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Comparative studies on toluene removal and pressure drop in biofilters using different packing materials

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Abstract: *To select the best available packing material for malodorous organic gases such as toluene and benzene, biofilter performance was compared in biofilters employed different packing materials including porous ceramic (celite), Jeju scoria (lava), a mixture of granular activated carbon (GAC) and celite (GAC/celite), and cubic polyurethane foam (PU). A toluene-degrading bacterium, Stenotrophomonas maltophilia T3-c, was used as the inoculum. The maximum elimination capacities in the celite, lava, and GAC/celite biofilters were 100, 130, and 110 g m^{-3} hr^{-1}, respectively. The elimination capacity for the PU biofilter was approximately 350 g m^{-3} hr^{-1} at an inlet loading of approximately 430 g m^{-3} hr^{-1}, which was 2 to 3.5 times higher than for the other biofilters. The pressure drop gradually increased in the GAC/celite, celite and lava biofilters after 23 day due to bacterial over-growth, and the toluene removal efficiency remarkably decreased with increasing pressure drop. Backwashing method was not effective for the control of biomass in these biofilters. In the PU biofilter, however, backwashing allowed maintenance of a pressure drop of 1 to 3 mm H_2O m^{-1} and a removal efficiency of > 80%, indicating that the PU was the best packing material for toluene removal among the packing materials tested.*

Key words: *Toluene, Biofilter, Packing material, Pressure drop, Clogging, Stenotrophomonas maltophilia*
PDF of full length paper is available online

Introduction

Aromatic hydrocarbons such as benzene, toluene, ethylbenzene and xylene are representative volatile organic compounds (VOCs), which can be harmful effect for human health. The substances emitting offensive odor may lead to unfavorable impact on human health and welfare (Kiared *et al.,* 1996; Celik *et al.,* 2008). Biofiltration technology has been exceedingly developed for the control of low concentration VOCs emitted from various industries due to its simple operation, relatively lower capital and operating cost than conventional technology such as physical and chemical treatment as well as the harmless final product such as water and carbon dioxide and environmentally friendly (van Groenestijn and Hesselink 1993; Lee *et al.,* 2002b; Kwon *et al.,* 2003; Lee *et al*., 2009; Lawansiri *et al.,* 2008; Chouy Chai *et al.,* 2009; Lawansiri *et al.,* 2008; Chouychai *et al.,* 2009). Applications of some researches for the control of odor and air pollutant from a variety of industries and public sources have been done (van Lith *et al.,* 1997; Wani *et al.,* 1997; Kam *et al.,* 2005; Kim *et al.,* 2005; Shim *et al.,* 2006). Whereas the biofilteration of VOCs have been used as a good treatment method, pressure drop and clogging caused by excess biomass growth in biofilter during extended operation are still problematic (Kwon *et al.,* 2003). Therefore, limiting excessive biomass growth is essential for the biofilter success (Devinny *et al*., 1999; Delhomenie *et al.,* 2003).

Recent researches in biofilteration have concentrated on the mechanical development of the packing material and reduction of pressure drop, which are the main operating cost of biofiltration (Iliuta and Larachi, 2004). The properties as ideal packing materials are required a high porosity to reduce head loss as well as a high specific surface area for the attachment of microorganisms. Compost, peat, wood chips, coconut fiber, pine leaves, celite, activated carbons, porous lava, polystyrene spheres, and polyurethane have been employed as packing materials in biofilters (Weber and Hartmans, 1995; Lee *et al.,* 2002b, 2005; Mendoza *et al.,* 2004; Kim and Sorial, 2007; Maestre *et al.,* 2007).

In this study, to select the best available packing material for VOCs control, it was compared the removal efficiency of toluene in biofilters packed with different packing materials, including porous ceramic (celite), Jeju scoria (lava), a mixture of granular activated carbon (GAC) and celite (GAC/celite), and cubic polyurethane (PU) foam. We also monitored pressure drop and clogging during toluene degradation.

