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SHORT COMMUNICATION

The Treatment of Waste-air Containing Mixed Solvent using a Biofilter 2. Treatment of Waste-air Containing Ethanol and Toluene in a Biofilter

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Abstract−An experiment for five stages of a biofilter-run was performed to investigate the effect of hydrophilic ethanol and hydrophobic toluene on the biodegradation of hydrophobic toluene and hydrophilic ethanol, respectively, when waste-air containing toluene and ethanol was treated by a biofilter. Removal efficiencies of toluene and ethanol began to decrease when inlet load surpassed 90 g/m^3/h and 100 g/m^3/h consistent with maximum elimination capacities of toluene and ethanol, respectively. At the end of the biofilter-run, removal efficiencies for toluene and ethanol were decreased and maintained at 65% and 40%, respectively. The concentration of toluene at 1st sampling port was raised by factor of two in the 3rd stage of the biofilter run when the inlet load of ethanol co-feed was increased by 1.5 times, while the process conditions of toluene were maintained the same as those of the 2nd stage of biofilter-run. According to the result of Mohseni and Allen, it may be interpreted that removal efficiency of hydrophobic toluene was affected by the presence of hydrophilic ethanol when high load of hydrophobic toluene was applied like that of the 1st sampling port of the biofilter. However it was not the case when a low load of hydrophobic toluene was applied like those of the 2nd, 3rd and 4th sampling ports since hydrophobicity of toluene is much less that of α-pinene. Thus, it may be suggested that biodegradation of hydrophobic VOC was interfered by hydrophilic VOC dissolved in the biolayer and the degree of interference was proportional to the inlet load of hydrophobic VOC as well as that of hydrophilic VOC and was inversely proportional to the solubility of hydrophobic VOC. However, it was inferred that the existence of hydrophobic toluene from waste-air can hardly inversely hinder the removal of hydrophilic ethanol in the biofilter when time-evolutions of hydrophilic ethanol concentrations of this experiment were compared with those of the previous experiment of biofilter to treat waste-air containing ethanol only.

Key words: Biofilter, Mixed Solvent, Degree of Interference, Solubility of Hydrophobic VOC, Hydrophilic Co-feed

INTRODUCTION

As a promising air pollution control technology biofiltration has emerged so as to treat waste air containing volatile organic compounds (VOCs) as well as odoriferous compounds. Many researchers have investigated biofilter performance to eliminate VOCs from waste gas [Ottengraf, 1986; Deshusses et al., 1995; Deshusses and Dunn, 1994; Deshusses and Hamer, 1993; Lim and Lee, 2003; Buchner, 1989; Leson and Winer, 1991; Sorial et al., 1995; Leson and Smith, 1997; Swanson and Loehr, 1997; Ottengraf and van den Oever, 1983; Zarook and Baltzis, 1994; Mohseni and Allen, 2000; Tang et al., 1995; Jorio et al., 1998; Hodge and Devinny, 1994, 1995; Shim et al., 1995; Arulneyam and Swaminathan, 2000; Auria et al., 1998; Christine et al., 2002; Lim and Park, 2004, 2005]. Performance of biofilter treating waste-air containing VOCs has been known to depend on their solubility in the biolayer of the biofilter [Ottengraf and van den Oever, 1983; Zarook and Baltzis, 1994]. Hydrophilic methanol and hydrophobic α-pinene existing in natural wood are major air pollutants generated from pulp and paper industries. The former is one of 189 hazardous air pollutants (HAP) referred to in Clean Air Act Amendment announced by EPA in 1990 and the latter is so hydrophobic that its water-solubility may be 5-10 mg/L. It was reported that removal efficiency of hydrophobic VOC (α-pinene) dropped more significantly up to 74% as the concentration of hydrophilic VOC (methanol) increased for the simultaneous biofilter-treatment of hydrophobic VOC (α-pinene) and hydrophilic VOC (methanol) than that for the sole treatment of hydrophobic VOC (α-pinene) of 91.2% [Mohseni and Allen, 2000]. Toluene and ethanol are designated as hydrophobic and hydrophilic VOCs, respectively, in order to perform experiments for the simultaneous treatment of hydrophobic and hydrophilic VOCs, since they have been emitted very frequently from various industries and their emissions have been under regulatory control even though hydrophobicity of toluene is much less than that of α-pinene (The water solubility of toluene is about 526 mg/L). The experiment for five stages of biofilter-run is performed to investigate the effect of hydrophilic ethanol and hydrophobic toluene on the biodegradation of hydrophobic toluene and hydrophilic ethanol, respectively, when waste-air containing toluene and ethanol is treated by a biofilter. Its results are compared to those of the works [Lim and Park, 2004, 2005], where transient behavior of biofilter to treat waste-air containing ethanol was observed under the same operating conditions of ethanol as the part two of the work adopts, and are evaluated in many respects.

