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Journal of Hazardous Materials B82 (2001) 233–245

Journal of Hazardous Materials

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Removal of styrene vapor from waste gases by a trickle-bed air biofilter

Chungsying Lu [a,*], Min-Ray Lin [b], Jingchung Lin [a]

[a] *Department of Environmental Engineering, National Chung Hsing University, Taichung 402, Taiwan*
[b] *Department of Mathematics and Science Education, National Taichung Teacher's College, Taichung 403, Taiwan*

Received 6 July 2000; received in revised form 22 August 2000; accepted 4 October 2000

Abstract

The trickle-bed air biofilter (TBAB) performance for the removal of high-strength styrene was evaluated under different gas flow rates and influent concentrations. Under pseudo-steady-state conditions, the elimination capacity increased but the removal efficiency decreased with the increase of styrene loading. More than 90 and 80% removal efficiencies were achieved for influent styrene loadings below 32 and 55 $g/m^3/h$, respectively. The TBAB appears to be an effective treatment process for controlling high-strength styrene emission under low-to-medium loading conditions, and the effectiveness could be maintained over 140 days of laboratory operation.

Keywords: Trickle-bed air biofilter; Styrene; Elimination capacity; Influent concentration; Empty-bed residence time

1. Introduction

Styrene is an important industrial chemical frequently encountered in the production of resins such as acrylonitrile–styrene (AS) or acrylonitrile–butadiene–styrene (ABS) and polymers such as polystyrene. In Taiwan, a large amount of styrene is released into the atmosphere during manufacture, transportation, use, and disposal every year. Losing these toxic pollutants to the ambient air may lead to an adverse environmental impact on the air quality and thus endanger public health and welfare.

More stringent requirements for the removal of volatile organic compounds (VOCs) in recent years necessitate the development of innovative, cost-effective treatment

* Corresponding author. Fax: +886-4-2862-587.
E-mail address: cslu@green.ev.nchu.edu.tw (C. Lu).

0304-3894/01/$ – see front matter
PII: S0304-3894(00)00347-2

alternatives. Traditional VOC control technologies such as carbon adsorption, liquid scrubbing, condensation, thermal incineration, and catalytic incineration have been commonly used to remove VOC vapors from waste gases. However, if the desired VOC removal efficiencies are achieved, these VOC control technologies may suffer from high operating costs and secondary waste stream issues [1].

The biofilter process is a relatively new application that has been proven to be more cost-effective than traditional technologies for treating low-strength and some medium-strength VOC waste gases [2]. The system consists of a filter bed usually comprised of natural organic media such as peat, compost, leaves, wood bark and/or soil. The bed moisture is kept at a constant level by humidification of influent air to maintain a biologically-active layer surrounding the media, known as the "biofilm". VOC-containing airstreams are transported to the air/biofilm interface, where they are absorbed into the biofilm and employed as carbon and/or energy sources by the microorganisms. A trickle-bed air biofilter (TBAB) is a type of biofilter process that employs synthetic, inorganic media and receives liquid nutrients and buffers through a nozzle system on the top of biofilter. The liquid flow rate is usually designed to provide the necessary moisture levels for microbial activity. Due to better control of the pressure drop across the bed, pH and nutrient, TBABs facilitate more consistent operation than do natural media biofilters. Furthermore, they do not suffer from the effects of aging as do natural media [3].

Smith et al. [4] evaluated the performance of two similar TBABs with high toluene loadings between 2.27 and 4.54 kg COD/m^3 per day. They found that a sustained toluene removal efficiency of 99% was achieved, and that the effectiveness could be maintained over 200 days. Rihn et al. [5] evaluated diethyl ether as a substrate for two TBABs and found that greater than 99% removal efficiencies could be obtained at a loading of 3.6 kg COD/m^3 per day. Sorial et al. [6] studied styrene removal from a waste gas by a pilot-scale TBAB. With influent concentrations as high as 160 ppmv (i.e. 160 ppm on a volume basis) and an empty-bed residence time (EBRT) as low as 0.67 min (influent loading = 4.26 kg COD/m^3 per day), removal efficiencies on the level of 94–99% were achieved. Lu et al. [7] evaluated the TBAB performance for treating both single and mixed benzene, toluene, ethylbenzene, and *o*-xylene (BTEX) waste gases. If the single BTEX was employed, the removal efficiencies of 90% could be achieved under the following loadings below: 64 g benzene/m^3/h, 110 g toluene/m^3/h, 53 g ethylbenzene/m^3/h, and 55 g *o*-xylene/m^3/h. If the mixed BTEX was employed, the removal efficiency of each compound could be above 90% under the BTEX loadings of below 96 g/m^3/h. In a parallel TBAB study for acrylonitrile (AN) removal conducted by the same authors [8], the removal efficiency of 95% was achieved for the AN loadings of below 490 g/m^3/h, and the effectiveness could be maintained over 180 days of laboratory operation.

