

CHARACTERISTICS OF BENZENE, TOLUENE, AND XYLENE GAS REMOVAL BY A BIOFILTER USING SCORIA

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ABSTRACT

The biodegradation experiments of benzene, toluene, and xylene (BTX) by bacillus culture were carried out. In biofilter experiments packed with scoria as a packing media, benzene and toluene were removed over 90% and xylene was removed over 80% with empty bed contact time (EBCT) between 10 and 60 seconds when each concentration was kept constantly at 600 ppmv. The maximum removal capacities of toluene, benzene and xylene were found to be 760 g/m³-hr, 720 g/m³-hr and 450 g/m³-hr, respectively. The pH in a drain water maintained between 6.0 and 7.0 during the continuous operation for 195 days. The pressure drop had a tendency to increase slightly according to the inlet gas concentration, but was maintained low at about 5 mmH₂O/m. The biofilter using scoria media showed an excellent efficiency of BTX removal, and would be able to apply in a full-scale system with economic advantage.

Key Words : Biofilter, Biofiltration. Scoria, Benzene, Toluene, Xylene, *Bacillus* sp.

1. INTRODUCTION

Benzene, toluene, and xylene (BTX) are important industrial solvents that are frequently encountered in industrial operations including the painting and varnishing industries. A large amount of BTX is released into the atmosphere from industrial processes, petroleum refining, petroleum making, containers, and storage tanks every year. BTX vapors are toxic to the liver, kidneys and the central nervous system when

they enter the body by skin contact or breathing (Martin et al., 1998; Fishbein, 1985; Dean, 1985). Losing these substances to the ambient air may lead to an adverse impact and endanger public health and environment.

Biofiltration is an air pollution technology that utilizes microorganisms present in the biofilter media to degrade the pollutants in a waste gas stream into water and carbon dioxide. It is a very cost-effective technology for the treatment of large volume of air contaminants with low concentrations of biologically degradable compounds, as compared with the traditional VOC control technologies such as incineration, ozonation, combustion, and adsorption (Ottengraf, 1986). Especially, it is environmentally friendly because the contaminants are completely converted into non-hazardous final products.

The capacity and the durability of the biofilter depend upon the property of the media, and the removal efficiency of the pollutant will be determined by the degradation ability and the specific growth rate of microorganisms. Thus, the packing material plays an important role in biofiltration. The selection of a proper media is very important for high efficient biofiltration. However its selection depends on several factors. The medium should provide an optimum environment for microbial populations. It should have desirable properties such as high porosity, appropriate pore size, good mechanical stability, and an ability to sorb water (Ergas et al. 1992). Another important factor affecting practical application of biofilter media is cost. The less expensive the packing medium, the more desirable it is.

A wide range of packing materials including peat (Acuña et al., 2002; Zilli et al., 2001; Auria et al., 1998), compost (Morgenroth, et al., 1996; Ergas, 1994; Hodge and Devanny, 1994), sugarcane bagasse (Sene et al., 2002), granular activated carbon (Kim, 2003; Den et al., 2002; Abumaizar, 1998), polystyrene particles (Kiared, 1996), porous inorganic matrix (Kim et al., 2000), and polyurethane foam (Thalasso et al., 2001; Moe and Irvine, 1999) have been used in biofilters. However these materials may cause several problems such as compaction and channeling, thus in turn strongly influencing biofiltration performance.

Scoria is abundantly found in Jeju Island, Korea, and especially inexpensive natural porous material. It is a volcanic stone generated from the eruption of a volcano. Since it has a high water holding capacity, a high porosity, and an appropriate

intensity, it is considered as a potential medium for biofiltration process. In this study, biofiltration of benzene, toluene and xylene (BTX) compounds has been investigated in a laboratory scale-down-flow biofilter packed with Jeju scoria as a new filter material. The effects of influential factors such as influent gas concentration and empty bed contact time (EBCT) on the removal efficiency were studied, and pH and pressure drop across biofilter bed were measured.

2. MATERIALS AND METHODS

A schematic diagram of a biofilter that was used in continuous experiments was shown in Fig. 1. The biofilter was made from acryl pipe with 150 cm of height and 10 cm of diameter. Scoria of 12 - 17 mm particle size was packed in the biofilter up to 1.2 m height. The packed volume and weight were 10 L and 6.3 kg, respectively. Five check valves were installed from the top of the reactor with 30 cm interval for gas sampling ports, for three media sampling ports were installed from the bottom of the reactor with 25 cm interval. The humidifier was made with acryl (18 cm of inside diameter, 150 cm of height), was packed with a number of pall rings in order to increase the efficiency of humidification. The vaporized BTX gases in the vaporizer were introduced into the top of the biofilter and flew downwards.