Materials and Methods

Packing materials: Four different types of packing materials were used in this study: celite (Ssang Yong Co. Ltd, Korea), lava (Jeju Island, Korea), a 1:1 (v/v) GAC (Kaya Activated Carbon Inc., Korea)/celite mixture, and cubic PU (Seilsponge, Korea) with a dimension of 1.0 cm × 1.0 cm × 1.0 cm. The mean particle sizes of celite, lava, and GAC were 9.0, 12.0, and 5.2 mm, respectively. The physical properties of each packing material were described in our previous studies (Cho *et al.,* 2000; Lee *et al.,* 2002a; Kwon *et al.,* 2003).

Inoculum Source: *Stenotrophomonas maltophilia* T3-c was used as the inoculum for the biofilters. This strain was cultured in LB medium (10 g l^{-1} tryptone, 5 g l^{-1} yeast extract and 5 g l^{-1} NaCl) and harvested by centrifugation at 7600 g for 5 min. The collected cells were resuspended in 0.4 l of minimal salt medium (1.5 g l^{-1} KH_2PO_4, 9 g l^{-1} $Na_2HPO_4 \cdot 12H_2O$, 3 g l^{-1} $(NH_4)_2SO_4$, 0.01 g l^{-1} $CaCl_2 \cdot 12H_2O$, and 0.15 g l^{-1} $MgSO_4$). Next, the immobilized packing materials

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were prepared by soaking them in the concentrated cell suspension.

Biofilter and operation conditions: Fig. 1 shows a schematic diagram of the biofilters used in this study. To compare toluene removal by biofilters packed with the four packing materials (celite, lava, GAC/celite and PU), four cylinder-type acrylic column biofilters (120 mm internal diameter × 1700 mm height) were built. Each biofilter bed was packed with 800 mm of *S. maltophilia* T3-c immobilized filter material (packing volume = 9.0 l). A grade 304 stainless steel screen (20 x 20 mesh) was placed on the bottom of the filter bed to support the packing material.

The biofilters were equipped with a drain storage tank, a circulation pump, and a liquid distributor for adding water. The top of the biofilter was sprayed six times per day with 2 l of tap water supplemented with mineral salts to supply the bacteria with nutrients and to prevent the packing material from drying. Wastewater accumulating in the drain storage tank was removed every two days.

Compressed air flowed through a volatilization chamber before entering the biofilter. The chamber was made of stainless-steel tube (20 mm internal diameter × 100 mm height) connected to a liquid injection system consisting of a peristaltic pump and a liquid toluene storage bottle. Toluene vapor was generated by injecting pure liquid toluene at 0.001-20 ml min^{-1} into the volatilization chamber air stream using a M930 peristaltic pump (Young-Lin Instrument Co. Ltd., Korea). The desired toluene concentration of the vapor was obtained by adjusting the injection rate of the solution and the flow rate of the air stream. Space velocity (SV) levels varied from 50 to 800 hr^{-1} and the toluene inlet concentration varied from 26 to 470 ppm (0.1-1.8 g m^{-3}). Toluene degradation in the biofilters was evaluated by measuring the inlet and outlet concentrations of toluene at different SVs and inlet concentrations.

Analytical methods: The toluene concentration was measured using a gas chromatograph (HP 5890 series II *plus*; Hewlett Packard Co., Wilmington, DE, USA), which was equipped with a flame ionization detector and a DB-WAX column (30 m × 0.32 mm × 0.25 μm, J and W Scientific, Folsom, CA, USA). The analytical conditions (Kwon *et al.,* 2003) were followed. The detection limit of this procedure was 0.1 ppm toluene. The pressure drop in the biofilter was measured using a H_2O manometer equipped between the inlet and outlet section of the filter bed.

Results and Discussion

Toluene degradation characteristics by biofilter packed with various media such as granular type of celite, lava, GAC/celite and porous polyurethane (PU) as biofilter media were investigated. The elimination of toluene in the biofilters immobilized with *S. maltophilia* T3-c was stabilized for 2-3 days operation. Except the PU biofilter, inlet toluene concentrations for the other three biofilters were randomly changed to maximum 1.8 g m^{-3} (470 ppm) and space velocity (SV) was changed stepwise from 50 to 150 hr^{-1} (Fig. 2A-C). As the result, the high toluene elimination efficiency over 95% by each three biofilter was maintained for 15-20 days. Then the elimination efficiencies for toluene by celite, lava and celite/GAC biofilter were gradually reduced from 95 to 40-50% for 35-42 days

