Research Article

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Effect of surfactant on styrene removal from waste gas streams in biotrickling filters

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Abstract

BACKGROUND: The performance of two biotrickling filters (BTFs) was evaluated for styrene removal from gas streams at the start-up period and at a pseudo-steady-state under various operating conditions.

RESULTS: The BTFs exceeded 99% removal efficiency within 19 days when the average inlet styrene concentration was 250 mg m^{-3} and gas empty bed retention time (EBCT) was 30.0 s. The effect of a surfactant, Triton X-100, on styrene removal was examined by comparative experiments in which one biofilter was fed with nutrient solution with the surfactant while the other without the surfactant, and the average organic loading rate of styrene was set at 65.3, 100.9 and 201.7 g styrene m^{-3} h^{-1}, respectively. Results showed that the corresponding average removal efficiency was 87%, 70% and 50% for the BTF without surfactant, while the average removal efficiency for the BTF with surfactant was 96%, 92% and 82%. Excessive biomass accumulation was observed in the medium when the styrene loading rate was high. However, the biomass density within the BTF medium when the surfactant was added remained stable during the whole period of the operation.

CONCLUSION: These results demonstrated that the use of Triton X-100 can improve the degradation of styrene and control the excessive biomass accumulation.

Keywords: biofilms; biofiltration of waste gases; biomass; bioreactors; volatile organic compounds; surfactant

INTRODUCTION

Styrene is commonly used as a starting material for synthetic polymers and a solvent in many chemical industries.[1] It is reported to have a significant adverse effect on human health because of its toxicity.[2] Styrene in waste gas is difficult to remove by typical wet-scrubbing processes due to its low water solubility and high vapor pressure.

Compared with the traditional physical and chemical waste gas treatment technologies, biological technology utilizes microbial metabolic reactions to convert the waste gas into carbon dioxide, water vapor, and organic biomass, so it is commonly viewed as a cost effective option to treat contaminants with large off-gas volumes but low concentrations.[3] What is used commonly in biotechnologies are biofilters, biotrickling filters (BTF) and bioscrubbers, while innovative biofilters including rotating drum biofilters and tubular biofilters have been developed recently.[4–8] In biotrickling filters, microorganisms attaches on the surface of filter-bed media as biofilms. When a gas stream passes through the medium, the target pollutants in the air phase were adsorbed into the biofilm where they are biodegraded into non-polluting products.[3] A typical BTF is relatively simple, with the bio-phase static and the liquid-phase moving. This allows for microorganisms whose growth cycle is long to also survive in the filter. When the concentration of target contaminants is high, the removal efficiency of BTFs is usually low because excessive biomass accumulation and consequent clogging and channeling result during long periods of operation. Although some biomass control methods have been reported, such as periodic backwashing and starvation in operation,[9,10] both methods need time for re-acclimation, which is time-consuming.

In order to improve the removal efficiency of BTFs at high organic loading rates, a nonionic surfactant, Triton X-100, was used. Surfactants are frequently used for remediation of hydrophobic organic compounds in contaminated soils and sediments.[11–13] Surfactants allow the insoluble matter to emulsify, wet, disperse, and dissolve thereby lowering the surface tension and the interfacial tension.[14] Non-ionic surfactants are known to be less toxic and could serve the dual purposes of increasing solubility of organic pollutants and limiting excess biomass growth.[15] Surfactants can improve toluene removal efficiency in bioreactors.[16] The effects of Triton-X-100 on styrene removal efficiency and biomass accumulation are rather interesting. In this

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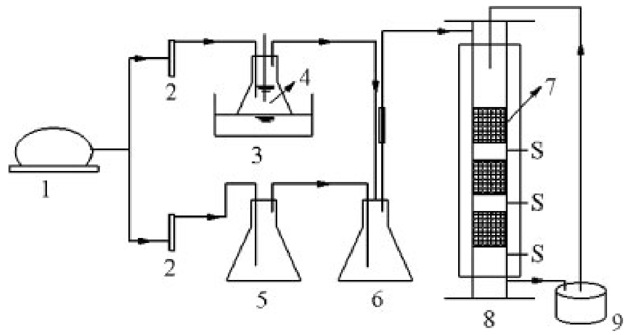

Figure 1. Schematic diagram of the biotrickling filter. (1) Air compressor; (2) flow meter; (3) water-bath device; (4) styrene; (5) glass humidifier; (6) mixed bottle; (7) polyurethane sponge medium; (8) biotrickling filter; (9) nutrient solution.

study, the biofiltration of styrene vapors was investigated using two separate but identical biotrickling filters, BTF1 and BTF2. After both BTFs were successfully started, we added Triton X-100 to the nutrient solution which was fed to BTF2, while all other operation conditions of the two BTFs remained same.