Bacillus sludge was used as inoculating microorganism into the biofilter. The sludge was centrifugated for 10 min at 3000 rpm, and washed before inoculation. The cell concentration of 15,000 mg/L as MLSS was inoculated to scoria medium through circulating by nutrient supplying pump for about 3 days. MLSS and MLVSS concentration of bacillus sludge inoculated in biofilter column were 6,720 and 5,130 mg/L, respectively. The final amount of micro-organisms attached to media was 0.517 g-MLVSS/g-dry material.

The composition of a mineral salts medium used in this study were (per 1 L) : KNO_3 , 0.2 g; K_2HPO_4 , 0.9 g; $(\text{NH}_4)_2\text{SO}_4$, 0.125 g; KH_2PO_4 , 0.6 g; MgSO_4 , 0.2 g; CaCl_2 , 0.07 g; FeSO_4 , 0.01 g; EDTA, 0.02 g. The nutrients were supplied into the reactor with 10 times dilution and sprayed from the top with a flow rate of 0.5 L/day in regular periods by a timer installed on pump.

Samples were taken at the top (inlet) and at the bottom (outlet) of the biofilter bed. The concentration of benzene, toluene, and xylene were analyzed by GC (Gas

chromatography, DS-1200) which installed FID (Flame Ionization Detector). Helium gas flowing at 1 mL/min was used as the carrier gas. The temperature of the oven was 70 °C at the beginning, and increased from 70 to 150 °C at a rate of 10 °C/min and maintained at 150 °C for 1 min. The temperatures of injector and detector were 150 °C and 250 °C, respectively. Pressure drop across biofilter bed was measured by a Durablock Manometer (DWYER, USA), and pH of the drain water from biofilter was measured using a pH-meter (ORION, model 420A).

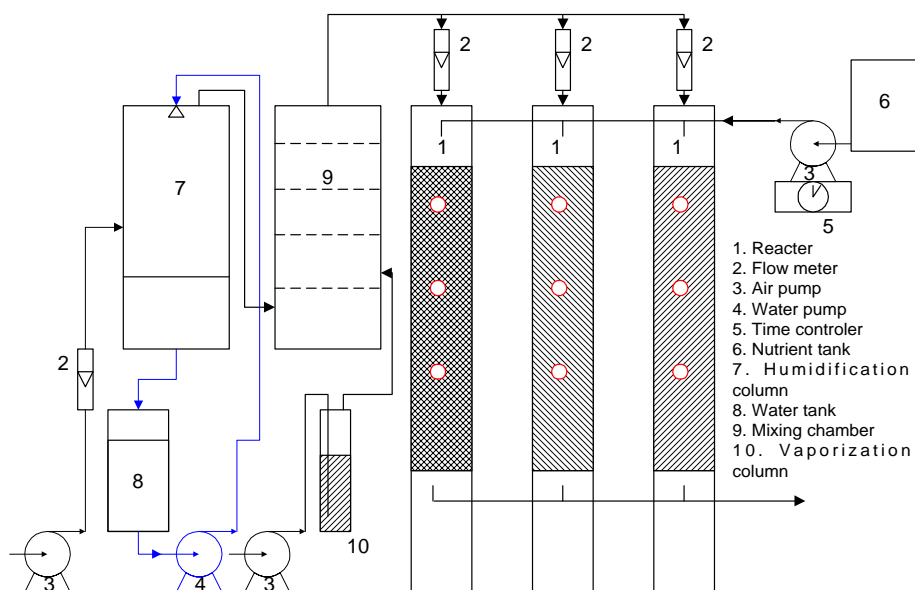


Fig. 1. Schematic diagram of biofilter.

3. RESULTS AND DISCUSSION

Fig. 2 shows the influent and effluent concentrations of each BTX compound along the operation time. Biofilter was operated continuously for 195 days in 3 different phases based on each BTX compound. The first phase began on day 1 and lasted 90 days. Toluene concentration in this phase was increased gradually from 40 ppmv to 1,200 ppmv (Fig. 2(a)). The second phase of the experiment was from day 91-150 using benzene as target gas (Fig. 2(b)). The last phase of the experiment started on day 151 by using xylene as target gas (Fig. 2(c)). At the beginning of the biofilter operation as shown in Fig. 2(a), toluene inlet concentration was fixed at 40 ppmv with 2 min EBCT for adaptation of microorganism and no toluene was detected after 3 days operation. From 9 days after the beginning of operation, the influent gas concentration was gradually increased and operated at 80, 100, 150, 300, 600, 1,000 and 1,200 ppmv for the continuous experiments. In this study, bacillus culture needed 2-4 days of adaptation time for all tested conditions. Kiared et al. (1997) reported

that acclimation period of 1 week needed when peat was used as the media. Compared to the study of Kiared et al.(1997), bacillus culture showed fast adaptation for toluene. This reason could be found not only from the property of scoria which provided good environment for microorganisms, but also from the characteristic of bacillus culture which possessed fast adaptation for toluene.

