and toluene were removed by fluidized bed bioreactor including VOC degrading microbial consortium. The optimum operating condition of the bioreactor was studied for the temperature, pH and liquid bed height. The removal efficiency was

measured in inlet concentration of 53(±5) ppm benzene and 48(±5) ppm toluene at 600 l/h of gas flow rate, and the maximum removal capacity was also obtained.

### EXPERIMENTAL

#### 1. Microorganisms

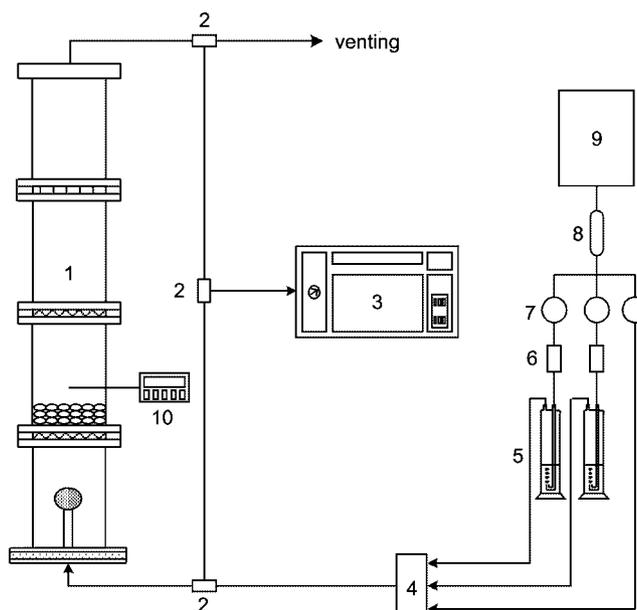


Fig. 1. The schematic diagram of a fluidized bed bioreactor.

- |                       |                         |
|-----------------------|-------------------------|
| 1. Reactor            | 6. Check valve          |
| 2. Sampling valve     | 7. Mass flow controller |
| 3. Gas chromatography | 8. Moisture trap        |
| 4. Mixing chamber     | 9. Air compressor       |
| 5. Vaporizer          | 10. Thermocouple        |

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<sup>‡</sup>This paper is dedicated to Professor Dong Sup Doh on the occasion of his retirement from Korea University.

Aerobic microbial consortium to degrade benzene and toluene was obtained from Yongho sewage water treatment plant in Busan and cultured at 30 °C, pH 7.0 for 3 months in the following medium [Moon et al., 2001] (g/l): 0.5 C<sub>6</sub>H<sub>6</sub>, 0.5 C<sub>7</sub>H<sub>8</sub>, 0.5 MgSO<sub>4</sub>·7H<sub>2</sub>O, 0.5 K<sub>2</sub>HPO<sub>4</sub>, 0.5 (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, 0.01 CaCl<sub>2</sub>, 0.001 FeCl<sub>3</sub>·6H<sub>2</sub>O, 0.001 MnCl<sub>2</sub>, and 0.0001 ZnSO<sub>4</sub>. From the sequencing of 16S rDNA by MicroID (Taejon, Korea), the major microorganism of the microbial consortium was identified as *Rhodococcus ruber*.

## 2. Fluidized Bed Bioreactor

A three phase fluidized bed bioreactor, as shown in Fig. 1, was used to remove benzene and toluene. The benzene and toluene was vaporized in a cylinder-type vaporization equipment where the glass bead was packed to 1/3 of the height to have a uniform distribution of the concentration after liquid benzene and toluene was added. The concentration was adjusted by air flow rate by using a Mass flow controller (5850E, Brooks Co.) and the gas entered the fluidized bed bioreactor (inner diameter=10 cm, height=210 cm). Gas disperser was installed to reduce the bubble size at the bottom of the bioreactor and gas distributor which had 500 1 mm inside diameter hole was installed over the gas disperser to distribute the inlet gas evenly. The inlet gas fluidized the carriers, then the bacteria in both carriers and solution oxidized the benzene and toluene. The gas volumetric flow rate was 600 l/h, the inlet concentration was 53(±5) ppm of benzene and 48(±5) ppm of toluene, and the liquid volume in the column was 11.78 l when the liquid bed height was 150 cm. The total cell concentration in solution of the bioreactor was 168 mg dry cell/l.