EXPERIMENTAL

The biofilter system and its packing media were designed and buffer and mineral solution was provided in the same way as in the work of Lim and Park [2004] to treat waste-air containing both hydrophilic ethanol and hydrophobic toluene. A schematic diagram

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of the biofilter process and feeding schedule of buffer solution are shown as Figs. 1 and 2, respectively. Temperature of biofilter column was loosely controlled between 26 °C and 35 °C by turning the heating band on and off to simulate natural temperature variation in the field as in Fig. 3.

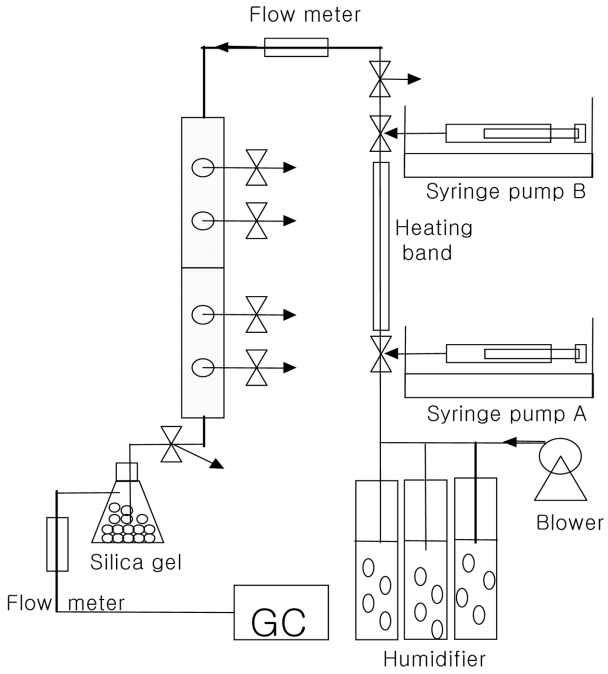

Fig. 1. Schematic diagram of biofilter treating waste-air containing mixed solvent.

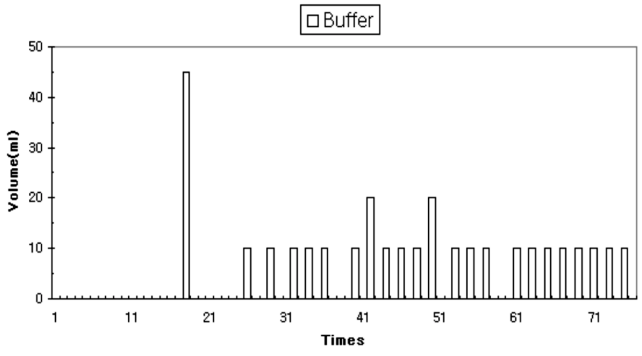

Fig. 2. Feeding schedule of buffer solution to biofilter.

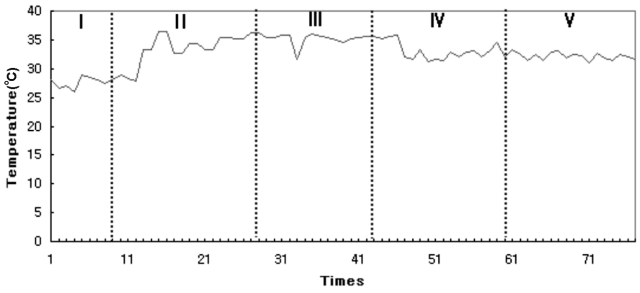

Fig. 3. Temperature schedule for the operation of biofilter.

Microorganisms of *Pseudomonas putida* (KCTC 1768) and *Burkholderia cepacia* G4 from Inje University were incubated and their mixed culture was inoculated before the count of microbes fixed on packing media were determined in the same way as in the work of Lim and Park [2005]. The biofilter was run at 26-35 °C under various operating conditions on toluene and ethanol as shown in Tables 1 and 2, respectively, for 39 days (total 77 times with measuring frequency of two times per day). Analytical methods to measure the concentration of ethanol and toluene from four sampling ports were also the same as in the work of Lim and Park [2005].

