Performance of biotrickling filters packed with structured or cubic polyurethane sponges for VOC removal

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Abstract

Two identical bench-scale biotrickling filters (BTFs), BTF 1 and BTF 2, were evaluated for toluene removal at various gas empty bed contact times (EBCTs) and organic loadings. BTF 1 and BTF 2 were packed with structured and cubic synthetic polyurethane sponges, respectively. At a constant toluene loading of 16 g/(m²·hr), toluene removal efficiencies decreased from 98.8% to 64.3% for BTF 1 and from 98.4% to 74.1% for BTF 2 as gas EBCT decreased from 30 to 5 sec. When the toluene loading increased from 35 to 140 g/(m³·hr) at a gas EBCT of 30 sec, the removal efficiencies decreased from 99.1% to 77.4% for BTF 1 and from 99.0% to 81.5% for BTF 2. The pressure drop for both BTFs increased with increased air flow rate, and did not significantly vary while the toluene loading was increased under similar operation conditions. BTF 1 and BTF 2 could start up successfully within 19 and 27 days, respectively, when packed with fresh sponge media, and the performances could be restored in 3–7 days after biomass was removed and wasted from the media. BTF 2 displayed higher removal efficiency even under shorter EBCT or higher loading rate than BTF 1 when other operation conditions were similar, while it showed lower pressure drop than BTF 1 during the whole period of operation. These results demonstrated that both BTFs could treat waste gas containing toluene effectively.

Key words: biofiltration; biotrickling filter; elimination capacity; empty bed contact time; sponge; volatile organic compound

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Introduction

The control of volatile organic compounds (VOCs) from waste gas streams of low concentration has attracted more and more public concern. Biofiltration, initially applied to control odor emissions in soil beds, has become an established technology for VOC emission control. Many investigations and applications of biofiltration technology on VOC removal were reported (Alonso et al., 2000; Devinny et al., 1999; Sorial et al., 1997; Yang et al., 2010). More recently, several novel biofilters capable of reaching higher removal efficiencies were developed (Cox et al., 2000; Li and Moe, 2005; Yang et al., 2003).

Due to the importance of packing media in biofiltration, many investigations or evaluations about various media have been reported (Devinny et al., 1999; Cai and Sorial, 2009; Kim et al., 2007; Koran et al., 2001; Li and Moe, 2005). Synthesis media including polyurethane sponges, polystyrene beads, polypropylene rings and ceramic pellets were used widely as packing media in biofilters (Devinny et al., 1999; Misiaczek et al., 2007; Moe and Irvine, 2000; Sakuma et al., 2006; Shareefdeen et al., 1993). Among these synthetic media, polyurethane sponges have been successfully applied in biotrickling filters (BTFs). Shareefdeen et al. (1993) conducted experiments using polyurethane foam plugs and shredded polyurethane foam mixed peat while choosing perlite and vermiculite as support media, concluding that the polyurethane foam did not have properties that were as favorable for biofiltration as other materials tested. Moe and Irvine (2000) reported methods for manufacturing and characterizing polyurethane sponges tested to be suitable for use in gas-phase biofilters. Leslous et al. (2004) found that polyurethane sponge filled with agar gel could not endure mechanical stress to which particles were subjected in fluidized beds for air biofiltration.

Polyurethane sponge cubes were also used as support medium in biofilters. Gabriel and Deshusses (2003) used spongy cubes in biofilters for control of odor emissions from waste gases in a wastewater treatment plant, which showed that such biofilters removed odors efficiently at gas
empty bed contact time (EBCT) of 1.6 to 2.2 sec. Shim et al. (2006) obtained high individual elimination capacity (EC) for benzene, toluene, and xylene in a polyurethane biofilter. Yang et al. (2003, 2006, 2008a, 2008b, 2010) reported that VOCs and biomass were more evenly distributed in media from rotating drum biofilters packed with single- or multi-layer sponge media.

Sponge media applied in BTFs have displayed many advantages over other media. However, there are few reported works on BTFs packed with different shapes of sponge media. Various shapes will affect significantly the purification process of biofilms and the performance of BTFs. Therefore, investigations and comparison on the performance of BTFs packed with various shapes of polyurethane sponges will lead to a better understanding of biofiltration.

