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Short Communication

Biodegradation of styrene laden waste gas stream using a compost-based biofilter

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Abstract

Biological treatment of waste gas styrene vapor was investigated in a three-stage bench-scale biofilter. Yard waste compost mixed with shredded hard plastics in a 25:75 v/v ratio of plastics:compost was inoculated with thickened municipal activated sludge. Microbial acclimation to styrene was achieved by exposing the system to an inlet concentration (C_{In}) of 0.25 g m^{-3} styrene and an empty bed retention time (EBRT) of 360 s for 30 days. Under steady-state conditions, maximum elimination capacity (EC) obtained was 45 g m^{-3} h^{-1} at a loading rate (L) of 60 g m^{-3} h^{-1} (C_{In} of 2 g m^{-3} and EBRT of 120 s). Reduction of retention time adversely impacted the performance resulting in the maximum EC of 39 and 27 g m^{-3} h^{-1} for EBRT of 60 and 30 s, respectively. Evaluation of the concentration profile along the bed height indicated dominance of first-order kinetics at $C_{In} \leqslant 0.45$ g m^{-3} and zero-order for higher concentrations.

Keywords: Biofiltration; Styrene; Waste gas; Biological treatment; Compost; VOC

1. Introduction

Volatile organic compounds (VOCs) are emitted into the atmosphere in large quantities from chemical and petrochemical industries. Styrene is included in this category and is regarded as toxic under Title III of US 1990 Clean Air Act Amendments. It is released into the environment during the manufacturing and application of its isomers including polystyrene, styrene–butadiene rubber, acrylonitrile–butadiene–styrene and copolymers resins. Natural sources include microbial and fungal metabolism (USEPA, 1999). Urban concentrations vary from city to city but are reported to be up to 15 ppb in some US cities (Howard, 1989). Acute exposure to styrene in humans results in mucous membrane and eye irritations as well as gastrointestinal disorders. Chronic contact causes a variety of discomforts including headache, fatigue, weakness, depression, and peripheral neuropathy (USEPA, 1999).

Biofiltration is a cost effective and reliable option in treating VOCs emitted from processes with large offgas volumes but low concentrations (Ottengraf et al., 1986). Biofilters are more effective in treating some

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hydrophobic compounds in comparison to other biological systems such as bioscrubbers and biotrickling filters (Van Groenestijn and Hesselink, 1993). Simplicity of design and ease of operation has also been cited as the reasons for popularity of biofilters (Zilli et al., 2001).

Biofiltration of styrene laden waste gas streams has been studied by a number of researches. The media used has included natural residues like peat (Arnold et al., 1997; Zilli et al., 2001), perlite (Paca et al., 2001) and specially preconditioned biomass pellets (Jorio et al., 2000). However, compost-based media has rarely been used as a packing material for biofiltration of styrene. Results show that mixed microbial cultures have better removal performance than pure cultures in the degradation of styrene (Arnold et al., 1997) but enriched styrene-degrading bacteria from activated sludge have also been tried as the initial seed for biofilters and biotrickling filters (Arnold et al., 1997; Chou and Hsiao, 1998; Pol et al., 1998; Juneson et al., 2001). The additional expense levied by enrichment can be compensated by shorter acclimation periods.

Filter media play an important role in the performance and steady operation of biofilters. Quality indicators include porosity, surface area, pressure drop and cost. Compost has been widely used for biofilter media because of its low cost and ease of availability. Negative aspects include the development of back pressure due to gradual compaction with time and aging effects due to microbial mineralization. As such, synthetic materials such as polystyrene spheres, perlite and glass have gained more popularity to offset excess compaction (Abumaizer et al., 1997, 1998).

In this bench-scale study, municipal activated sludge was used to cultivate compost media without any enrichment process. The reactor was packed with yard waste compost-based media mixed with shredded hard plastics as the bulking agent. The objectives of this study were (a) estimation the acclimation period of biofilter for degradation of styrene inoculated by activated sludge; (b) evaluation of the variation of operational parameters on system performance.