As a cell carrier, 580 g of biosands (Crystal biosand, Chung woo art system), which were made of 15% SiO<sub>2</sub> and 85% H<sub>2</sub>O, had the diameter of 2.0-3.0 mm, specific surface area of 539 m<sup>2</sup>/g, and density of 1,270 kg/m<sup>3</sup> was used in the experiment. Among the various cell carriers, the biosand showed excellent odor removal capacity [Oh et al., 1998].

The biosand was pretreated with UV and was inoculated with the microbial consortium in 200 ml medium. The solution including the biosand was maintained at 30 °C, 160 rpm in the shaker for 48 hours, and then added in the bioreactor, and initial total cell concentration was 212 mg dry cell/l.

The concentration of benzene and toluene was measured with gas chromatograph (HP 4890D, Hewlett Packard) using FID and HP-5MS column. Initial injection temperature was 120 °C, and increased to 150 °C at the rate of 5/min, and the detector temperature was set as 280 °C. As a carrier gas, 1.1 ml/min of N<sub>2</sub> was used.

In this study, the removal efficiency, inlet loading rate, removal capacity and biodegradation rate were calculated according to the following formulae;

$$\text{Removal efficiency } (\eta) = 100 \cdot (C_{in} - C_{out}) / C_{in} \quad (1)$$

$$\text{Inlet loading rate} = C_{in} Q / V \quad (2)$$

$$\text{Removal capacity} = \eta C_{in} Q / 100 \cdot V \quad (3)$$

$$\text{Biodegradation rate} = (C_0 - C_t) / (t \cdot \text{dry cell weight}) \quad (4)$$

## RESULTS AND DISCUSSION

Fig. 2 shows the effect of temperature on benzene and toluene degradation by microbial consortium in flask culture at 225 mg/L

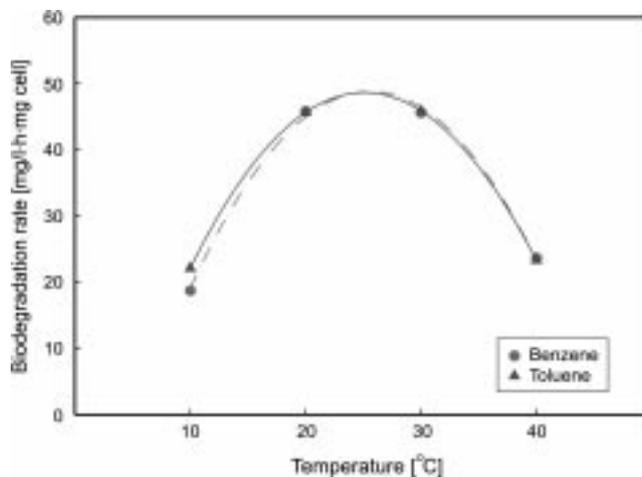


Fig. 2. The effect of temperature on biodegradation rate of benzene and toluene by microbial consortium in flask culture at 225 mg/l benzene, 220 mg/l toluene and pH 7.0.

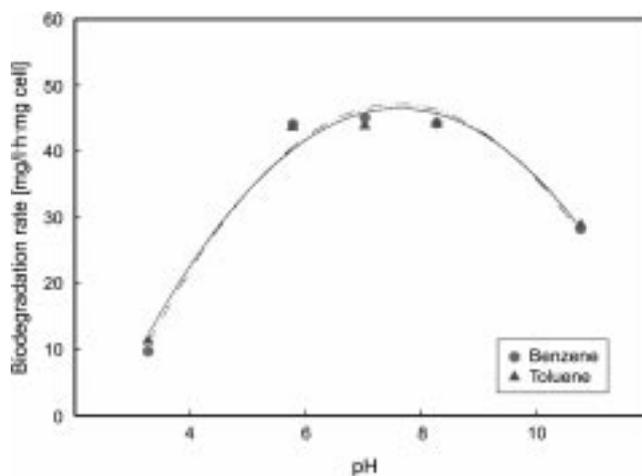


Fig. 3. The effect of pH on biodegradation rate of benzene and toluene by microbial consortium in flask culture at 225 mg/l benzene, 220 mg/l toluene and 30 °C.