RESULTS AND DISCUSSION

1. Time Evolutions of Hydrophobic Toluene Concentrations and the Effect of Hydrophilic Ethanol Co-feed at Four Sampling Ports

Transient behavior of toluene concentrations measured at the position of feed inlet and four sampling ports of biofilter is shown as in Fig. 4. After excess amount of 45 ml buffer solution was poured into the biofilter at 18th time of 2nd stage operation (9-26 times) of biofilter, abnormal largest concentration peaks appeared first at the 1st sampling port. For the lower sampling port less abnormal peaks of toluene concentration appeared than that of the upper sampling port as shown in Fig. 4. It was noticed that all abnormal peaks of toluene concentrations from four sampling ports appeared at the same time unlike those of ethanol concentrations from this part two of the work as well as those of ethanol concentrations from part one of the work [Lim and Park, 2004]. It was also observed that removal efficiency of toluene was diminished due to such flooding effects of excess supply of buffer solution as temporary loss of interface between waste-air and biolayer and reduced effective height of biofilter as shown in Fig. 5. Later, the removal efficiency of toluene

Table 1. Theoretical values of operating condition on toluene from each stage of biofilter-run

Theoretical value \ Stage (times)	1st stage (1-8)	2nd stage (9-26)	3rd stage (27-42)	4th stage (43-58)	5th stage (59-77)
m (μl/min)	0.29	0.58	0.58	1.16	2.32
Q (L/min)	0.25	0.5	0.5	0.5	1.0
C_{go} (ppm)	278	278	278	556	556
C_{go} (g/m^3)	1	1	1	2	2
τ (min)	2.98	1.49	1.49	1.49	0.75
Inlet load (g/m^3/h)	20.13	40.27	40.27	80.54	160

※ m: toluene injection rate at a syringe pump, Q: air flow rate, C_{go}: feed concentration, τ: EBCT (effective height: 0.38 m)

Table 2. Theoretical values of operating condition on ethanol from each stage of biofilter

Theoretical value \ Stage (times)	1st stage (1-8)	2nd stage (9-26)	3rd stage (27-42)	4th stage (43-58)	5th stage (59-77)
m (μl/min)	0.83	1.67	2.5		5.0
Q (L/min)	0.25	0.5	0.5	0.5	1.0
C_{go} (ppm)	1,450	1,450	2,180	2,180	2,180
C_{go} (g/m^3)	2.62	2.62	3.93	3.933.93	
τ (min)	2.98	1.49	1.49	1.49	0.75
Inlet load (g/m^3/h)	52.75	105.50	158.26	158.26	316.51

※ m: ethanol injection rate at a syringe pump, Q: air flow rate, C_{go}: feed concentration, τ: EBCT (effective height: 0.38 m)

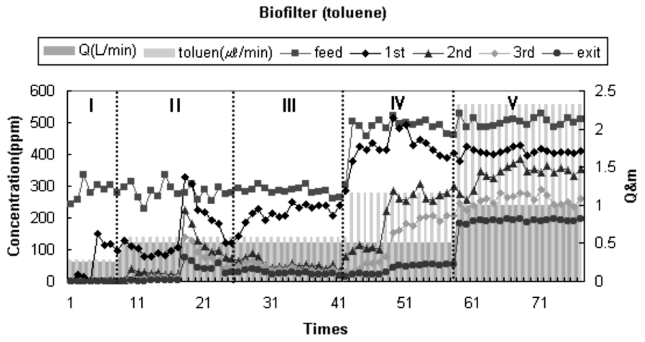

Fig. 4. Various toluene concentrations of biofilter at each sampling port versus experimental times.

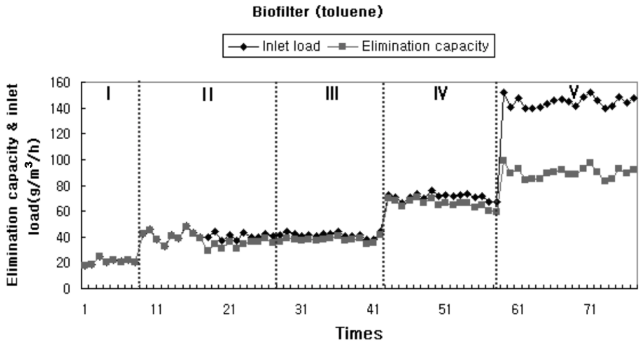

Fig. 6. Elimination capacity (g/m^3/h) and inlet load versus times.