The EBCT was changed from 45 to 5 sec with a fixed concentration of BTX at 600 ppmv. Up to 10 sec EBCT, toluene was removed over 90%. When EBCT was further reduced to 5 sec, toluene was removed about 50%. Also, the inlet toluene concentration was reduced half to 300 ppmv and EBCT was reduced two-fold to 5 sec in order to make the same condition of loading rate into the biofilter as that of 10 sec EBCT at 600 ppmv. With the concentrations at less than 300 ppmv, over 98% of benzene and toluene were removed. Xylene was removed 95% with concentrations at less than 100 ppmv. These results indicated that EBCT was able to be reduced at low influent gas concentrations within the limited loading rate. Hence, this results in an economic advantage in reducing size of the system.

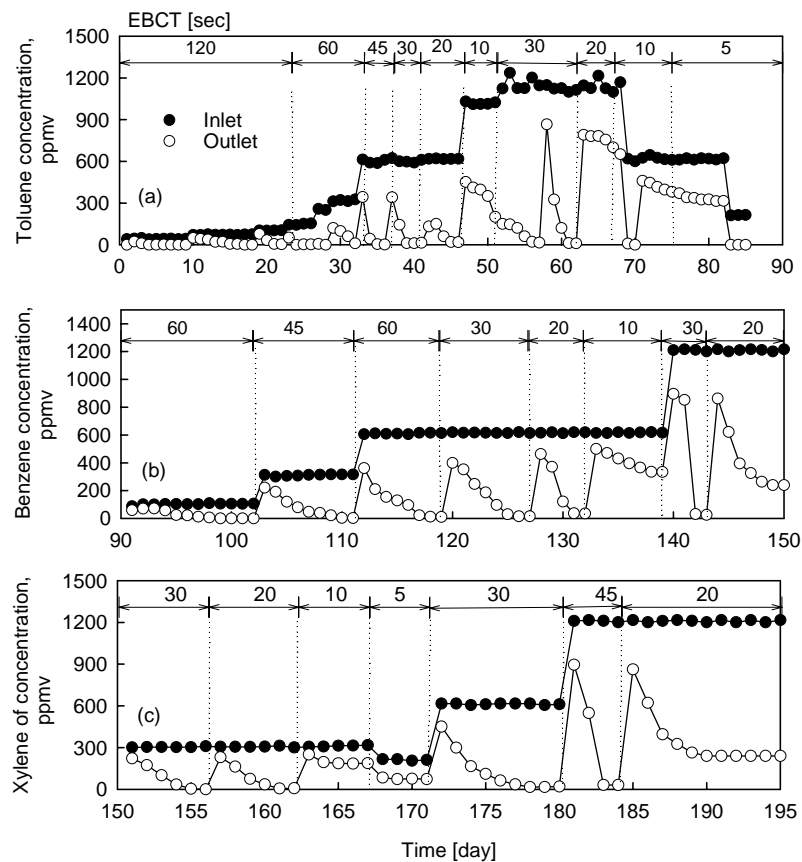


Fig. 2. Influent and effluent concentrations of (a) toluene, (b) benzene, and (c) xylene during operating time in biofilter.

For benzene shown in Fig. 2(b), the experiment started at 100 ppmv benzene with 1 min EBCT, since bacillus sludge was thought to be adapted through that of toluene removal. After 4 days operation, no benzene could be detected, and this phenomenon lasted more than 10 days. The continuous experiments were executed with the increase of benzene concentrations at a range of 100 – 1,200 ppmv. The adaptation time of bacillus sludge was more increased, compared with that of toluene, but it could be done in 4-5 days. In Fig. 2(c), the influent concentration of xylene was initially operated at 300 ppmv with 45 sec EBCT. This operation was done by the assumption that bacillus culture had been already adapted adequately. The influent xylene was gradually changed with the increase of concentrations at 300, 600 and 1,200 ppmv. For xylene, bacillus culture required more adaptation time (4-7 days) than those for other two gases.