of benzene, 220 mg/L of toluene, and pH 7.0. The biodegradation rates were maximum at 20-30 °C, and those of benzene and toluene showed similar values for all the temperature ranges tested. Fig. 3 shows the effect of pH on benzene and toluene biodegradation in flask culture at 225 mg/l benzene, 220 mg/l toluene and 30 °C. The biodegradation rate of benzene was highest at pH 7.0 as 45.2 mg/l-h-mg cell and that of toluene was highest at pH 8.0 as 44.4 mg/l-h-mg cell. The neutral pH region of pH 6-8 showed higher degradation rates of benzene and toluene than acidic and basic region.

Fig. 4 shows the degradation rates of benzene and toluene for the initial concentrations in flask culture at 30 °C and pH 7.0. At lower concentration, the degradation rates increased monotonically as the concentration of benzene and toluene increased. However, degradation rates reached steady state over 300-400 ppm, and the degradation rate of toluene was higher than that of benzene.

Fig. 5 shows the effect of bed temperature on the removal efficiency of benzene and toluene by a fluidized bed bioreactor. The optimum operating temperature was 30 °C and the removal effi-

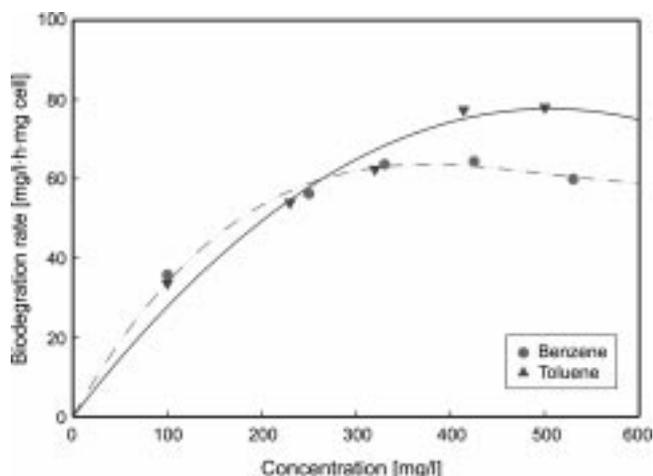


Fig. 4. The biodegradation rate of benzene and toluene for the initial concentrations by microbial consortium in flask culture at 30 °C and pH 7.0.

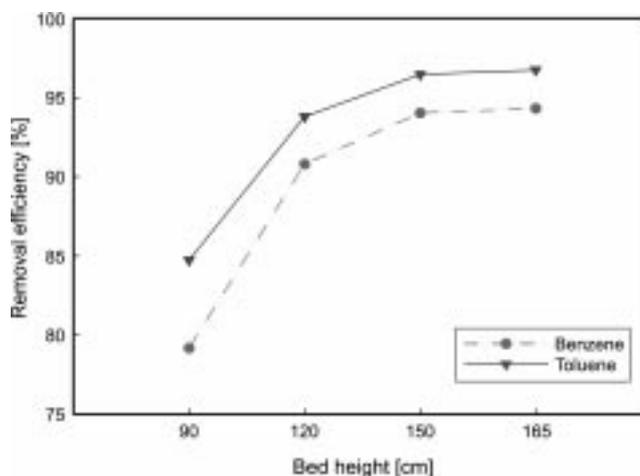


Fig. 7. The effect of liquid bed height on the removal efficiency of benzene and toluene by a fluidized bed bioreactor at 30 °C and pH 7.0.