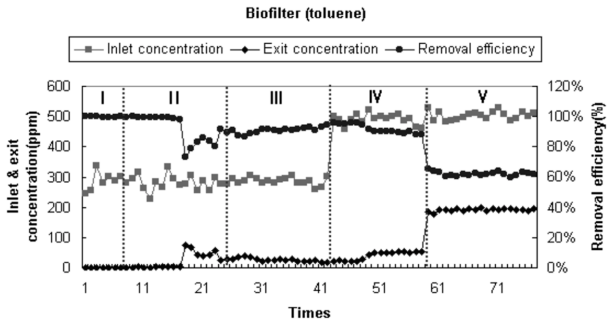

Fig. 5. Removal efficiency, inlet and exit concentrations versus times.

was observed to recover the status prior to flooding.

The inlet load of ethanol co-feed was increased by 1.5 times while the process conditions of toluene were maintained the same as those of the 2nd stage of operation at the 3rd stage of operation (43 times-58 times). The concentration of toluene at the 1st sampling port was raised by a factor of two at the 3rd stage of biofilter-run, while the effect of hydrophilic ethanol co-feed was observed negligible on transient behavior of hydrophobic toluene concentrations at the 2nd, 3rd and 4th sampling ports, compared to those of the 2nd stage of biofilter-run as shown in Fig. 4, except for abnormal peak of toluene concentration due to flooding effect, in the similar manner to the previous investigation treating mixed solvent vapors with hybrid system composed of a biofilter and photo-catalytic reactor [Lim and Park, 2005] where the concentration of toluene at 1st sampling port was raised by 50% with the same operating conditions. Mohseni and Allen [2000] reported that removal efficiency of hydrophobic VOC (α-pinene) dropped more significantly up to 74% as the concentration of hydrophilic VOC (methanol) increased for the simultaneous treatment of hydrophobic VOC (α-pinene) and hydrophilic VOC (methanol) than that for the sole treatment of hydrophobic VOC (α-pinene) of 91.2%. According to the result of Mohseni and Allen [2000] it may be interpreted that removal efficiency of hydrophobic toluene was affected by the presence of hydrophilic ethanol when high load of hydrophobic toluene was applied like that of the 1st sampling port of the biofilter. However, it was not the case when a low load of hydrophobic toluene was applied like those of the 2nd, 3rd and 4th sampling ports since hydrophobicity of toluene is much less that of α-pinene. Thus, it may be suggested that biodegradation of hydrophobic VOC was interfered by hydrophilic VOC dissolved in biolayer, and the degree of interference was proportional to the inlet load of hydrophobic VOC as well as that of hydrophilic VOC and was inversely proportional to the solubility of hydrophobic VOC.

Time-evolutions of removal efficiency and elimination capacity versus inlet load are shown as in Figs. 5 and 6, respectively. The removal efficiency of toluene maintained almost 100%, as in Fig. 5, in early biofilter-run. However, in the end of the biofilter-run its removal efficiency was maintained at 65% after it began to decrease when inlet load surpassed the maximum elimination capacity of 90 g/m^3/h, as in Figs 6 and 7.

2. Time Evolutions of Hydrophilic Ethanol Concentrations and the Effect of Hydrophobic Toluene Co-feed at Four Sampling Ports

Transient behavior of ethanol concentrations measured at the position of feed inlet and four sampling ports of the fore-said biofilter is shown as in Figs. 8a, 8b and 8c.

After excess amount of 45 ml buffer solution was poured into the biofilter at 18th time of 2nd stage operation (9-26 times) of biofilter, abnormal largest concentration peak appeared first at the 1st sampling port. For the lower sampling port less abnormal peak appeared later with a time interval than that of the upper sampling port as shown in Figs. 8a, 8b and 8c. The removal efficiency of ethanol was observed to decrease due to such flooding effects of excess supply of buffer solution and to recover the status prior to flooding later as shown in Figs. 9a and 9b from the part one of this work [Lim and Park, 2004], which was similar fashion to that of toluene.

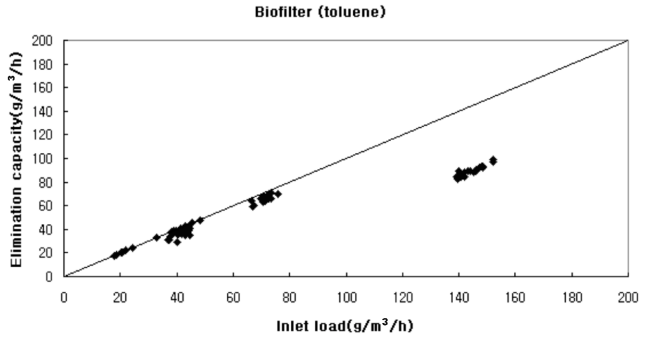

Fig. 7. Elimination capacity (g/m^3/h) versus inlet load of toluene at the exit of biofilter.