2. Materials and methods

2.1. Experimental set-up

A three-stage downward flow bench-scale biofilter constructed from galvanized iron was used in this study (Fig. 1). The column had an inner diameter of 8 cm with

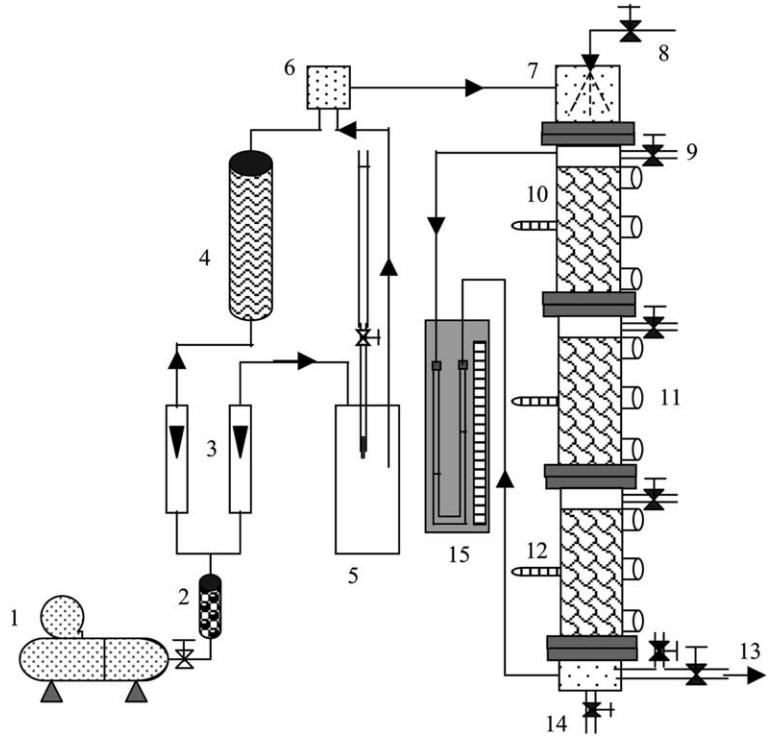

Fig. 1. Schematics of biofilter system (1—compressor, 2—carbon filter, 3—rotameter, 4—humidifier, 5—pollutant vessel with styrene injector, 6—mixing chamber, 7—inlet, 8—nutrient, 9—gas sampling port, 10—biofilter bed, 11—bed sampling port, 12—thermometer, 13—outlet, 14—leachate, 15—manometer).

an effective bed height of 120 cm. Perforated steel plate plenums (pore diameter = 2 mm) placed between sections acted as a support for the packing material as well as for gas flow redistribution. A 7-cm space in between the sections allowed for representative gas sampling. Provision of sampling ports at the top, midpoint and the end of each section allowed bed media access. The synthetic waste gas stream was prepared by passing compressed air through a granular activated carbon canister to retain residual oil and particles. The air stream was then sparged through a 15 l water container equipped with heated element for adjusting gas stream temperature and humidification. Pollutant vapor was prepared by introducing low flow air stream into a container receiving drop-wise styrene feed from a burette. After humidification, the main air stream was mixed with the stream containing pollutant vapor to generate feed air with the needed concentration. Changing water temperature in the humidifier allowed humidity control of influent gas stream and biofilter material. Temperature control of the bed material to 30 ± 1 °C was achieved by using a heated tape wrapped around the exterior of the reactor wall. Bed water content was maintained at 60–65% during the study period.

Bed media was prepared by mixing yard waste compost (Gol & Giah Compost Co., Tehran) and shredded high-density plastics (1.5×1.0 cm) as bulking agent to produce a 25:75 v/v ratio of compost-bulk agent with the porosity and density of 54% and 0.611, respectively. In preparing the packing medium, thickened activated sludge obtained from municipal wastewater treatment plant (Tehran Water & Wastewater Co.) was added to this mixture to increase microbial density and improve homogeneity of compost particles and bulking agents. Nutrient and buffering solution was used according to the quantity of inlet carbon to keep the C:N:P ratio around 100:5:1. The nutrient solution had the following composition (per liter of tap water): 0.694 g KH_2PO_4, 0.854 g K_2HPO_4, 1.234 g $(NH_4)_2SO_4$, 0.46 g $MgSO_4 \cdot H_2O$, 0.176 g $CaCl_2 \cdot 2H_2O$, 0.001 g $FeSO_4 \cdot 7H_2O$, and 5 ml trace element solution consisting of 60 mg l^{-1} H_3BO_3, 40 mg l^{-1} $CoCl_2 \cdot 6H_2O$, 20 mg l^{-1} $ZnSO_4 \cdot 7H_2O$, 6 mg l^{-1} $MnCl_2 \cdot 4H_2O$, 6 mg l^{-1} $NaMoO_4 \cdot 2H_2O$, 4 mg l^{-1} $NiCl_2 \cdot 6H_2O$, 2 mg l^{-1} $CuCl_2 \cdot 2H_2O$ with an overall pH around 6.9 ± 2 (Ergas et al., 1994).