MATERIALS AND METHODS

Experimental apparatus

Two identical biofilters, BTF1 and BTF2, were used to remove styrene from waste gas streams in this study. Both BTFs consisted of a cylindrical column with an inner diameter of 10 cm and a height of 80 cm in which three similar cylindrical medium serums with a height of 10 cm were packed. A gas distribution system and a nutrient solution supply system were used for each of the BTFs. The schematic diagram of the systems is shown in Fig. 1.

Seeding microorganisms

The activated sludge taken from the oxidation ditch of Jinxia Municipal Wastewater Treatment Plant was used for seeding the BTFs. Any enrichment procedures for the sludge were not carried out.

Medium composition

Filter media have a major impact on bacteria activity and pressure drop across the reactor.[17] Porosity, surface area, cost, and mechanical properties of media are important parameters. Many types of media have been used in laboratory-scale biofilters, including activated carbon,[18] compost[19,20] and perlite.[21,22] Polyurethane sponge is considered to be a highly suitable packing material for biofiltration.[23] Serums of reticulated polyurethane sponge (10 pores per inch) were used as the medium in this study, which has a large surface area, good permeability, and high compressive strength.

Nutrient supply

During the whole experimental period, the nutrient and buffering solution was used to supply various nutrient elements to microbes and to prevent the pH value from changing significantly in the medium bed. The composition of the solution per liter of tap water was as follows: 8.40 g K_2HPO_4, 2.80 g KH_2PO_4, 1.32 g $MgSO_4$, 150.00 g NH_4Cl, 7.20 g $NaHCO_3$, 0.03 g $FeCl_3$, 0.80 g $CaCl_2$, 0.02 g $CuCl_2{\cdot}4H_2O$, 0.04 g $MnCl_2{\cdot}4H_2O$, 0.26 g $CoCl_2{\cdot}6H_2O$, 0.0002 g folic acid, 0.00069 g nicotinic acid. A liquid spraying device was installed at the top of each BTF, and biofilter leachate was circulated and sprayed over the medium bed periodically to provide nutrients and water for the microbes.

Analytical methods

Styrene concentration in gas samples was measured using an HP 5890 gas chromatograph (HP5890, Series II, Hewlett-Packard, Palo Alto, California), using a capillary column HP-VOC (60 m× 320 µm ID× 1.8 µm). The flow rates were 30 mL min^{-1} for H_2 and 350 mL min^{-1} for air, and N_2 was used as the carrier gas at a flow rate of 30 mL min^{-1}. The temperature at the GC injection, oven and detection ports were 120 °C, 120 °C and 250 °C, respectively. The pressure drop was measured using a U-tube water manometer connected to the top of the BTF, with an operation range 0–5 kPa (0–50 cm H_2O).

Experimental plan

The experimental plan was designed to investigate the performance of BTFs treating styrene waste gas under different organic loading rates (65.3, 100.9 and 201.7 g styrene m^{-3} h^{-1}, different EBCTs (30.0, 15.0 and 7.5 s), and different gas flow rates (140, 280 and 560 L h^{-1}) in the presence or absence of nonionic surfactant. To ensure the biofilters were at pseudo-steady-state so that the experimental results under the different operating parameters were comparable, the BTFs were operated in the reference condition with an inlet styrene concentration of 250 mg m^{-3} and EBCT of 30 s before and after changing the value of an operating parameter.

RESULTS AND DISCUSSION

Start up

The performance of the BTFs was evaluated in terms of removal efficiency (RE) and elimination capacity (EC).