The TBAB has been proven to be an effective process for treating various VOCs from waste gases by the foregoing investigators. However, studies on the biofiltration of gases containing high concentrations of styrene are still unattainable from the literature. The objectives of this research were, therefore, to investigate the TBAB performance for influent gases with styrene concentrations from 150 to 600 ppmv and EBRTs from 1.5 to 6 min. Experimental results obtained herein provide useful information concerning the design criteria and operation of TBAB for controlling high-strength styrene emission.

2. Materials and methods

2.1. Experimental set-up

The bench-scale TBAB was constructed and the experimental set-up for treating styrene waste gas is shown in Fig. 1. The TBAB was made of glass and consisted of six sections. Each section was 15 cm long with an internal diameter of 10 cm. A 10 cm head space was designed for the styrene waste gas inlet and for housing a nutrient spray nozzle, while a 10 cm bottom space was designed for the outlet of treated air and leachate. Leachate was collected at the bottom of the TBAB. Each section of the TBAB was filled with a 1.11 l packing material which supported a thin biological film. The packing material consisted of coal particles with an average pore size of 20.34 μm, a specific surface area of 0.66 m^2/g, and an equivalent-volume diameter of 2.1 cm. The void fraction of TBAB before biofilm attachment was 0.6. The TBAB was placed within a temperature control box (Model CH-502, Chin Hsin, Taipei, Taiwan) to regulate the temperature of styrene waste gas and bioreactor in the range of $25 \pm 2^{\circ}C$.

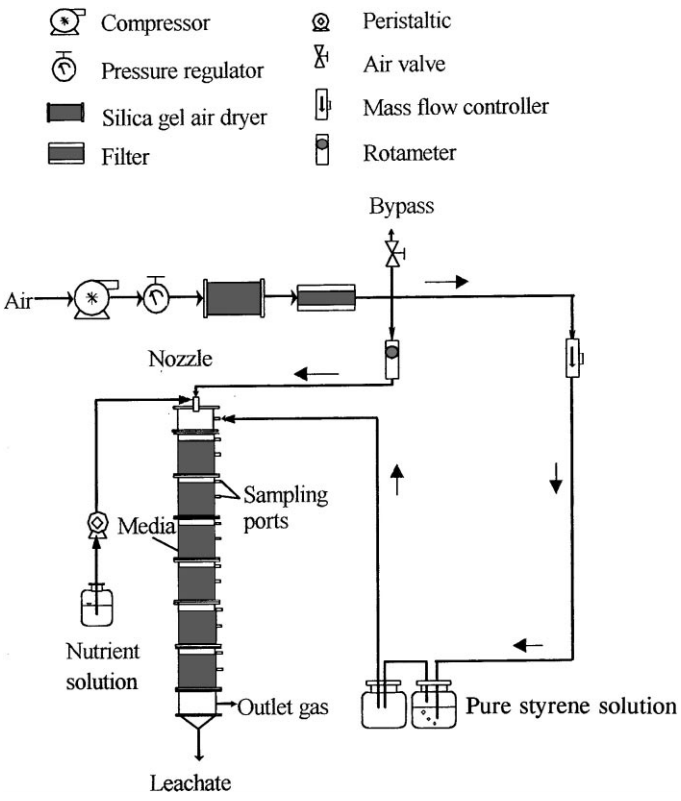

Fig. 1. Schematic diagram of the trickle-bed air biofilter for the removal of styrene from waste gases.

Table 1
Chemical compositions of the nutrient feed[a]

Constituents	Unit	Concentration
KH_2PO_4	g/l	1.19
$Na_2HPO_4{\cdot}12H_2O$	g/l	3.13
KNO_3	g/l	3.88
$(NH_4)_2SO_4$	g/l	2.58
$FeSO_4{\cdot}7H_2O$	g/l	0.28
$MgSO_4{\cdot}7H_2O$	g/l	0.35
$NaHCO_3$	g/l	0.90
$MnSO_4{\cdot}7H_2O$	mg/l	1.52
$Na_2MoO_4{\cdot}2H_2O$	mg/l	1.0
$CaCl_2$	mg/l	3.0
Vitamin B_1	mg/l	10.0

[a] For the case of 150 ppmv styrene.