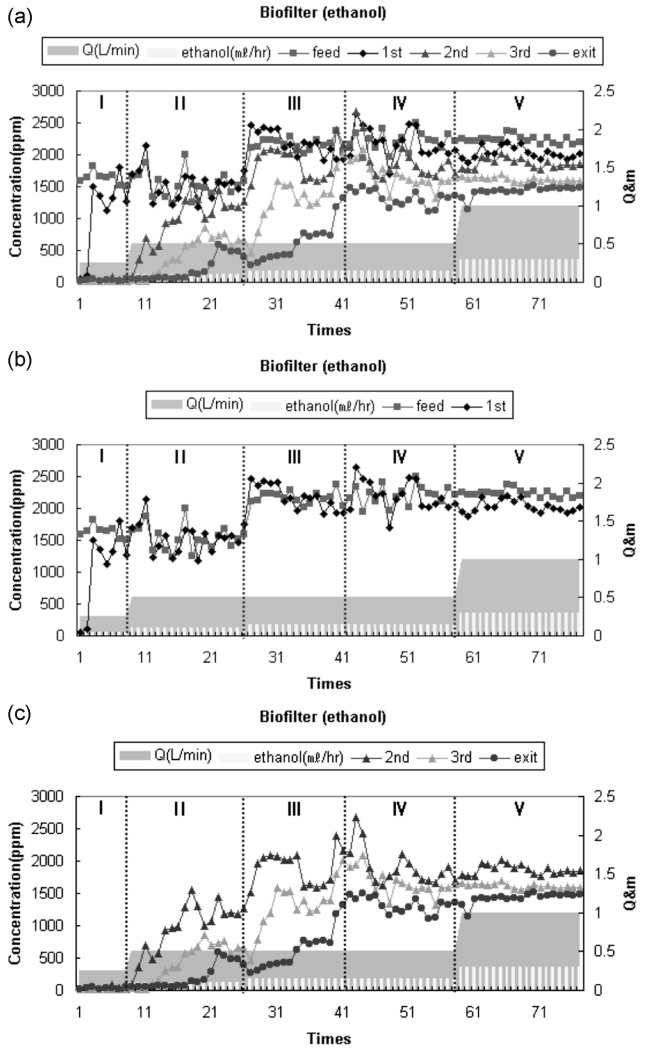

Fig. 8. (a) Various ethanol concentrations of biofilter at each sampling port versus experimental times. (b) Various ethanol concentrations of biofilter at feed inlet and 1st sampling port. (c) Various ethanol concentrations of biofilter at 2nd, 3rd and exit sampling ports.

At 4th stage of biofilter-run (43-58 times) process conditions of ethanol were maintained as the same as those of the 3rd stage of operation except for temperature of biofilter. 10 ml of buffer solution was provided intermittently to the biofilter except for the 43rd time and 50th time of biofilter-run when each of 20 ml buffer solution was provided. Each time-evolution of ethanol concentration at each sampling port was observed sharply increased around these two times (i.e., 43rd time and 50th time) of 4th stage of biofilter-run and lowered, as in Figs. 8a, 8b and 8c. It may be attributed that relatively large amount of buffer solution provided at 43rd time and 50th time of biofilter-run caused flooding in the biofilter, and it recovered the status prior to the flooding in consideration that experimental times of abnormal concentration peaks of ethanol (in particular, from 2nd sampling port) shown at 4th stage of biofilter-run were exactly matched with those (i.e., 43rd time and 50th time) when each of 20ml-buffer solution was provided to the biofilter.