2.2. Experimental schedule

Performance of biofilter in treatment of styrene was evaluated for more than a year by varying airflow rate and styrene inlet concentrations. During the start-up of biofilter which lasted 65 days, the inlet concentration was kept low to allow gradual microbial adaptation. Upon the establishment of pseudo-steady-state operation, different parameters were varied to evaluate the performance. The superficial gas velocity was increased to 35.8, 71.2 and 143.2 m h^{-1} with empty bed retention times (EBRTs) of 120, 60 and 30 s and styrene inlet concentration was varied between 0.14 and 2.54 g m^{-3}. Each of the experimental sets lasted for about a month.

2.3. Analytical methods

Gaseous samples were collected in 5 l Tedlar bags by connecting the bag port onto the tube connected to the biofilter sampling ports. The bags were flushed with charcoal-filtered air prior to sampling. Analyses were done within 3 h using a gas chromatograph (SRI 110 Inc.) equipped with a flame ionization detector and a 30 m stainless steel capillary column (GM 6210). Operating conditions were as follows: temperature schedule (injector 200 °C, oven 220 °C, detector 230 °C) and N_2 carrier gas at 8 ml min^{-1}. Gas samples of 1 ml containing styrene were withdrawn from Tedlar bags using 2 ml gas-tight syringe (Series A-2, VICI, Inc.) and were injected immediately into the GC unit for concentration determination. Inlet and outlet concentrations reported represent an average of two sequential samples.

3. Results and discussion

3.1. Start-up

After inoculation of the biofilter with thickened municipal activated sludge, the system was started with low airflow rate (Q) of 0.06 m^3 h^{-1} corresponding to an EBRT of 360 s with an inlet styrene concentration of 0.25 g m^{-3}. Gradual increase in removal efficiency was observed (Fig. 2) reaching to about 79% on day 20 and almost 100% after about 2 months. The results are consistent with the reported acclimation periods of several weeks to several months (Juneson et al., 2001). Inoculation of the biofilter media with adapted microbial aggregates greatly reduces the acclimation time of biofilter (Shareefdeen and Baltzic, 1994; Acuna et al., 1999; Jorio et al., 2000) to as low as 12 days (Arnold et al., 1997). Experiments conducted with pasteurized media under similar conditions confirmed styrene biodegradation.

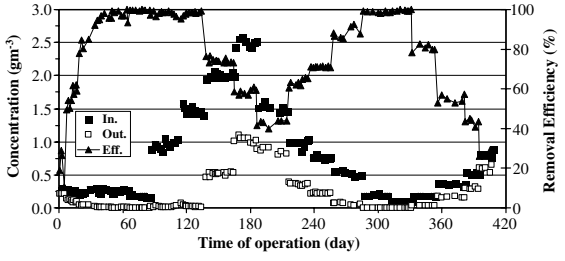

Fig. 2. Overall performance of compost-based biofilter in the removal of styrene.

No styrene removal was observed for 30 days of continuous monitoring.

Steady-state conditions were observed after day 35 of the operation. As shown in Fig. 3a, the majority of the removal occurred in the first two sections of the biofilter with section 3 contributing <20% by day 35 and <10% by day 57.