In this study, performances of BTFs packed with either structured polyurethane sponge plugs or cubes for toluene removal from waste gas streams were evaluated and compared under different gas EBCTs and toluene loading rates. The startup performance and pressure drop in the operation of BTFs were also examined. These investigations could be helpful to properly design and operate full-scale BTFs packed with polyurethane sponges.

1 Materials and methods

1.1 Experimental apparatus

Two identical biofilters at bench-scale were used and designated as BTF 1 and BTF 2 in this study. A schematic diagram of the BTFs is presented in Fig. 1. Transparent plexiglas pipe with inner diameter of 10 cm was used to construct the BTFs. Each BTF column height was 65 cm and was equally divided in three sections. A perforated plexiglass plate (Φ 10 cm) was positioned at the base of each section. This enabled the system to evenly distribute incoming gas steams and nutrient solution. A layer of sponge media of 10 cm high was packed on each plate. At the top of the BTFs, there was a 10 cm high headspace for nutrient feed and gas distribution. At the bottom of the BTFs, a 10 cm high housing was also left to collect the leachate and discharge the effluent gas stream. Sample ports were placed in the plexiglass pipes between the bed sections for gas sampling and pressure drop monitoring.

Both BTFs were operated in co-current mode with the gas and nutrient flowed downward. The gas stream, fed by an air compressor, was divided into two streams. One was delivered into a humidifier to be fully saturated, while the other passed through a flask containing liquid toluene reagent (98.5%, analytic reagent grade, Xiangke Chemical Factory, China). Both airflow rates were regulated with two precalibrated flow meters (LZB-10, Yuyao Yinhuan Flowmeter Co., Ltd., China), respectively. Then the gas mixture was delivered to the top of the BTF and successively passed through three layers of sponge media. The contaminants in the waste stream were absorbed into and biodegraded by biofilms in the BTF. The purified gas stream exited out of the BTF through the outlet at the bottom of the BTF.

A nutrient solution was periodically supplied to the BTFs, and was sprayed onto the media through a nozzle.
installed at the upper headspace. A microprocessor-based time controller was used to control a nutrient pump (HQB-3900, Zhejiang Sunsun Industry Co., Ltd., China) as needed. The liquid solution flowed downwards through the media and was discharged at the base of the BTF.

Each BTF was also equipped with an automatic water bath system surrounding the plexiglass pipe, with a water temperature of 25°C.

### 1.2 Model volatile organic compound

Toluene was selected as the model VOC in this study for the following reasons: First, toluene is commonly emitted from a number of industries throughout the world. Second, it was used as a model VOC in many prior investigations (Cox et al., 2000; Kim and Sorial, 2007; Sorial et al., 1997; Yang et al., 2008b; Zhu, 2000; Zhu et al., 2004). Therefore, the results from this study could be compared to other biofilters.

### 1.3 Sponge media

Open-pore reticulated polyurethane sponges with a pore size of 10 pores per cm (Shenzhen Jiechun Filter Material Co., Ltd., China) were used as packing media in this study. The polyurethane sponges have a porosity of 95.3% and an apparent density of 28 kg/m³. The sponge media were either structured cylindrical plugs with a diameter of 10 cm or cubes with a side length of 2 cm. In BTF 1, each section was inserted with one structured cylindrical sponge plug within which an incoming gas stream was distributed evenly. In BTF 2, each section was randomly packed with a 10 cm high layer of sponge cubes. Both BTFs had a total medium height of 30 cm and a medium bed volume of 2.36 L.

### 1.4 Nutrients

The nutrient solution for the BTFs consisted of macronutrients (nitrate and phosphate), micronutrients, and vitamins. The nutrient solution for the BTFs consisted of the following main compounds in tap water (mg/L): NaNO₃ 400, KH₂PO₄ 9.25, and K₂HPO₄ 28.16. Sodium bicarbonate was used as a buffer to prevent large deviation in the pH of the nutrient solution. When in operation, the pH of the nutrient solution was maintained at 7.0 ± 0.2. The feed rate was kept at 4.5 L/day. The nutrient solution provided necessary bed moisture content and nutrients for biofilms within the sponge media.