Both BTFs were initially acclimated to styrene by passing low concentration of 250 mg m^{-3} and low gas flow rate of 280 L h^{-1} for 19 days to obtain sufficient biomass in the filter bed, corresponding to an EBCT of 30.0 s. Although visible biomass growth was seen on the surface of the medium from the first day, the removal efficiency of both BTF1 and BTF2 were very low during the first 5 days. The removal efficiency of BTF1 reached 90% on day 7 and BTF2 on day 8. The RE of both BTF1 and BTF2 reached 99% on day 18. Rene *et al.*[24] reported that the styrene removal efficiency reached 90% on day 17 in a fungal monolith bioreactor. During the 19 days of the start-up period, the average styrene loading was maintained at 29.7 g m^{-3} h^{-1}, and the pressure drop in both BTF1 and BTF2 was less than 30 Pa for the whole bed height. The start-up performance of the two BTFs is shown in Fig. 2.

Effect of styrene inlet concentration on gaseous styrene removal

After the two BTFs were successfully started-up, we added 1 mL Triton X-100 to 15 L nutrient solution, which was fed to BTF2. The flow rate fixed at 280 L h^{-1} and EBCT fixed at 30.0 s. The average styrene concentration was increased from 250 mg m^{-3} to 550, 850 and 1700 mg m^{-3} for both BTF1 and BTF2. The BTFs were run in the reference conditions (inlet styrene concentration of 250 mg m^{-3} and EBCT 30.0 s) for recovery experiments before once again changing the concentration. The removal efficiency of BTF1 and BTF2 at different organic loading rates is presented in Fig. 3.

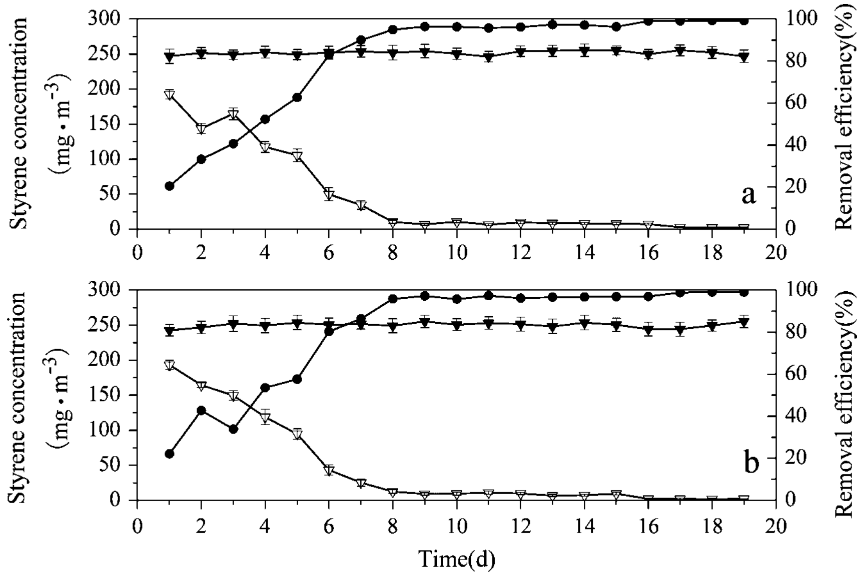

Figure 2. The start-up performance of BTF1 (a) and BTF2 (b). ▼ Inlet concentration; ▽ outlet concentration; ● removal efficiency.