3.2. Vertical concentration gradient

Evaluation of styrene concentration gradient along the bed height showed steady decline with increasing distance from the inlet (Fig. 3b and c). At low C_{In} of 0.2 g m^{-3}, the profiles show an exponential decrease for all EBRTs, an indication of a first-order kinetics with respect to styrene concentration. This is consistent with the results obtained by other researchers for this range of concentrations for biofilter (Ottengraf et al., 1986) and biotrickling filters (Loy and Flauger, 1996). At higher inlet concentration of 1 g m^{-3}, there was an apparent shift to zero-order kinetics. Similar results have been reported by some researchers (Ottengraf et al., 1986) while others report a much lower inlet concentration of 0.1 g m^{-3} for the onset of zero-order kinetics (Jorio et al., 2003; Zilli et al., 2004). The apparent differences may be attributed in part to the variations of microbial dynamics in different sections and the corresponding specific activities and metabolic pathways utilized by the dominant strains involved in the degradation of styrene. Degree of homogeneity of microbial density can also be a factor as it affects the availability and/or deficiency of substrate for degradation.

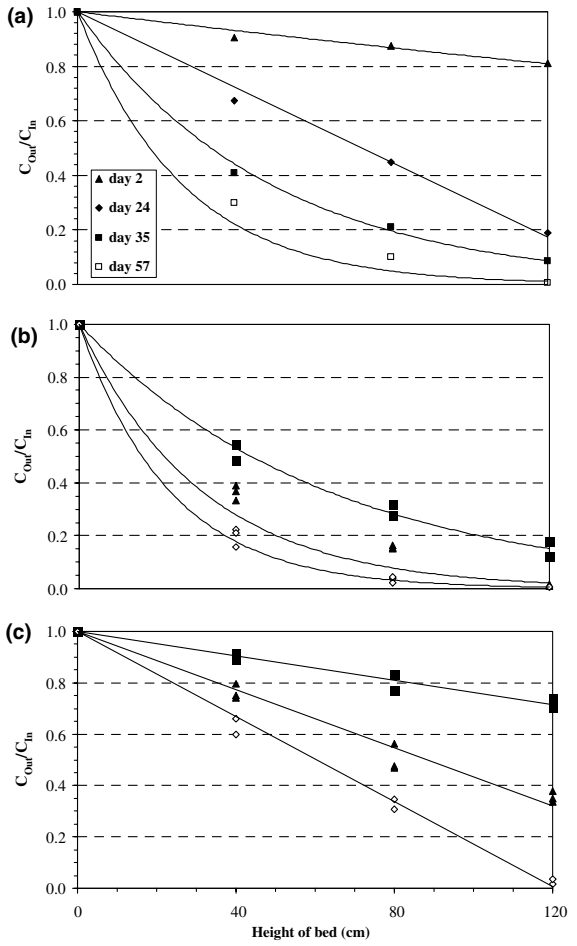

Fig. 3. Styrene concentration profile along the bed height during the start-up period with C_{In} of 0.25 g m^{-3} and EBRT of 360 s (a) and at steady-state condition with EBRT of 30 (■), 60 (▲) and 120 (◇) with C_{In} of 0.2 (b) and 1 g m^{-3} (c).

3.3. Kinetics

In general, for biofiltration systems, oxygen concentration required for the aerobic metabolism of the microorganisms is not limiting the utilization rate of compound by the microbial flora as modeled by Michaelis–Menten type kinetic relationship. As such, the rate of biodegradation or elimination capacity (EC) can be described by the following modified equation:

$$\mathrm{EC} = \frac{V_{max} \times C_{In}}{K_m + C_{In}}$$

where V_{max} is the maximum biodegradation rate (g m^{-3} h^{-1}) and K_m is the saturation (Michaelis–Menten) constant (g m^{-3}). Depending on the concentration of substrate, the rate can be first-order ($C_{In} \ll K_m$) or zero-order reaction ($C_{In} \gg K_m$). The results shown in Fig. 4 illustrate inverse relationship between EC and EBRT indicating limitations of diffusion rate of styrene into the biomass (Jorio et al., 2003). Zero-order kinetics seems to prevail at inlet concentrations above 1.55 g m^{-3} which are consistent with results suggested in the modeling studies of other hydrophobic and hydrophilic VOCs (Ottengraf et al., 1986).