Compressed air was passed first through a silica gel air dryer to remove moisture and oil and then through a filtration device (Gelman Science, Product No. 12144, Japan) to remove particulate matter. After purification, the major airstream mixed with a 2.4 ml/min nutrient solution was delivered into the headspace of TBAB by a nozzle system. The minor airstream was passed through two glass bottles containing pure styrene solution (99%) and air, respectively, to produce styrene waste gas. The styrene vapor was then mixed with the major airstream in the headspace and passed into the bed with the flows directed downwards. The EBRT was controlled by regulating the major airstream rate using a rotameter, while the influent styrene concentration was controlled by regulating the minor airstream rate using MKS mass flow controllers (model 247C four channel read-out mass flow controller, Andover, MA, USA). The variations of influent styrene concentration were within 10%. The nutrient feed contained inorganic salts and vitamins vital to the growth of attached microorganisms and $NaHCO_3$ as a pH buffer to maintain pH values of 7–8. The compositions of nutrient feed for a 150 ppmv styrene feed are listed in Table 1, while those of other runs were increased according to the carbon mass ratio of influent styrene to that of 150 ppmv styrene.

Microbial seed was used to start the process. Activated sludge which had a density of 1.02 kg/l, suspended solids (SS) of 7000 mg/l, and volatile suspended solids (VSS) of 4700 mg/l was obtained from the secondary clarifier of a wastewater treatment plant in Taichung Industrial Park (Taichung, Taiwan). The suspended solids were allowed to settle for 4 h and the supernatant was discarded to obtain concentrated sludge. The seeding step consisted of mixing 500 l of concentrated sludge with coal particles and 125 g $CaCO_3$. The addition of $CaCO_3$ was used to prevent the acidification inside TBAB. The coal particles with biological attachment were placed into TBAB for about 3 h. The TBAB was then fed with a 150 ppmv styrene, and operated at a 6 min EBRT for a period of 3 weeks to attain the pseudo-steady-state conditions. Pseudo-steady-state conditions were assumed when the daily changes in the styrene removal efficiency were within 10% for three successive days.

2.2. Analytical methods

Styrene concentrations in the air stream were measured using a gas chromatograph (China Chromatography 8900 Series, Taipei, Taiwan) equipped with a flame ionization detector (FID). A 30 m SUPELCO Fused Silica capillary column (0.53 mm i.d.; 1 μm film thickness) was used for all experiments. Sampling ports were located at TBAB inlet and outlet, and between each section of TBAB. A 0.5 l effluent air sample was collected using 1 l teflon bag. One-tenth milliliter of the air sample was taken from this bag using a gas-tight syringe and injected into gas chromatograph. The GC/FID was operated at an injection temperature of 200°C, a detector temperature of 250°C, and an oven temperature of 140°C.

The following parameters were determined as suggested in the standard methods of APHA, AWWA, and WEF [9]: soluble chemical oxygen demand (SCOD, 5220D), and SS (2540D) and VSS (2540G). The pH value of nutrient solution and leachate was measured by a digital pH meter (SUNTEX SP-701). Carbon dioxide and oxygen were determined by precision gas detector tubes (Kitagawa, Model 126SA for CO_2 and Model SA-16 for O_2, Japan). Pressure drops across the bed were measured using an oil-filled manometer (Dwyer Model 400, MI, USA).

A special design tweezers was used to remove coal particles from the TBAB so that the attached biofilm was undisturbed upon coal particle removal. Mass of the attached biofilm per unit volume of coal particle (X_a) was evaluated by drying coal particles before and after biofilm attachment at 80°C for 24 h. The difference between the two measurements divided by volume of coal particle was equal to X_a. The mean X_a of each section was obtained by averaging the measured X_a of six–eight sample tests.