### 1.5 Seed cultures

The fresh activated sludge was taken from a secondary sedimentation tank at Changsha Guozhen Wastewater Treatment Co., Ltd., China. The activated sludge was used for seeding the BTFs.

### 1.6 Operation of biotrickling filters

Two experimental runs were carried out separately in BTF 1 and BTF 2. Both BTFs completed the initial startup in the first run (from day 1 to day 32). In the second run (from day 33 to day 129), the impact of gas empty bed contact time (EBCT) on the BTF performance was studied at a constant toluene loading. The third run (from day 130 to day 206) was conducted to evaluate the impact of toluene loading on the BTF performance at a gas EBCT of 30 sec. During the experimental period, the pressure drop was also monitored. Then, the performances of the BTFs during the periods of start-up and recovery after biomass removal (as following mentioned in Section 1.7) under different conditions were investigated.

In order to check the reproducibility and pseudo steady-state condition of the BTFs, a set of reference condition was used in this study. When in operation, the BTFs were resumed to the reference level for about one week before and after a change of an operating parameter. The reference condition was set at gas EBCT of 30 sec or toluene loading of 35 g/(m³/hr) during the second and third runs, respectively.

Both gas EBCT and toluene loading rate were important parameters for evaluating the performance of biofilters. In the second run, the effect of gas EBCT on biofilter performance was evaluated at a constant toluene loading rate of 16 g/(m³/hr) after both BTFs achieved stable toluene removal efficiencies. All toluene loading rates and gas EBCTs in this study were based on the medium volume in the BTFs. Both BTFs were evaluated at different gas EBCTs listed in Table 1. All other operating parameters remained constant. The experimental period (97 days) of both BTFs was divided into eight successive stages, i.e., stages A (day 33–44), B (day 45–59), C (day 60–70), D (day 71–84), E (day 85–91), F (day 92–107), G (day 108–122) and H (day 123–129). Among these stages, stages E and G were carried out at the reference condition, and the recovery experiment was conducted after biomass was wasted from the media in stage H. Gas EBCT was varied by adjusting incoming gas flow rate passing the BTF. When the gas flow rate was changed, all other parameters including toluene loading rates, nutrient solution concentration, nutrient solution feed rates, etc., remained unchanged.

In the third run, toluene loading was changed by controlling the air flow rate to the flask containing the liquid toluene. A toluene feed loading was obtained through accurate control of the ratio of flow rate of gas stream into the toluene flask to that into the humidifier. Similarly, when toluene loading was changed, all other parameters remained constant.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Duration (day)</th>
<th>Nutrient solution rate (L/day)</th>
<th>Gas flow rate (L/hr)</th>
<th>EBCT (sec)</th>
<th>Toluene concentration (mg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>33–44</td>
<td>4.5</td>
<td>280</td>
<td>30</td>
<td>135</td>
</tr>
<tr>
<td>B</td>
<td>45–59</td>
<td>4.5</td>
<td>360</td>
<td>15</td>
<td>67</td>
</tr>
<tr>
<td>C</td>
<td>60–70</td>
<td>4.5</td>
<td>840</td>
<td>10</td>
<td>45</td>
</tr>
<tr>
<td>D</td>
<td>71–84</td>
<td>4.5</td>
<td>1120</td>
<td>7.5</td>
<td>34</td>
</tr>
<tr>
<td>E</td>
<td>85–91</td>
<td>4.5</td>
<td>280</td>
<td>30</td>
<td>135</td>
</tr>
<tr>
<td>F</td>
<td>92–107</td>
<td>4.5</td>
<td>1700</td>
<td>5</td>
<td>22</td>
</tr>
<tr>
<td>G</td>
<td>108–122</td>
<td>4.5</td>
<td>280</td>
<td>30</td>
<td>135</td>
</tr>
<tr>
<td>H</td>
<td>123–129</td>
<td>4.5</td>
<td>280</td>
<td>30</td>
<td>135</td>
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</tbody>
</table>

BTF: bench-scale biotrickling filter; EBCT: empty bed contact time.
1.7 Methods of removing biomass from sponge media

The procedure for removing biomass from the sponge media in BTF is as follows. After a BTF was stopped because of microorganism overgrowth, the sponge media was taken out of the column, and put in a container with deionized water. The excessive biomass within the media was removed by squeezing the media repeatedly. Afterwards, the cleaned media was put back in the column which was seeded or started up again.