Time-evolution of elimination capacity versus inlet load is shown as in Fig. 10. In early time-evolution of removal efficiency as in Fig. 9a, it maintained almost 100%. However, it began to decrease when inlet load surpassed, as in Fig. 10, 100 g/m^3/h consistent with maximum elimination capacity shown as in Fig. 11. At the end of the biofilter-run removal efficiency was decreased and maintained at 40%. During the whole biofilter-run, time-evolution of removal

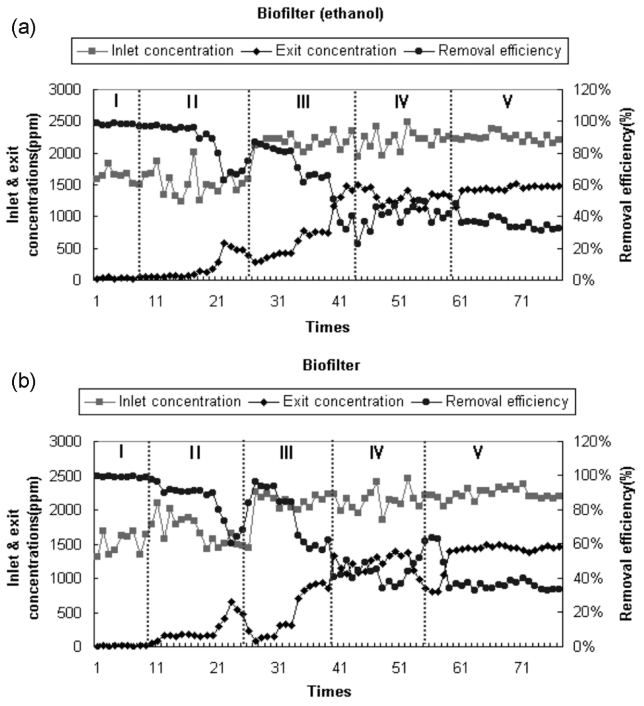

Fig. 9. (a) Removal efficiency, inlet and exit concentrations versus times. (b) Removal efficiency, inlet and exit concentrations versus times [Lim and Park, 2004].

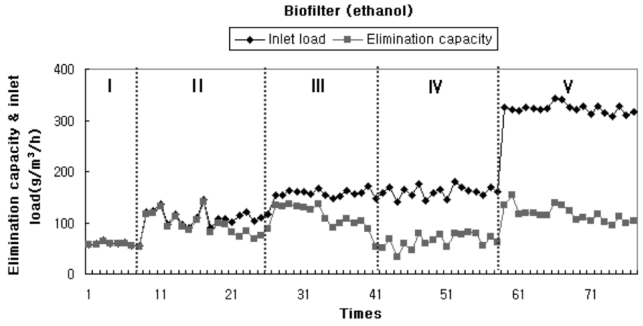

Fig. 10. Elimination capacity (g/m³/h) and inlet load versus times.

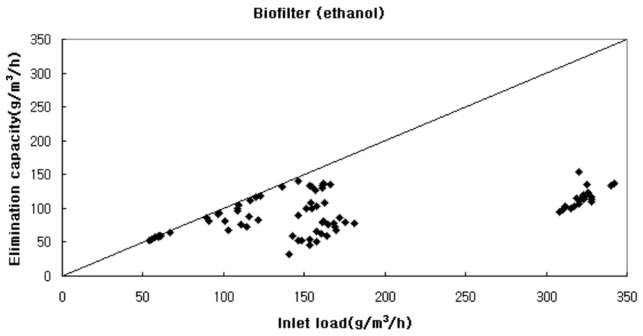

Fig. 11. Elimination capacity (g/m³/h) versus inlet load of ethanol at the exit of biofilter.

efficiency was quite similar to that as shown in Fig. 9b from part one of this work [Lim and Park, 2004] where waste-air contained only ethanol. This comparison inferred that the existence of hydrophobic toluene from waste-air can hardly affect the removal of hydrophilic ethanol in the biofilter. Moreover, this inference was consistent with the experimental result that the effect of hydrophobic toluene co-feed was observed negligible, of Lim and Park [2005] where the ethanol concentrations, in 4th stage of hybrid system-run, at every sampling port remained almost the same as in the previous stage of hybrid system-run even though the inlet load of toluene co-feed was increased by factor of two.

3. Analysis of Packing Media

The density of packing media was 0.40 and the initially neutral pH of packing media from 2nd and 4th sampling ports changed to 5.65 and 6.20, respectively, at the end of biofilter experiments. Moisture contents of the media from these two sampling ports were 52.50% and 54.45%, respectively. The total bacterial numbers (TBN) for the 1st, 2nd and 3rd sampling ports were 1.72×10^9/g, 9.03×10^8/g and 6.15×10^8/g, respectively, in such a way that TBN was decreased as the effective height was increased. Microbes were observed by fluorescence microscope (16×100) (Axiolab, Xeiss, Germany) UV filter (G365, LP395, FT420) as shown in Fig. 12.