2.3. Carbon balance analysis

The carbon balance of each run was analyzed. For the pseudo-steady-state conditions, the carbon balance in the TBAB can be expressed as

$$Q_a C_i = Q_a C_e + Q_a C_c + \alpha Q_l C_b + \beta Q_l C_l \quad (1)$$

where Q and C represent the flow rate and concentration, respectively, $Q_a C_i$ and $Q_a C_e$ are the influent and effluent carbon rates of waste gas, $Q_a C_c$ is the effluent carbon rate of CO_2 gas; $\alpha Q_l C_b$ is the biomass production rate equivalent to carbon utilization rate; and $\beta Q_l C_l$ is the effluent carbon rate of leachate. Assuming that the net accumulation of attached biomass in the TBAB is equal to zero (i.e. biomass production rate = biofilm detachment rate), the C_b level can be estimated from VSS of leachate. According to Swanson and Loehr [10] the predominant microorganisms in the biofilter decomposing VOCs are heterotrophs such as bacteria and fungi. A typical cellular composition for a heterogeneous microbial population can be represented as $C_5H_7NO_2$ [11]. The conversion factor of biomass to carbon concentration (α) is thus equal to 0.53. The C_l level can be approximated from SCOD of leachate and the conversion factor of SCOD to total organic carbon (TOC), β is equal to 0.3. The carbon recovery (R) is defined as the percentage ratio of the sum on the right-hand size of Eq. (1) to $Q_a C_i$.

Table 2
The operating conditions of each runs[a]

Run no.	EBRT (min)	Q_a (l/min)	C_i (ppmv)	L (g/m^3/h)
1	6.0	1.11	150	6
2	3.0	2.21	150	12
3	1.5	4.43	150	24
4	6.0	1.11	300	12
5	3.0	2.21	300	24
6	1.5	4.43	300	48
7	6.0	1.11	600	24
8	3.0	2.21	600	48
9	1.5	4.43	600	96

[a] EBRT: empty-bed residence time; Q_a: gas flowrate; C_i: influent concentration; L: influent loading.

2.4. Experimental plan

The experimental plan was designed to investigate the TBAB performance for treating high-strength styrene waste gas under different EBRTs (1.5, 3 and 6 min) and influent styrene concentrations (150, 300 and 600 ppmv). The operating conditions of each run are summarized in Table 2, in which the influent styrene loadings ranged from 6 to 96 g/m^3/h. The experiments were conducted by following the order of runs in Table 2 and each run was operated about 2 weeks to achieve the pseudo-steady-state conditions. Upon attainment of pseudo-steady-state conditions, the TBAB was sampled and the results are discussed as follows.

3. Results and discussion

Fig. 2 shows the elimination capacity (removal rate) and removal efficiency of styrene waste gas as a function of influent styrene loading. Symbols represent the experimental data while lines depict the regression results. A regression coefficient of 0.99 was obtained in the analysis. The elimination capacity increased as influent styrene loading increased, but an opposite trend was observed for the removal efficiency. More than 90 and 80% removal efficiencies were achieved for influent styrene loadings below 32 and 55 g/m^3/h, respectively. The styrene emission from the production of resins and polymers in Taiwan may be as high as 300 ppmv. From an operating perspective, for 88% styrene removal efficiency, the EBRT of this TBAB should be $\geq$3 min. If 76% styrene removal efficiency is desired, the EBRT of this TBAB must be $\geq$1 min. As a result, the TBAB appears to be an effective process treating high-strength styrene under low to medium loading conditions.

Fig. 3 shows the styrene concentration profiles against the incremental TBAB length. As can be seen, the styrene concentration profiles were lower at a longer EBRT because the attached microorganisms could take a longer time to decompose styrene in the waste gas. The styrene removal efficiency decreased as the influent styrene concentration increased. It was also seen that the styrene removal efficiency was highest in the first section and gradually decreased in the subsequent sections. The average ratios of styrene removal in

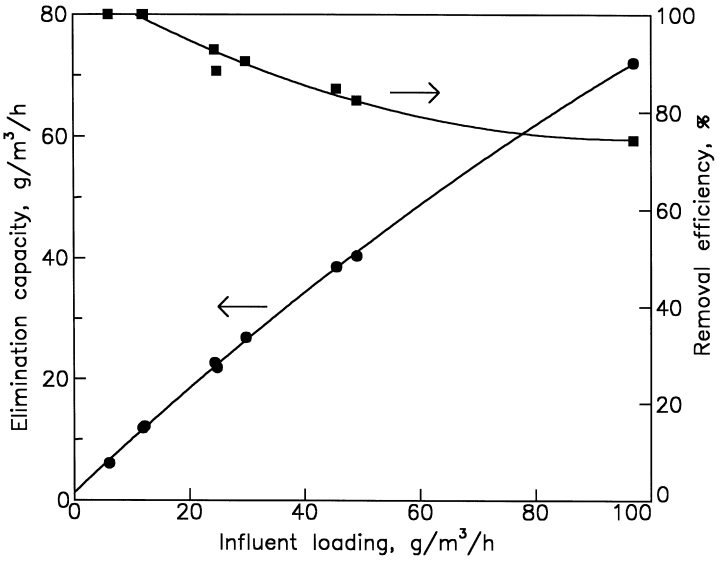

Fig. 2. The elimination capacity and removal efficiency of styrene waste gas as a function of influent loading: (●) elimination capacity; (■) removal efficiency.