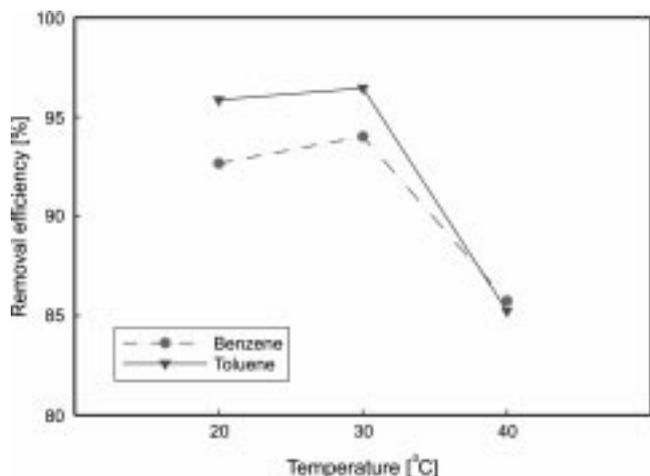


Fig. 5. The effect of bed temperature on the removal efficiency of benzene and toluene by a fluidized bed bioreactor at pH 7.0 and 150 cm liquid bed height.

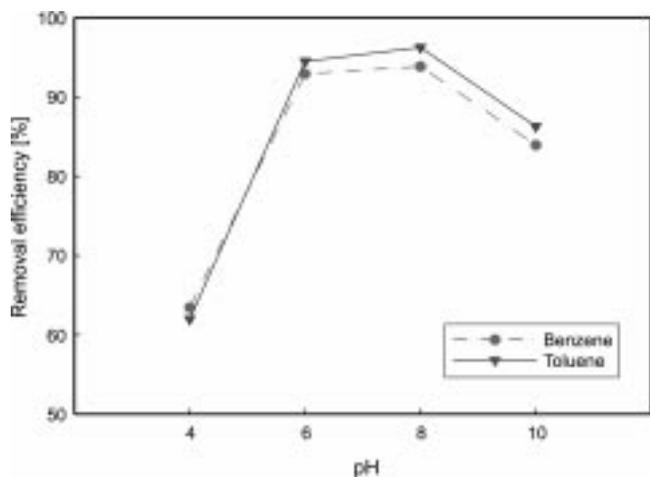


Fig. 6. The effect of solution pH on the removal efficiency of benzene and toluene by a fluidized bed bioreactor at 30 °C and 150 cm liquid bed height.

ciency of toluene was higher than that of benzene at the optimum temperature. From the data, the bioreactor needs temperature control in the heat of summer if it is installed in the plant. Fig. 6 shows the effect of solution pH on the removal efficiency of benzene and toluene by a fluidized bed bioreactor. The removal efficiency of benzene and toluene was higher at neutral pH region of pH 6-8 and that was significantly reduced in acidic region.

Fig. 7 shows the effect of liquid bed height on the removal efficiency of benzene and toluene by a fluidized bed bioreactor. As the liquid bed height increased from 90 cm to 165 cm, the removal efficiency was increased significantly due to the increased residence time of gas (42.4 s to 77.7 s) in the solution. However, the increase of the removal efficiency was minimal between 150 cm and 165 cm of liquid bed height, and the optimum liquid bed height was thought to be 150 cm.

Fig. 8 shows the effect of input gas velocity on the removal efficiency of benzene and toluene. The removal efficiency of benzene

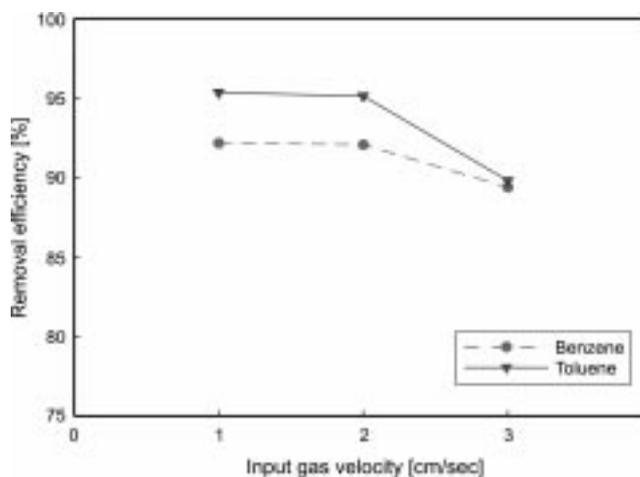


Fig. 8. The effect of input gas velocity on the removal efficiency of benzene and toluene by a fluidized bed bioreactor at 30 °C and pH 7.0.