The effect of EBCT on removal efficiency for each BTX is shown in Fig. 3. The EBCT was changed from 60 to 5 sec with a fixed concentration of BTX at 600 ppmv. Up to 10 sec EBCT, benzene and toluene were removed over 90% while xylene was removed 80%. When EBCT was further reduced to 5 sec, benzene and toluene were removed about 50% while xylene was removed 40%. This decrease in BTX removal was thought to be from a two-fold loading rate.

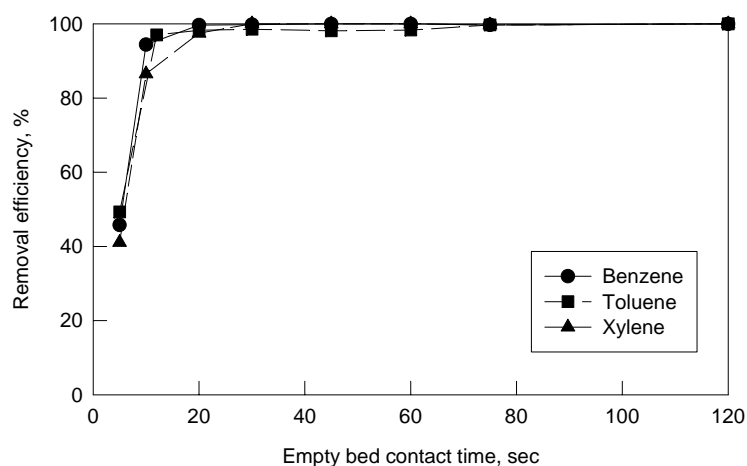


Fig. 3. Removal efficiency of benzene, toluene and xylene as a function of EBCT. (EBCT : 5 - 60 sec, influent concentration : 600 ppm).

Fig. 4 shows the individual elimination capacity of each BTX compound as a function of inlet loading rate. As shown in Fig. 4, the elimination capacity was an

increasing function of the inlet load and reached a maximum. This result revealed that a biofilter system used in this study have the maximum loading rate without inhibition to microorganisms for all tested conditions. This is a very important factor in design and operation of a biofilter system, and depends on both the operation condition and the type of media (Angela, 1996). The maximum removal capacities obtained in this study were 760, 720 and 450 $\text{g/m}^3\text{-hr}$ for toluene, benzene and xylene, respectively. The maximum removal capacities for toluene and benzene were almost the same, but that for xylene was lower.

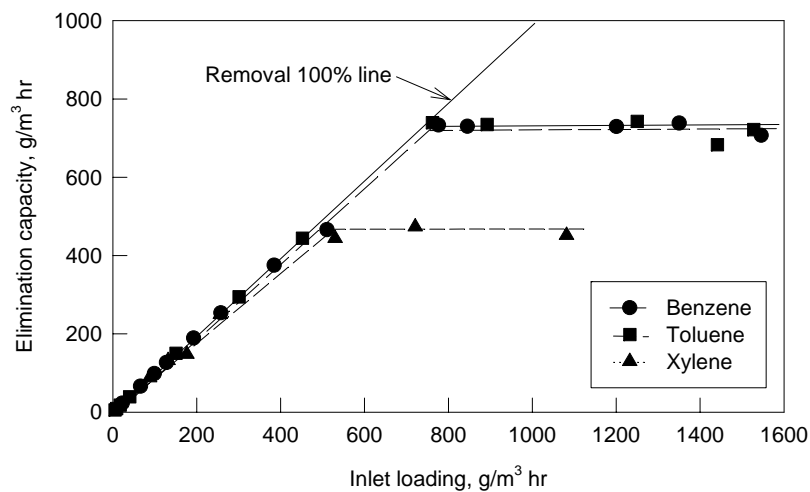


Fig. 4. Elimination capacity of benzene, toluene and xylene as a function of inlet loading rate.