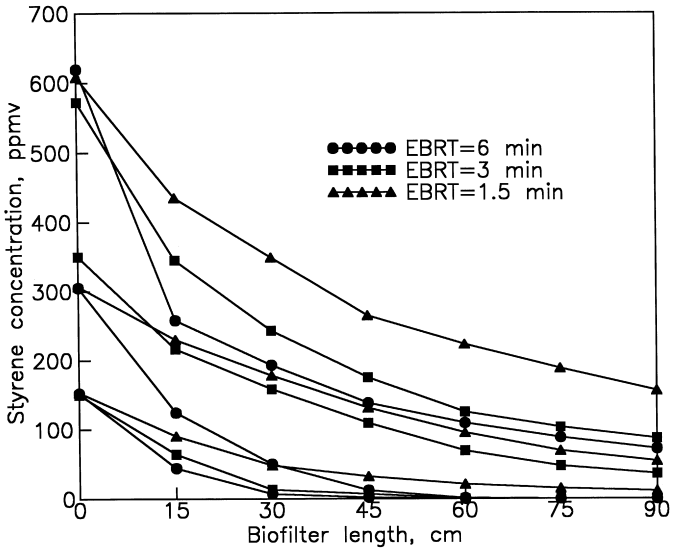

Fig. 3. The concentration profiles of styrene waste gas against the incremental biofilter length.

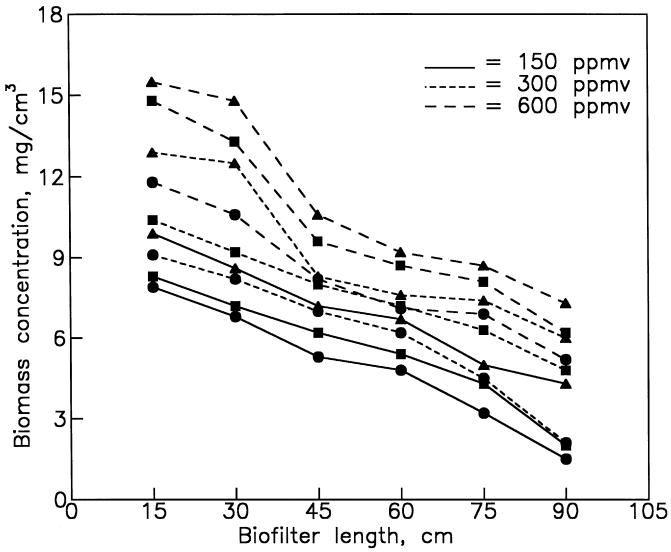

Fig. 4. The test results of masses of the attached biofilm per unit volume of coal particle: (▲) 1.5 min; (■) 3 min; (●) 6 min.

the upper half of TBAB to overall styrene removal were equal to 0.77, 0.85, and 0.94 for EBRTs of 1.5, 3, and 6 min, respectively, indicating that most biological reactions occurred in the earlier sections of TBAB. These results may be due to the fact that more carbon sources, moisture contents and nutrients were present in the earlier sections of the TBAB, which caused a higher metabolic reaction and thus led to a faster biodegradable rate.

The test results of masses of the attached biofilm per unit volume of coal particle (X_a) are shown in Fig. 4. The X_a value increased with an increase of influent styrene concentration or a decrease of EBRT (an increase of influent styrene loading). This was because the biofilm growth was directly related to styrene elimination capacity. As shown in Fig. 2, with a higher influent styrene loading, the elimination capacity was higher resulting in production of more attached microorganisms. The X_a value was highest in the first section and decreased in the subsequent sections, which agreed with the decrease of styrene elimination capacity from section to section as shown in Fig. 3. The X_a values ranged from 1.5 to 15.5 mg/cm^3.