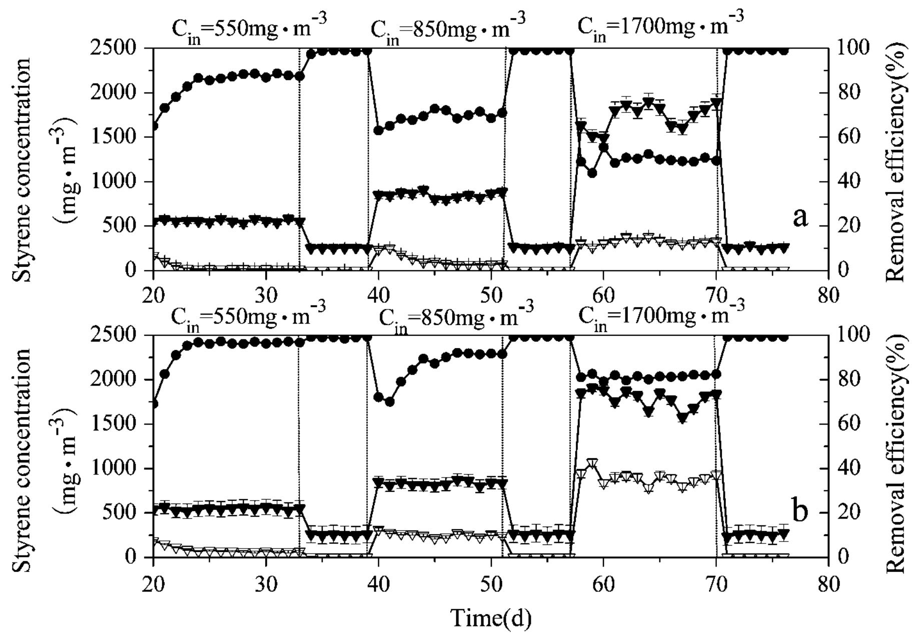

Figure 3. Effect of inlet concentration on the removal efficiency of BTF1 (a) and BTF2 (b) ▼ Inlet concentration; ▽ outlet concentration; ● removal efficiency; C_{in}: styrene inlet concentration.

From Fig. 3 it can be seen that when inlet styrene concentration was increased to 550, 850 and 1700 mg m^{-3}, corresponding to average organic loading rates of 65.3, 100.9 and 201.7 g styrene m^{-3} h^{-1}, the average RE of BTF1 was 87%, 70% and 50%, corresponding to the EC was 54.8, 70.6 and 100.9 g styrene m^{-3} h^{-1}, while the average RE for BTF2 was 96%, 92%, and 82%, corresponding to EC of 62.6, 92.8 and 165.4 g styrene m^{-3} h^{-1}. During all three recovery experiments, the RE of both BTFs reached 99%; during this period, we found that the increase in loading rate also enhanced biomass growth within the medium of BTF1. This was reflected in an increase in the pressure drop values from nearly 30 Pa on day 20, to about 130 Pa on day 70, while the pressure drop values of BTF2 only increased to 80 Pa. From these data, we can conclude that the use of Triton X-100 can improve the degradation of styrene, and prevent the growth of microorganisms to a certain degree, thereby avoiding excessive accumulation of microbes.

Effect of gas EBCT on styrene removal

During this period we continued to supply Triton X-100 to BTF2. The effect of EBCT on the styrene removal efficiency of BTFs was investigated at an average styrene loading rate of 237.3 g styrene m^{-3} h^{-1}. BTF1 and BTF2 were simultaneously evaluated at gas EBCT values of 30.0, 15.0 and 7.5 s, respectively. And the corresponding inlet concentrations were about 2000, 1000 and 500 mg m^{-3}. The RE of BTF1 and BTF2 at standard operating conditions are presented in Fig. 4.