CONCLUSION

After excess amount of 45 ml buffer solution was poured into the biofilter, less abnormal peaks of toluene and ethanol concentrations appeared for the lower sampling port at the same time as and later with a time interval than, respectively, those of the upper sampling port. The removal efficiencies of toluene and ethanol were observed to decrease due to such flooding effects of excess supply of buffer solution and was observed to recover the status prior to flooding later. In the early stage of the biofilter-run removal efficiencies of toluene and ethanol maintained almost 100%. However, they began to decrease when inlet load surpassed 90 g/m³/h and 100 g/m³/h consistent with maximum elimination capacity of toluene and ethanol, respectively. At the end of the biofilter-run removal efficiencies for toluene and ethanol were decreased and maintained at 65% and 40%, respectively.

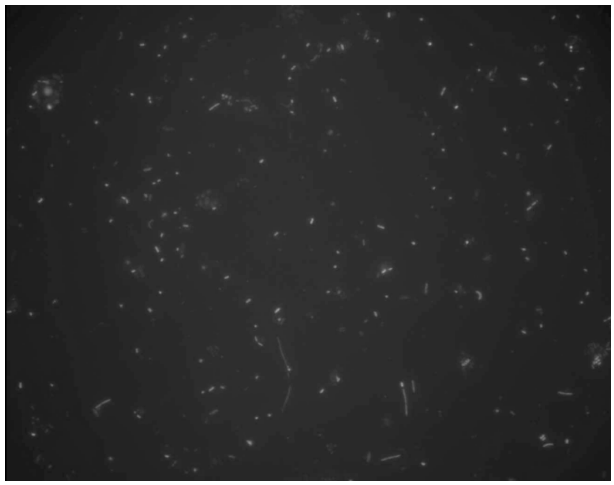

Fig. 12. Microbes observed by fluorescence microscope (16×100) (Axiolab, Xeiss, Germany).

Contrary to the result of Mohseni and Allen [2000], at the 3rd stage of the biofilter run, the effect of hydrophilic ethanol co-feed was observed negligible on transient behavior of hydrophobic toluene concentrations at the 2nd, 3rd and 4th sampling ports, compared to those of the 2nd stage of biofilter run, except for abnormal peak of toluene concentration due to flooding effect. However, saturated concentration of toluene was raised by factor of two at the 1st sampling port at the 3rd stage of biofilter run. According to the result of Mohseni and Allen [2000], it may be interpreted that removal efficiency of hydrophobic toluene was affected by the presence of hydrophilic ethanol when high load of hydrophobic toluene was applied like that of 1st sampling port of biofilter. However, it was not the case when low load of hydrophobic toluene was applied like those of 2nd, 3rd and 4th sampling ports, since hydrophobicity of toluene is much less that of α-pinene.

Thus, it may be suggested that biodegradation of hydrophobic VOC was interfered by hydrophilic VOC dissolved in the biolayer and the degree of interference was proportional to the inlet load of hydrophobic VOC as well as that of hydrophilic VOC and was inversely proportional to the solubility of hydrophobic VOC. However, it was inferred that the existence of hydrophobic toluene from waste-air could hardly inversely affect the removal of hydrophilic ethanol in the biofilter when the time-evolution of hydrophilic ethanol concentrations of this experiment was compared with those of the previous experiments of Lim and Park [2004, 2005] to treat waste-air containing ethanol.

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REFERENCES

Arulneyam, D. and Swaminathan, T., "Biodegradation of Ethanol Vapor in a Biofilter," *Bioproc. Eng.*, **22**, 63 (2000).

Auria, R., Aycaguer, A. C. and Devinny, J. S., "Influence of Water Content on Degradation Rates for Ethanol in Biofiltration," *J. Air Waste Manag. Assoc.*, **48**, 65 (1998).

Bohn, H. L., *Soil Treatment of Organic Waste Gases*, Soils for Management of Organic Wastes and Wastewaters, ASA-CSSA-SSSA, Madison, WI, pp. 607-618 (1977).

Buchner, R., *Auswirkungen Verschiedener Betriebszustande in der Biologischen Abluftreinigung am Beispiel von Biofiltern*, Ph. D. Thesis, T. U. Wien, Austria (1989).

Christine, P., Domenech, F., Michelena, G., Auria, R. and Revah, S., "Biofiltration of Volatile Ethanol using Sugar Cane Bagasse Inoculated with *Candida utilis*," *Journal of Hazardous Materials*, **B89**, 253 (2002).