The microbial yield coefficient (Y) is defined as the biomass of cells formed per unit of substrate utilized, which depends on the nature of substrate and species present as well as the environmental conditions of TBAB [11]. The apparent (or observed) Y values of each run are listed in Table 3. As can be seen, Y was much less than unity, indicating that the amount of cells produced in the TBAB is usually much smaller than the amount of styrene removed. The apparent Y value decreased as EBRT decreased or influent styrene concentration increased indicating that the aerobic growth within the TBAB was less efficient under a higher loading condition. In this study, the influent COD loading ranged from 0.45 to 7.14 kg COD/m^3 per day. The resulting Y values ranged from 0.02 to 0.37 with an average value of 0.11 which is close to the typical value of 0.08 for BTEX removal by TBAB systems in the COD loading range of 0.45–6.2 kg COD/m^3 per day [12]. However, the Y values for TBAB systems were

Table 3
The apparent microbial yield coefficients of each run[a]

Run no.	SRR (g per day)	BPR (g per day)	*Y*
1	1.03	0.39	0.38
2	2.03	0.35	0.17
3	3.85	0.27	0.07
4	2.07	0.37	0.18
5	4.57	0.30	0.07
6	6.86	0.24	0.03
7	3.72	0.30	0.08
8	6.56	0.22	0.03
9	12.26	0.25	0.02

[a] SRR: substrate removal rate; BPR: biomass production rate; *Y*: apparent yield coefficient (g-biomass/g-substrate).

much smaller than the typical value of 0.6 for activated sludge systems in the COD loading range of 0.3–3 kg COD/m^3 per day [13]. Sorial et al. [12] offered two possible reasons to explain the differences in the *Y* value between TBAB systems and activated sludge systems. First, the biofilm may contain some entrapped air that may hinder the diffusion of nitrate ions within the biofilm and will eventually retard the growth of microbial cell. Second, the well-controlled experiments may provide a very ideal environment for effective composing the biomass.

The pressure drop across the bed (*P*) plays an important role in determining the amount of energy needed by the blowers to force the VOC contaminated gas through the bed. The *P* value should not be too high since this will result in higher energy requirements [14]. Fig. 5

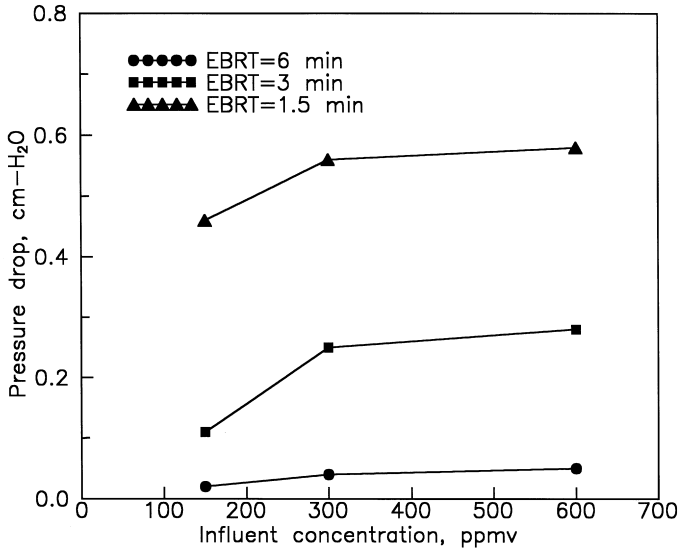

Fig. 5. The pressure drops across the bed under different EBRTs as a function of influent styrene concentration.

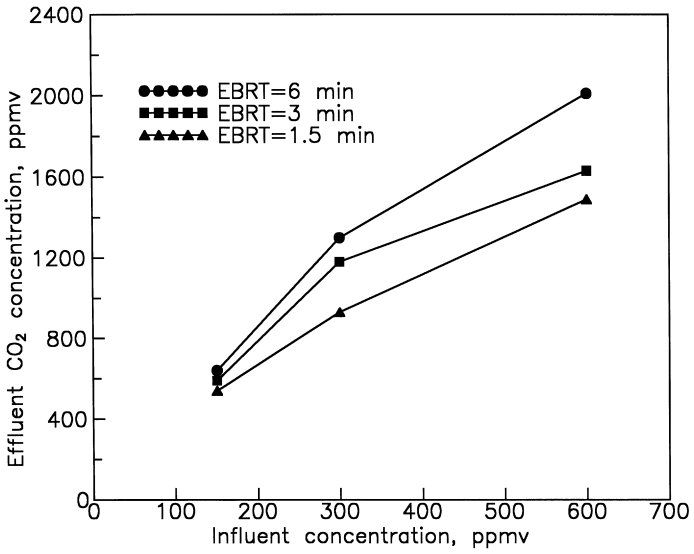

Fig. 6. The CO_2 concentrations of outlet gas under different EBRTs as a function of influent styrene concentration.