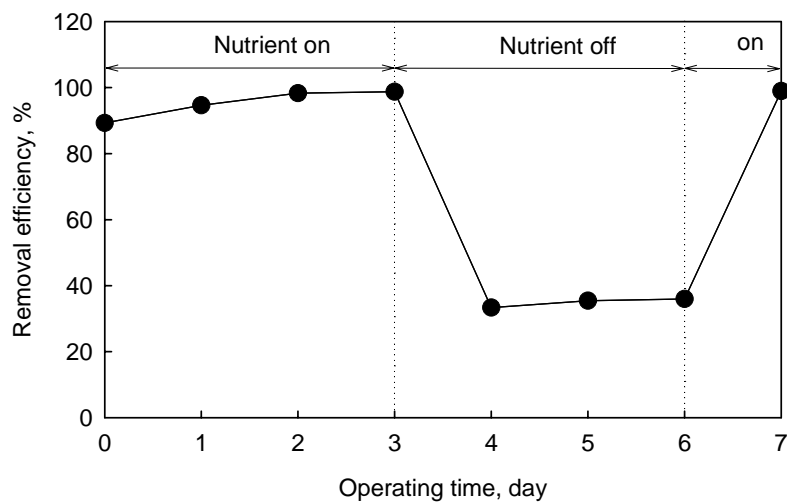


Fig. 5. The effect of removal efficiency of toluene with nutrient solution and without nutrient solution. (Influent concentration of toluene : 1200 ppmv, EBCT : 30 sec).

It has been known that the capacity of a biofilter decreases in a long-run operation when nutrients are not supplied by power failure and other operational problems, and recovering the capacity is time-consuming even though enough nutrients are supplied again (Maria et al., 1999). In Fig. 5, the removal efficiency is shown when the supply of nutrients was shut down in continuous operation and the time to recover the removal efficiency is shown when the supply of nutrients resumed. At an influent toluene concentration of 1200 ppmv with 30 and 40 sec EBCT, the removal efficiency declined from 99% to 40% when the supply of nutrients was shut down for 3 days and it recovered up to 98% within one day when the supply of nutrients resumed. This result indicated that the supply of nutrients other than toluene as the carbon source largely affected the removal efficiency and the removal efficiency quickly recovered in a day when scoria as the media was used in a biofilter.

Variations of pH and pressure drop in the biofilter during continuous operation are shown in Fig. 6. The pH maintained constantly between 6.0 and 7.0 only by intermittent spray of nutrients. The pressure drop across the biofilter bed maintained low at the average of 5 mmH₂O/m during the continuous operation.

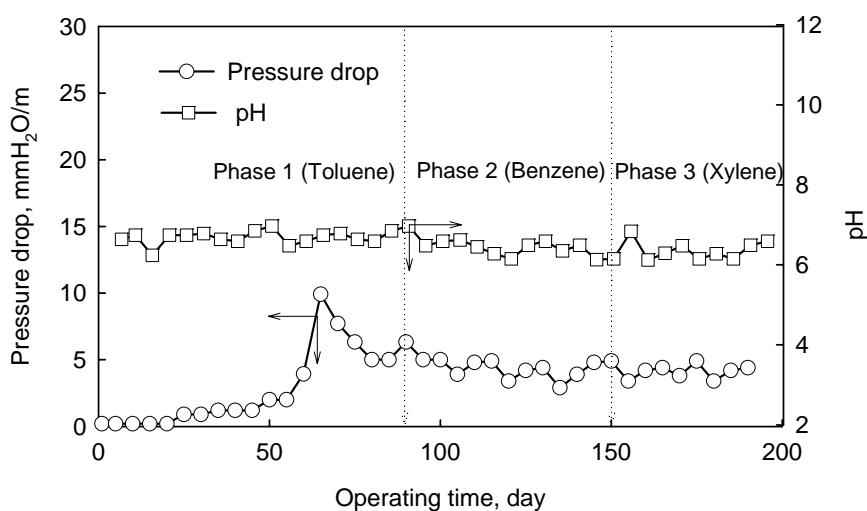


Fig. 6. Pressure drop and pH variations in the biofilter.

4. CONCLUSION

Biofiltration of BTX compounds has been investigated in a laboratory scale-down-flow biofilter packed with a new filter material. Various inlet concentrations and gas flow rates has been tested. Benzene and toluene were removed over 90 % and xylene

was removed over 80 % with EBCT between 10 and 60 seconds when each concentration was kept constantly at 600 ppmv. With 5 seconds of EBCT, benzene and toluene at the concentrations below 300 ppmv were removed over 98 % and xylene below 100 ppmv was removed over 95 %. The maximum removal capacities of toluene, benzene and xylene were found to be 760 g/m³-hr, 720 g/m³-hr and 450 g/m³-hr, respectively..

The pH in a drain water maintained between 6.0 and 7.0 during the continuous operation for 195 days. The pressure drop had a tendency to increase slightly according to the inlet gas concentration, but was maintained low at about 5 mmH₂O/m. From all the results, the biofilter using scoria showed an excellent efficiency of BTX removal, and would be able to apply in a full-scale system with economic advantage. Scoria exhibited good physical and mechanical properties as justified by the low pressure drop through the filter bed recorded during all the operation of the biofilter.

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