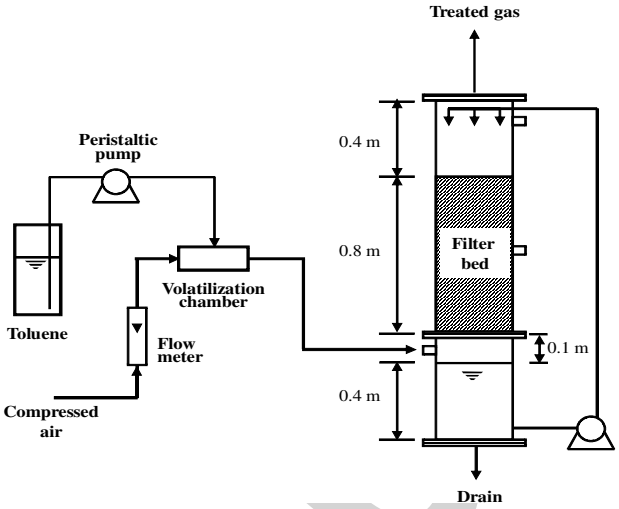

Fig. 1: Schematic diagram of the experimental apparatus

of operation. In the PU biofilter, the space velocity was changed stepwise from 200 to 800 hr^{-1}. The elimination efficiency over 95% was maintained under the condition of above 400 hr^{-1} for 10 days of initial operating period when inlet toluene concentration of below 1 g m^{-3} (260 ppm) was injected. Although the SV was changed from 600 to 800 hr^{-1}, the elimination efficiency of toluene was maintained above 80%. In the other three biofilters, the elimination efficiency was significantly decreased as extending operation period, but it was maintained steady for 30 days in the PU biofilter.

Fig. 3 shows elimination capacity at various inlet loadings in the biofilters packed with celite, lava, GAC/celite, and PU. The maximum elimination capacities in the celite, lava, and GAC/celite biofilter were 100, 130 and 110 g $m^{-3}hr^{-1}$, respectively. However, the elimination capacity for the PU biofilter was approximately 360 g $m^{-3}hr^{-1}$ at an inlet loading of approximately 430 g $m^{-3}hr^{-1}$, which was 2 to 3.5 times higher than for the other biofilters.

Fig. 4 presents the changes in the pressure drop in the various biofilters during operation. During the first 15 days, the pressure drop was not significantly different for the various biofilters. After 15 days, however, the pressure drop gradually increased in the order of GAC/celite < celite < lava. In particular, the pressure drop in the GAC/celite biofilter at a SV of 150 and 300 hr^{-1} dramatically increased to 230 and 750 mm H_2O m^{-1}, respectively at 25 days. However, in the PU biofilter, when the SV was 300 hr^{-1}, there was a low pressure drop of approximately 1 to 3 mm H_2O m^{-1}. In the celite, lava, and GAC/celite biofilters, the elimination efficiency for toluene was closely associated with the pressure drop (Fig. 2, 4). During the initial operating period, where there was a lower pressure drop, the elimination efficiency remained above 95% (Fig. 2). After 15 to 20 days, when the pressure drop began to increase, the elimination efficiency decreased (Fig. 2). After 30 to 40 days, when there was a dramatic increase in the pressure drop, the elimination efficiency decreased to 40 to 65% (Fig. 2). The increase in the pressure drop in the biofilters was due to clogging as the result of biomass growth on the packing materials (Iliuta and Larachi, 2004; Mendoza *et al.,* 2004; Kim and Sorial, 2007).