shows the test results of *P* value. The *P* value significantly increased as EBRT decreased because the *P* value has a linear relationship with the superficial gas flow rate in a packed filter [15]. The *P* value increased with the increase of influent styrene concentration. This can be attributed to the fact that more microorganisms were produced for a higher influent styrene concentration as shown in Fig. 4, which might have minimized the external porosity of coal particles and thus led to higher pressure drops. The TBAB was operated about 20 weeks from start-up to Run 9. During this period, the pressure drop increased from 0.02 to 0.56 cm of water. These values are lower than those reported for a natural-bed biofilter [16], indicating the TBAB effectiveness for treating high-strength styrene could be maintained over 140 days of laboratory operation.

The CO_2 concentrations of clean air into TBAB were below 30 ppmv which are significantly lower than those of waste gas and can be neglected in the following analysis. Fig. 6 presents the test results of CO_2 concentrations in the outlet gas. The data shows that, at the same EBRTs, the CO_2 concentration significantly increased as the influent styrene concentration increased. This trend was because CO_2 is the end product of aerobic microbial metabolism, therefore, the CO_2 production should be directly related to styrene elimination capacity. As shown in Fig. 3, with a higher influent styrene concentration, the elimination capacity was much higher leading to production of more CO_2. On the other hand, with the same influent styrene concentrations, the CO_2 concentration increased as EBRT increased because more styrene is oxidized at a longer EBRT. The CO_2 concentrations ranged from 540 to 2010 ppmv. The oxygen content of outlet gas was higher at a shorter EBRT or with a lower styrene feed and in the range of 15.6–20.9% meaning that the oxygen content inside TBAB was adequate for metabolic reactions through the study.

The leachate production rate was about 3 l per day. Fig. 7 shows the test results of the soluble chemical oxygen demand (SCOD) in leachate. The SCOD of leachate can be related

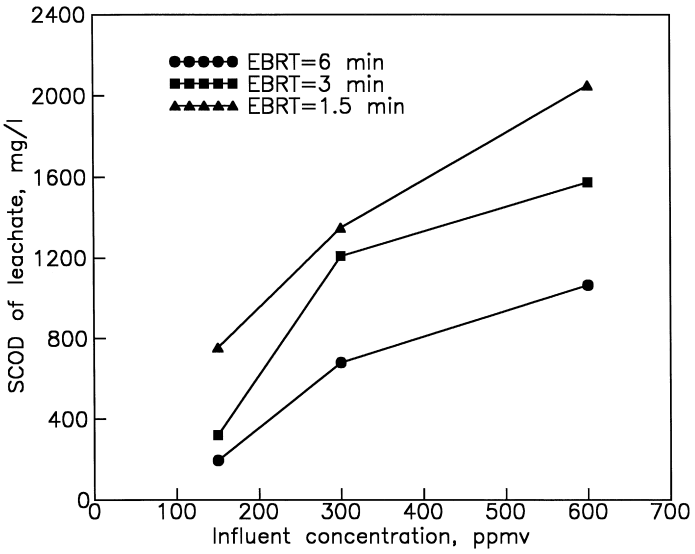

Fig. 7. The SCOD of leachate under different EBRTs as a function of influent styrene concentration.

to the dissolved styrene waste gas. It was seen that the SCOD of leachate increased as an increase of influent styrene concentration or a decrease of EBRT indicating that the dissolved styrene waste gas in leachate increased with the increase of influent styrene loading. The SCOD of leachate ranged from 195 to 2050 mg/l. Fig. 8 shows the test results of SS and VSS

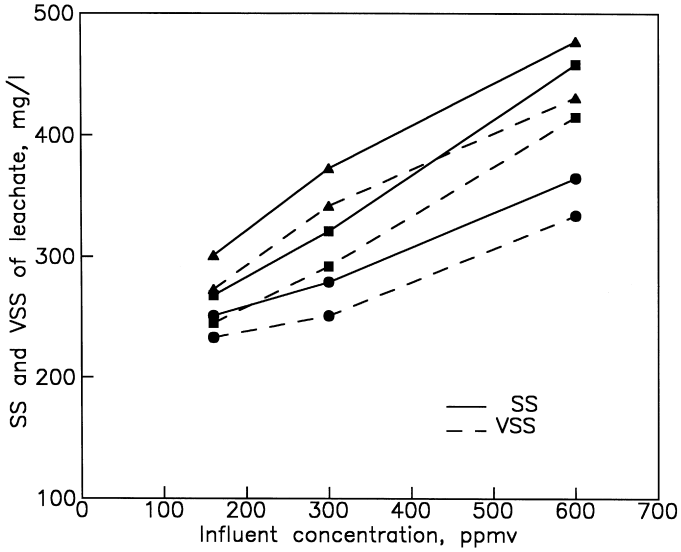

Fig. 8. The SS and VSS of leachate under different EBRTs as a function of influent styrene concentration: (▲) 1.5 min; (■) 3 min; (●) 6 min.