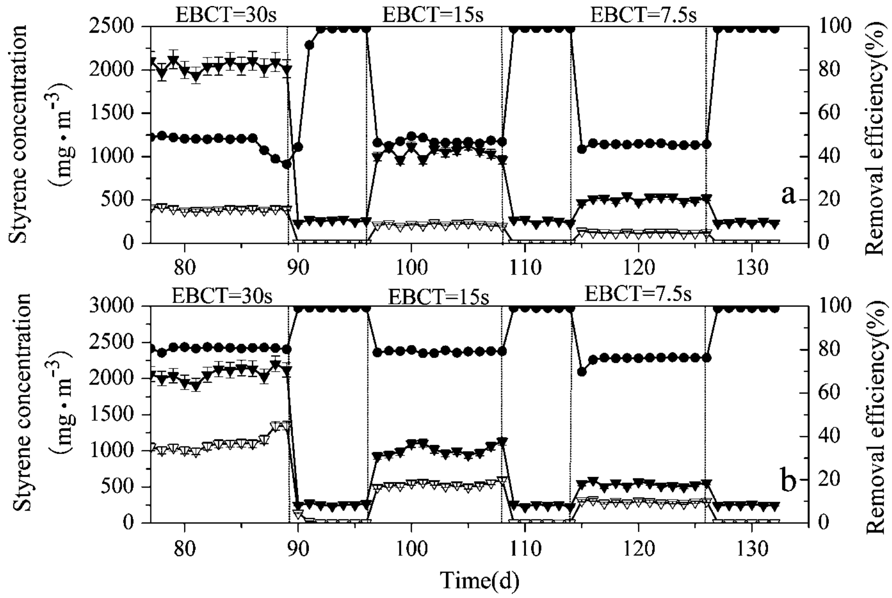

Figure 4. Effect of EBCT on the removal efficiency of BTF1 (a) and BTF2 (b). ▼ Inlet concentration; ▽ outlet concentration; ● removal efficiency.

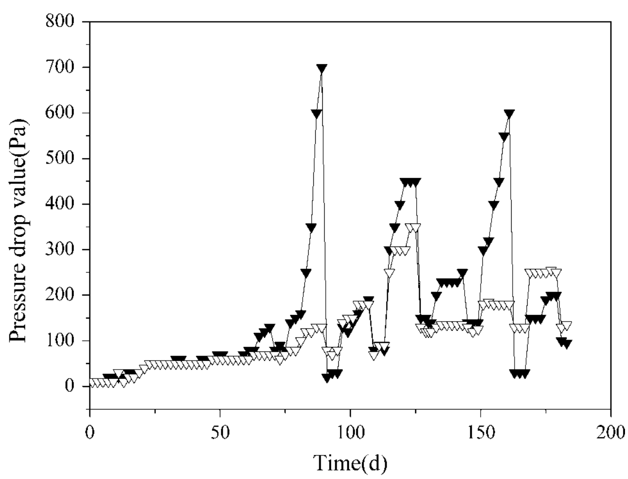

Figure 5. Profiles of pressure drop during the operation of BTF1 and BTF2. ▼ Pressure drop of BTF1; ▽ pressure drop of BTF2.

From Fig. 4 it can be seen that when the average inlet concentration increased to 2000 mg m^{-3}, the average RE of BTF1 was about 48%, while the corresponding EC was 113.9 g styrene m^{-3} h^{-1}. When the average RE of BTF2 was approximately 80%, the corresponding EC value was 189.8 g styrene m^{-3} h^{-1}.

From Fig. 5 it can be seen that the pressure drop values of BTF1 increased from 140 Pa on day 76 to 700 Pa on day 88, and that RE of BTF1 dropped from 48% to 36%. Dense biofilms were attached to the medium.

So, BTF1 was stopped on day 88, and the medium was taken out of the filter and put in a container filled with tap water. Yang *et al.*[25] squeezed the medium repeatedly for excess biomass removal in an investigation of rotating drum biofilters. The media were then put back to BTF1, and started up again on day 88 in reference conditions. During this period the pressure drop values of BTF2 only increased to about 120 Pa. After the excess biomass was removed, the pressure drop values of BTF1 rapidly dropped to 10 Pa. On day 88 on which the media of BTF1 were cleaned, the RE of BTF1 reached lows of 44% at reference conditions. After 24 h acclimation, the RE of BTF1 rapidly increased to 91%, and ivory white biofilm was seen on the surface of the media of BTF1.

After running under reference conditions for 7 days to confirm that the biofilters were at pseudo-steady-state, the average inlet concentration of both BTFs was decreased to 1000 mg m^{-3}, while the gas flow rate was increased to 560 L h^{-1}, and the corresponding EBCT was 15.0 s. As presented in Fig. 4, the RE of both BTF1 and BTF2 decreased slightly. The average RE dropped to 46% in BTF1 and 79% for BTF2, while the average EC of BTF1 and BTF2 was 109.2 and 187.5 g styrene m^{-3} h^{-1}. At the same time, we found that as the gas flow rate was increased, the pressure drop values of the BTFs also increased. The pressure drop of BTF1 increased to about 200 Pa on day 108 and for BTF2 was 180 Pa. After 6 days of recovery experiment, the average inlet concentration of both BTFs were decreased to 500 mg m^{-3}, while the gas flow rates were increased to 1120 L h^{-1}, corresponding an EBCT of 7.5 s. The average RE of BTF1 dropped to 45% and BTF2 to 76%, and the average EC of BTF1 and BTF2 was 106.8 and 180.4 styrene g m^{-3} h^{-1}, respectively. During this period, the pressure drop values of both BTFs increased rapidly as Fig. 5 shows.