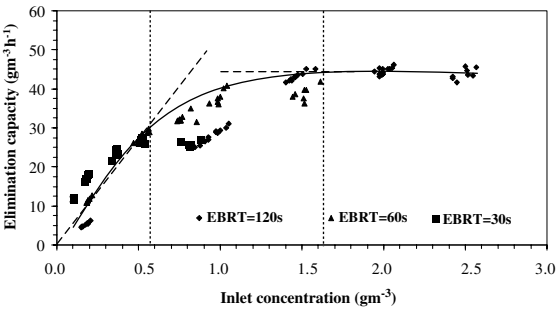

Fig. 4. Evaluation of elimination capacity of biofiltration of styrene at different inlet concentrations.

Table 1
Summary of experimental results of styrene biofiltration

EBRT (s)	C_{In} (g m^{-3})	L (g m^{-3} h^{-1})	RE (%)
120	0.4		99
60	0.2	<12	99
30	0.1		99
120	1.0		98
60	0.5	12–30	88
30	0.25		81
120	1.5		98
60	0.75	30–45	71
30	0.37		55
120	2.0		74
60	1.0	45–60	63
30	0.5		43

3.4. Retention time and loading rate

The effect of retention time on biofilter performance at organic loading rates (L) of 12–60 g m^{-3} h^{-1} was studied by varying EBRT from 30 to 120 s and a summary of results is shown in Table 1. As shown in Table 1, not much was gained as a result of increase in EBRT from 30 to 120 s at $L < 12$ g m^{-3} h^{-1}, but considerable change from RE of 98–81% was observed with further decrease to 30 s at organic loads between 12 and 30 g m^{-3} h^{-1}. Further increase in inlet load above 30 g m^{-3} h^{-1} resulted in proportional decrease in performance.

Fig. 5 shows that at both EBRT of 60 and 120 s for $L < 30$ g m^{-3} h^{-1}, elimination capacity remains very close to 100% removal rate (dotted line in the figure). Further reduction of EBRT to 30 s resulted in an onset of deviation from 100% removal rate at a lower inlet load. Maximum values of EC obtained were 45, 39 and 27 g m^{-3} h^{-1} at EBRT of values of 120, 60 and 30 s, respectively. The values are close to reported maximum EC of 50 g m^{-3} h^{-1} for styrene with a novel biomass as filter bed at EBRT of 127 s (Jorio et al., 2000) but lower than the value of 63 g m^{-3} h^{-1} at EBRT of 30 s reported for peat biofilter inoculated by cultivated bacteria (Zilli et al., 2003). Back pressure along biofilter column did not altered with increasing organic loading rates and was measured with lower variations at higher flow rates due to high porosity of mixture of bed material. The maximum pressure drop of about 20 mm H_2O was observed across biofilter bed.

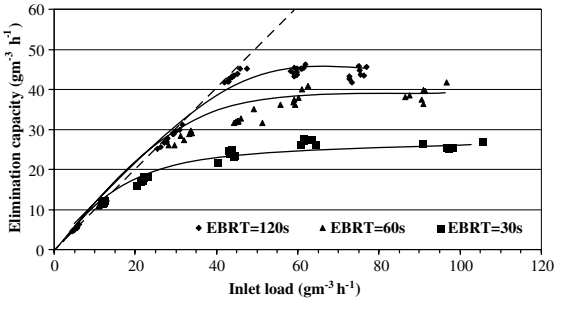

Fig. 5. Effect of styrene inlet load on EC of biofilter at different hydraulic detention times.

4. Conclusions

- Acclimation period of less than 30 days for biodegradation of styrene in biofilters is possible for non-cultivated microbial flora as reported by other researchers.
- EC of biofilter increased directly with $C_{In} \leqslant 0.45$ g m^{-3} at all of the EBRTs, although increasing the gas flow rate resulted better performance. Also respect to inlet concentration, biofilter shows zero-order kinetics with comparison of the data with Ottengraf's model.
- Zero-order kinetics seems to prevail at inlet concentrations above 1.55 g m^{-3}.
- Maximum EC of 45 g m^{-3} h^{-1} was obtained at EBRT of 120 s and C_{In} of 2 g m^{-3} with an average RE of 74%. For EBRT of 60 s at $C_{In} \leqslant 0.53$ g m^{-3}, RE was about 88% resulting in an effluent concentration (C_{Out}) of 0.063 g m^{-3} (15 ppm) which is lower than the than the ACGIH threshold limit value (0.085 g m^{-3}) expressed as a time-weighted average.

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