Table 4
The carbon balance analysis of each run[a]

Run no.	Q_aC_i (mg C/min)	Q_aC_e (mg C/min)	Q_aC_c (mg C/min)	αQ_lC_b (mg C/min)	βQ_lC_l (mg C/min)	R (%)
1	0.661	0	0.348	0.143	0.053	82
2	1.3	0	0.639	0.129	0.086	66
3	2.66	0.191	1.17	0.135	0.204	64
4	1.33	0	0.707	0.101	0.183	75
5	3.24	0.312	1.28	0.110	0.326	63
6	5.33	0.937	2.02	0.112	0.364	64
7	2.69	0.313	1.09	0.089	0.287	66
8	4.95	0.753	1.78	0.082	0.435	62
9	10.6	2.71	3.23	0.090	0.553	62

[a] Q_aC_i: influent carbon rate of waste gas; Q_aC_e: effluent carbon rate of waste gas; Q_aC_c: effluent carbon rate of CO_2 gas; αQ_lC_b: biomass production rate equivalent to carbon utilization rate; βQ_lC_l: effluent carbon rate of leachate; R: carbon recovery.

in leachate. The VSS of leachate can be related to the sloughed biofilm. As can be seen, the SS and VSS also increased with the increase of influent styrene loading. The mass ratios of VSS to SS were about 0.9 indicating that most SS in the leachate were from sloughed biofilm. The SS and VSS of leachate ranged from 251 to 477 mg/l and from 235 to 431 mg/l, respectively.

The analytic results of carbon balance are listed in Table 4. The heterogeneous microorganisms in the TBAB are unable to utilize the oxidation of inorganic chemicals for their energy and carbon requirements. Therefore, $NaHCO_3$ which was added into TBAB as a pH buffer was not included in the following analysis. As can be seen in Table 4, the carbon recoveries (R) were in the range of 62–82%. There are three possible reasons not included in the analysis that could explain the differences between influent and effluent carbon rate. First, some intermediate products were present in the effluent waste gas. Second, some of the produced CO_2 was dissolved into leachate. Third, some detachments of aged biofilm were accumulated in the TBAB. These three reasons may lead to the underestimate of effluent carbon rate in the effluent waste gas (C_e), effluent CO_2 gas concentration (C_c), and biofilm growth quantity (C_b). The carbon balance analysis also showed that most of the carbon in the effluent was from effluent CO_2 gas and sloughed biofilm for low influent loadings and from effluent waste gas and CO_2 gas for high influent loadings. This trend indicates that the dissolved styrene and its derivatives in leachate were not significant in the TBAB. The carbon recoveries were higher at a longer EBRT or for a lower styrene concentration feed. This may be attributed to the fact that the above-mentioned departures were not significant under low influent loading conditions.

4. Conclusions

The following conclusions could be drawn from the results presented in this study:

1. More than 90 and 80% removal efficiencies were achieved for influent styrene loadings below 32 and 55 g/m^3/h, respectively, indicating the TBAB appears to be an effective

process for treating high-strength styrene waste gas under low to medium loading conditions.
2. Most styrene removals occurred in the earlier sections of the TBAB.
3. The apparent microbial yield coefficients indicated the amount of cells produced in the TBAB were much less than the amount of styrene removed, and the aerobic growth within the TBAB is less efficient under a higher loading condition.
4. The biomass concentration (X_a), system pressure drop (P), and SCOD, SS and VSS of leachate increased with an increase of influent styrene loading.
5. Pressure drops across the bed ranged from 0.02 to 0.56 cm of water which were relatively lower than those reported for a natural-bed biofilter.
6. From carbon balance analysis, most effluent carbons were from effluent CO_2 gas and SS of leachate for a lower influent loading and from effluent styrene waste gas and CO_2 gas for a higher loading condition.

Acknowledgements

Support from the National Science Council, Taiwan (NSC 87-2218-E-005-001) is gratefully acknowledged.

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