From what has been discussed above, we found that clogging occurred due to excess biomass growth when the BTF was operated under high organic loading rates. Clogging of medium bed led to an increase in pressure drop and decrease in styrene degradation efficiency. The use of a surfactant played an important role not only in the degradation of styrene but also in control of biomass growth, as shown by comparison of the performances of BTF1 and BTF2.

Effect of gas flow rate on styrene removal

During this period we continued to supply Triton X-100 to BTF2. The effect of gas flow rate on the styrene removal efficiency was investigated at an average styrene concentration of 1500 mg m^{-3}. BTF1 and BTF2 were simultaneously evaluated at gas flow rates of 140, 280 and 560 L h^{-1}. The corresponding average organic loading rates were 88.98, 178.0 and 355.9 g styrene m^{-3} h^{-1}. The RE of BTF1 and BTF2 at standard operating conditions are presented in Fig. 6.

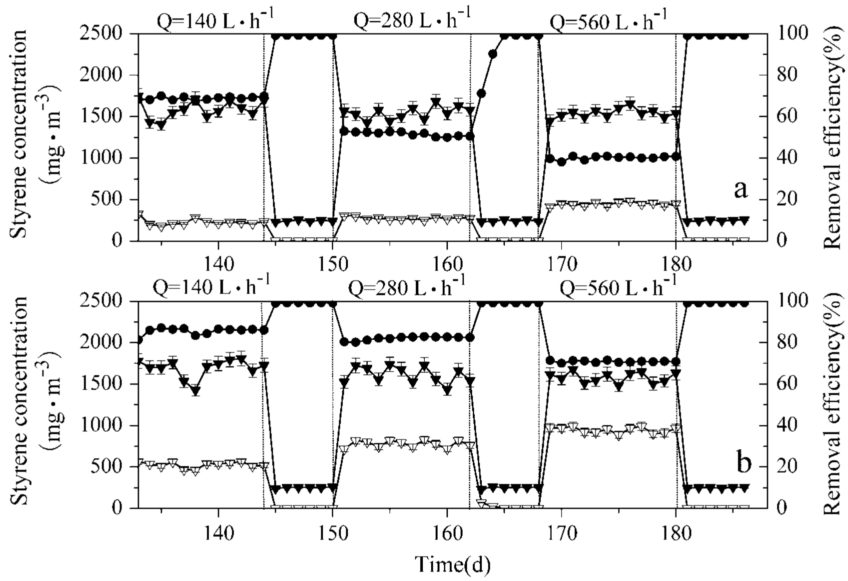

Figure 6. Effect of gas flow rate on the removal efficiency of BTF1 (a) and BTF2 (b). ▼ Inlet concentration; ▽ outlet concentration; ● removal efficiency (Q: gas flow rate).