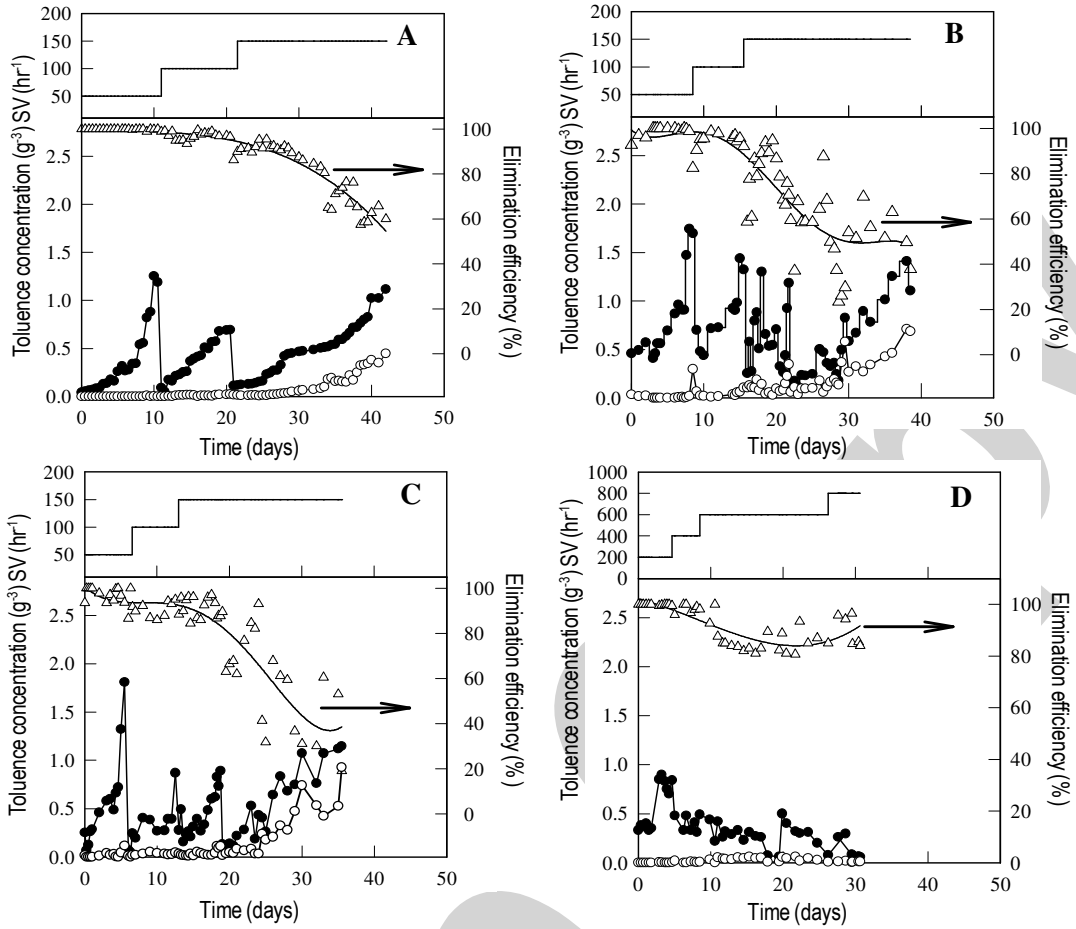

Fig. 2: Toluene degradation in the biofilters packed with different packing materials. A. celite, B. lava, C. GAC+celite, D. PU, Symbols: ●, inlet concentration; ○, outlet concentration; △, removal efficiency

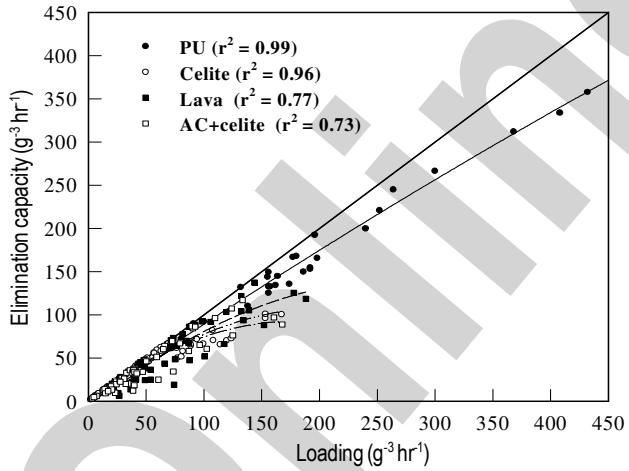

Fig. 3: Elimination capacity for toluene by *S. maltophilia* T3-c in various biofilters. Symbols: ●, PU; ○, celite; ■, lava; □, GAC/celite mixture