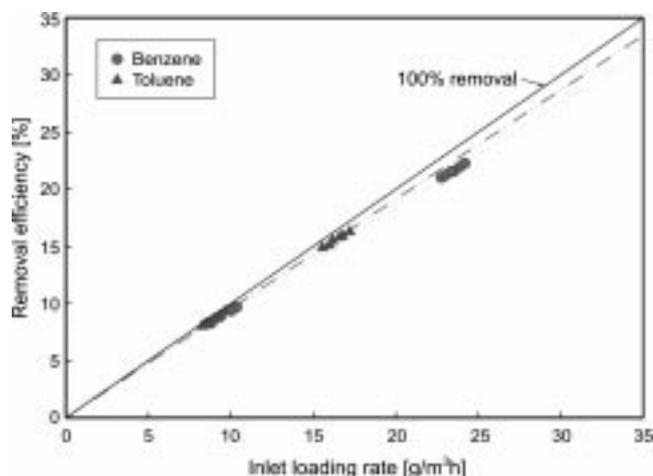


Fig. 9. The removal capacity of benzene and toluene for the inlet loading rate by a fluidized bed bioreactor at 30 °C, pH 7.0 and 150 cm liquid bed height.

and toluene was similar at 1 and 2 cm/s; however, that was reduced to 90% at 3 cm/s of gas velocity. From the data, the residence time of gas in the bioreactor was an important parameter in the operation of fluidized bed bioreactor.

Fig. 9 shows the removal capacity of benzene and toluene by a fluidized bed bioreactor for inlet loading rate. The diagonal line means 100% removal of benzene and toluene. The average removal efficiency of benzene was 94% for inlet concentration of 53(±5) ppm benzene and that of toluene was 96% for an inlet concentration of 48(±5) ppm toluene at 600 l/h of gas volumetric flow rate. The maximum removal capacity in this experimental condition was 22.3 g/m<sup>3</sup>·h for benzene and 16.3 g/m<sup>3</sup>·h for toluene.

The removal efficiency of benzene and toluene in a bench scale biofilter including granular activated carbon and yard waste compost was 53-92%, 94-98%, respectively, at inlet concentration of 4 ppb benzene and 26 ppbv toluene, and 7.0 m<sup>3</sup>/min [Webster et al., 1996]. The removal efficiency of benzene and toluene in a compost-biofilter with barks was 72%, 92%, respectively, at inlet concentration of 10-85 g/m<sup>3</sup> and gas flow rate of 320 ml/min [Yun et al., 2002]. The removal efficiency of benzene and toluene in the fluidized bed bio- reactor including biosands was better than that in the literature value, even though inlet concentrations and gas flow rates were different. From the research, it was shown that the fluidized bed bioreactor including VOC degrading microbial consortium and biosands was very effective for removing the benzene and toluene in gas phase under the high loading rate.

## CONCLUSIONS

The microbial consortium which was obtained from sewage treatment plant degraded benzene at the rate of 45.2 mg/l-h-mg cell in 30 °C and pH 7.0, and toluene at the rate of 44.4 mg/l-h-mg cell in 30 °C and pH 8.0. The optimum operating condition of the fluidized bed bioreactor including the microbial consortium was 30 °C, pH 7.0 and 150 cm of liquid bed height. The average removal efficiency

was 94% for inlet concentration of 53(±5) ppm benzene and 96% for an inlet concentration of 48(±5) ppm toluene at 600 l/h of gas volumetric flow rate. The maximum removal capacity in the experimental condition was 22.3 g/m<sup>3</sup>·h for benzene and 16.3 g/m<sup>3</sup>·h for toluene.

## ACKNOWLEDGMENTS

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## NOMENCLATURE

- $C_{in}$  : inlet concentration [ppm]  
 $C_{out}$  : outlet concentration [ppm]  
 $C_0$  : initial concentration in flask culture [ppm]  
 $C_t$  : concentration at time t in flask culture [ppm]  
 $Q$  : gas volumetric flow rate [l/h]  
 $t$  : cell culture time [h]  
 $V$  : liquid volume of the solution in bioreactor [l]  
 $\eta$  : removal efficiency [%]

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