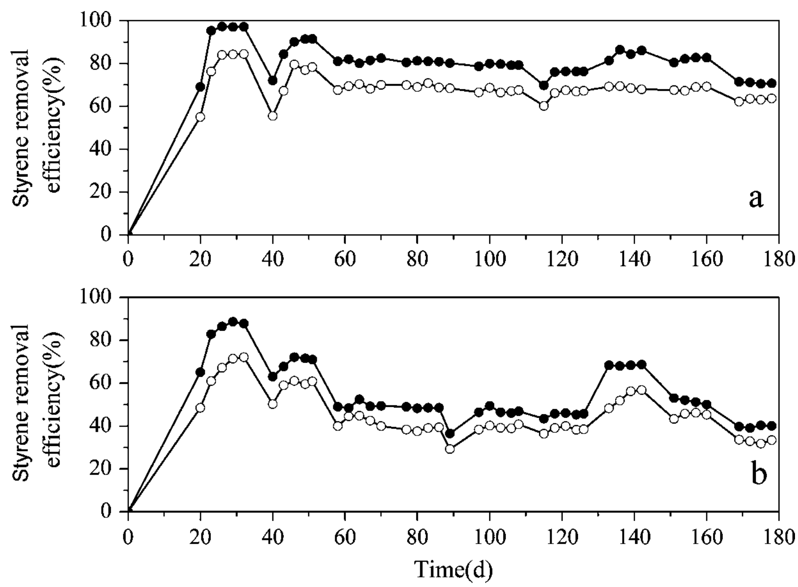

Figure 7. Styrene removal efficiency along the height of of BTF1 (a) and BTF2 (b) during the experiment. ○ RE of the upper and middle layers; ● RE of all three layers.

From Fig. 6 it can be seen that when increased gas flow rate to 140 and 280 L h^{-1}, corresponding to EBCT of 60.0 and 30.0s, the average RE of BTF1 were 70% and 52%, corresponding to EC 62.3 and 92.5 g styrene m^{-3} h^{-1}, while the average RE for BTF2 were 86% and 82%, corresponding to EC 76.5 and 145.9 g styrene m^{-3} h^{-1}.

From Fig. 5 it can be seen that the pressure drop of BTF1 increased suddenly to 550 Pa on day 162, while the styrene RE dropped from 53% to 50%. In consideration of this high pressure drop, BTF1 was stopped on day 162, and the media were cleaned again, the process was as previously described. The media were then put back to BTF1, and started up again on day 162 under reference conditions.

After running under reference conditions for 6 days to confirm that the biofilters were at pseudo-steady-state, the gas flow rate of both BTFs was increased to 560 L h^{-1}, and the corresponding EBCT was 15 s. As presented in Fig. 6, the average RE dropped to 40% in BTF1 and 71% in BTF2, while the average EC of BTF1 and BTF2 was 142.4 and 252.7 g styrene m^{-3} h^{-1}, respectively. The pressure drop throughout the experiment in BTF1 and BTF2 is shown in Fig. 5.

Examination of the performance of BTF1 and BTF2 under the various operating conditions shows that the use of Triton X-100 can increase the styrene degradation efficiency of the BTF under all conditions.

Degradation along the bed height

Figure 7 shows the RE across different bed layers of BTF1 and BTF2. As the height of the media of the BTFs in this study is only 30 cm, the study only measured the styrene removal of the second and third layer. From Fig. 7, it can be concluded that the degradation efficiency of the third layer contributes far less than the first and second layer. That is to say, under certain conditions, if the height of the medium exceeds a certain value, the degradation efficiency is not increased.

CONCLUSIONS

(1) BTFs inoculated with activated sludge and packed with polyurethane foam successfully reached 90% and 99% RE within 8 and 17 days, respectively, at an average inlet concentration 250 mg m^{-3} of styrene and a gas EBCT of 30 styrene s.

(2) At a constant gas EBCT of 30.0 s, styrene RE of the BTFs decreased with increased organic loading rate, while the EC increased. When operating at an average loading rate of 237.3 g styrene m^{-3} h^{-1}, the effects of EBCT and inlet styrene concentration on styrene RE and EC of the BTFs are not significant. When operating at a constant inlet concentration, decrease of gas EBCT can directly decrease the styrene RE of BTFs. For both BTF1 and BTF2, the maximum EC was reached when operated at inlet styrene concentration of 1500 mg m^{-3} and EBCT of 15.0 s. The maximum average ECs for BTF1 and BTF2 were 142.4 and 252.7 g styrene m^{-3} h^{-1}. From the statistics we obtained, it is concluded that the use of Triton X-100 can enhance styrene RE significantly during each operation period.

(3) Styrene RE increases as the height of the medium beds increased to a certain degree, but the degradation efficiency of the lowest layer of the medium bed is not affected significantly in this study. The styrene RE of BTF2 in which the surfactant is applied is higher than that of the other BTF.

(4) Pressure drop across the medium bed increases when gas flow rate or styrene inlet concentration increase due to consequent accumulation of excess biomass within the medium bed, while the use of the surfactant can prevent clogging within the medium bed.

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