Deshusses, M. A., Hamer, G. and Dunn, I. J., "Behaviour of Biofilters for Waste Air Biotreatment. 1. Dynamic Model Develpement," *Environ. Sci. Technol.*, **29**, 1048 (1995).

Deshusses, M. A. and Dunn, I. J., *Modelling Experiments on the Kinetics of Mixed-solvent Removal from Waste Gas in a Biofilter*, Proceedings of the 6th European Congress on Biotechnology (L. Alberghina, L. Frontali and P. Sensi eds.), Elsevier Science B. V., 1191-1198 (1994).

Deshusses, M. A. and Hamer, G., "The Removal of Volatile Ketone Mixtures from Air in Biofilters," *Bioprocess Engineering*, **9**, 141 (1993).

Hodge, D. S. and Devinny, J. S., "Biofilter Treatment of Ethanol Vapors," *Environmental Progress*, **13**(3), 167 (1994).

Hodge, D. S. and Devinny, J. S., "Modeling Removal of Air Contaminants by Biofiltration," *Journal of Environmental Engineering*, **121**(1), 21 (1995).

Jorio, H., Kiared, K., Brezenski, R., Leroux, A., Viel, G. and Heitz, M., "Treatment of Air Polluted with High Concentrations of Toluene and Xylene in a Pilot-scale Biofilter," *J. Chem. Technol. Biotechnol.*, **73**, 183 (1998).

Leson, G. and Winer, A. M., "Biofiltration: An Innovative Air Pollution Control Technology for VOC Emissions," *J. Air & Waste Manage. Assoc.*, **41**, 1045 (1991).

Leson, G. and Smith, B. J., "Petroleum Environmental Research Forum Field Study on Biofilters for Control of Volatile Hydrocarbons," *Journal of Environmental Engineering*, **123**(6), 556 (1997).

Lim, K. H. and Lee, E. J., "Biofilter Modeling for Waste Air Treatment: Comparisons of Inherent Characteristics of Biofilter Models," *Korean J. Chem. Eng.*, **20**(2), 315 (2003).

Lim, K. H. and Park, S. W., "The Treatment of Waste-air Containing Mixed Solvent using a Biofilter; 1. Transient Behavior of Biofilter to Treat Wasye-air Containing Ethanol," *Korean J. Chem. Eng.*, **21**(6), 1161 (2004).

Lim, K. H. and Park, S. W., "Treatment of Mixed Solvent with Hybrid System Composed of Biofilter and Photo-catalytic Reactor," *Korean J. Chem. Eng.*, **22**(1), 70 (2005).

Mohseni, M. and Allen, D. G., "Biofiltration of Mixtures of Hydrophilic and Hydrophobic Volatile Organic Compounds," *Chemical Engineering Science*, **55**, 1545 (2000).

Ottengraf, S. P. P., *Exhaust Gas Purification, Biotechnology* (H. J., Rehm, G. Reed, eds.), VCH, Weinheim, Germany, Vol. 8, pp. 426-452 (1986).

Ottengraf, S. P. P. and van den Oever, A. H. C., "Kinetics of Organic Compound Removal from Waste Gases with a Biological Filter," *Biotechnology and Bioengineering*, **25**, 3089 (1983).

Shim, J. S., Jung, J. T., Sofer, S. and Lakhwala, F., "Oxidation of Ethanol Vapors in a Spiral Bioreactor," *J. Chem. Technol. Biotechnol.*, **64**, 49 (1995).

Sorial, G. A., Smith, F. L., Suidan, M. T. and Biswas, P., "Evaluation of Trickle Bed Biofilter Media for Toluene Removal," *Journal of the Air & Waste Management Association*, **45**, 801 (1995).

Swanson, W. J. and Loehr, R. C., "Biofiltration: Fundamentals, Design and Operations Principles, and Applications," *Journal of Environmental Engineering*, **123**(6), 538 (1997).

Tang, B., Hwang, S. J. and Hwang, S., "Dynamics of Toluene Degradation in Biofilters," *Hazardous Waste & Hazardous Materials*, **12**(3), 207 (1995).

William, T. O. and Miller, F. C., "Biofilters and Facility Operations," *Biocycle*, **33**, 75 (1992).

Zarook, S. M. and Baltzis, B. C., "Biofiltration of Toluene Vapor Under Steady-state and Transient Conditions: Theory and Experimental Results," *Chemical Engineering Science*, **49**, 4347 (1994).