The biomass growth on the celite packing material operated for each 10th, 20th and 40th day was shown in Fig. 5A. During the subsequent period, the biofilm growth created increasing resistance to gas flow through the porous bed because the void space available for convection was gradually reduced. Clogging caused the appearance of a black color in the biofilter due to anaerobic

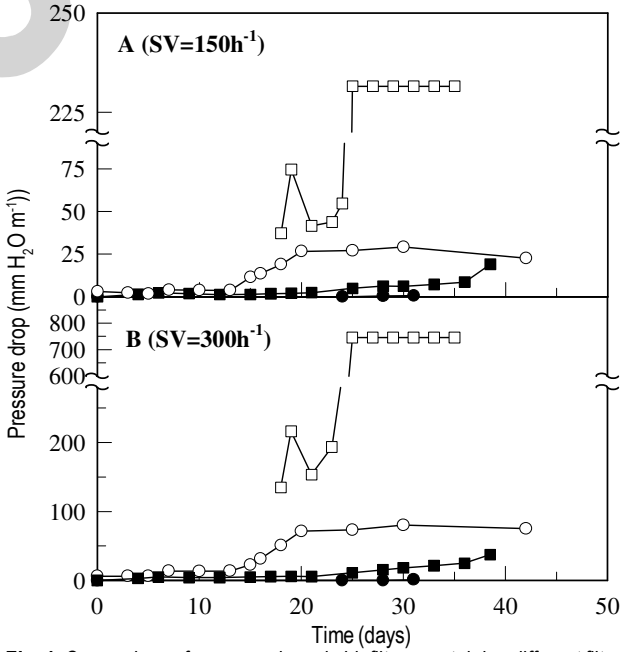

Fig. 4: Comparison of pressure drops in biofilters containing different filter media. A. SV = 150 h^{-1}, B. SV = 300 h^{-1}. Symbols: ●, PU; ○, celite; ■, lava; □, GAC/celite mixture

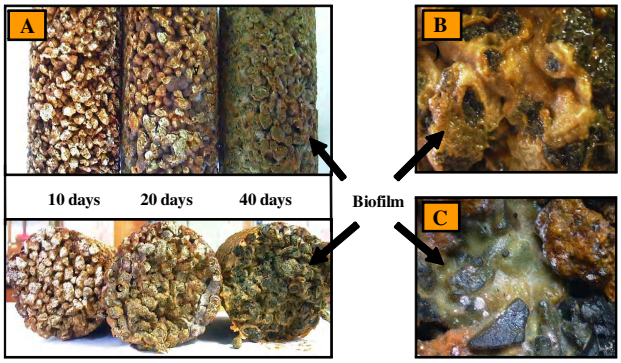

Fig. 5: Biomass grown on various packing materials. A. Celite at 10th, 20th and 40th day, B. Lava at 40th day, C. GAC/celite mixture at 40th day

degradation of the biomass. This occurred not only in the celite biofilter but also in the lava and GAC/celite biofilters (Fig. 5 B,C).

Physical, chemical, and biological treatments are generally used to remove excess biomass in biofilters (Iliuta and Larachi, 2004). Physical methods consist mainly of stirring the bed or draining the accumulated biomass by backwashing with water (Wübker *et al.,* 1997; Laurenzis *et al.,* 1998; Delhomenie *et al.,* 2003). Chemical methods use reagents that directly lyse the biomass or weaken the chemical bonds between the biofilm and the surface of the packing material (Diks *et al.,* 1994; Schonduve *et al.,* 1996; Chen and Stewart, 2000; Delhomenie *et al.,* 2003). Biological methods use microorganisms that can feed on or degrade the excess biomass in the filter bed (Iliuta and Larachi, 2004). In this study, to remove excess biomass from the packing materials, we attempted to use backwashing by spraying water on the filter and then passing compressed air through it. In the celite, lava and GAC/celite biofilters, this method was not effective at removing the biomass, but it was effective in the PU biofilter. The results suggest that it was possible to maintain the microbial activity in the PU filter during the long-term degradation of toluene because of the easy removal of excess biomass by backwashing.

Acknowledgments

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