1.8 Analytical methods

Measurements for the gas phase included influent and effluent concentrations of toluene samples. Toluene concentrations were determined using gas chromatography (GC) (HP 6890N, Series II, Hewlett-Packard, Palo Alto, USA) equipped with a flame ionization detector. A HP-5 capillary column (30 m × 0.25 μm ID × 0.25 μm, Agilent, USA) was used for the analysis. Nitrogen was employed as the carrier gas and was supplied at a flow rate of 9 mL/min. The temperatures at the GC injector, oven and detector were set at 200, 120 and 150°C, respectively.

Gas samples for toluene measurement were collected daily. A certain volume of 3–8 mL of each sample from the sample ports in the BTFs was taken into a gas dilute bottle, when necessary. Each diluted or undiluted sample was collected into a gas-tight syringe and injected into the GC. The sample volume injected into the GC was 1.0 mL. The sample from a same port was taken and measured in triplicate.

The pressure drop was monitored using water manometers connected to both ends of the columns. The manometer scale had a sensitivity of 1 mm H₂O (9.8 Pa). The flow rates of the air streams flowing through the packed columns were checked and adjusted to the controlled flow rate before recording.

2 Results and discussion

2.1 Comparison of startup performances of BTFs

In the first run, both BTFs were operated at influent toluene concentration of about 135 mg/m³ and a gas flow rate of 280 L/hr during the startup period. The corresponding toluene loading rate and gas EBCT were 16 g/(m³-hr) and 30 sec.

Figure 2 summarizes the startup process of both BTFs. The removal efficiency of BTF 1 exceeded 99% and stabilized within 19 days after the startup. BTF 2 reached only 95% removal efficiency on day 19, yet achieved over 99% removal efficiency on day 27.

Misiaczek et al. (2007) reported that a shorter start-up period (7 days) was obtained in a trickle bed reactor packed with polypropylene high flow rings under a gas EBCT of 177 sec and influent toluene concentration of about 130 mg/m³.

Fig. 2 Performance on startup of the two BTFs under gas EBCT of 30 sec and influent toluene concentration of 135 mg/m³.

2.2 Stabilization, reproducibility and re-startup of performances of BTFs

After successful startup of the two BTFs, the stability and reproducibility of the performances were examined under the same conditions mentioned as above from day 1 to day 32 (Fig. 2). The similarity in the toluene removal efficiencies and concentration profiles between BTF 1 and BTF 2 demonstrated that both BTFs performed in a similar manner.

During the whole duration of the BTF operation, the concentration of toluene in the influent streams deviated from the target value because of the fluctuation of gas flow rate in the two parts of stream. However, the variation of influent concentration did not lead to any obvious effects on BTF performance. This established a basic reference for properly evaluating and comparing the effects of gas EBCTs and influent toluene loads on BTF performances in the following experiments. At the same time, the result also indicated that both BTFs could resist the influent shock loading pretty well.

The re-startup performances of both BTFs under the continuous loading conditions were also investigated to obtain a better approach for re-startup at the successive stage of the experiment. Biomass in the media were washed and wasted from the biofilters before the re-startup.

Figure 3 shows the recovery performances of both BTFs after the first removal of excessive biomass accumulated within the media on day 124. It can be seen from that the removal efficiency enhanced rapidly and stabilized above 99% after short acclimation stage. The recovery duration was 3 days for both BTFs. The second recovery performances of both BTFs were conducted on day 197, the recovery duration was 5 and 7 days for BTF 1 and BTF 2, respectively (Fig. 4). The washing of the media and the consequent wasting of biomass from the BTFs had almost no effect on the viability of biofilms because VOC removal performance of the BTFs was soon restored.

These results showed that both BTFs packed with either sponge media could re-startup successfully in short duration and recovered rapidly. However, BTF 1 performed better than BTF 2 during the initial startup and recovery.
periods. It was obvious that biomass within the structured sponge plugs distributed more evenly than that within the sponge cubes. Furthermore, this would provide even distribution of gas streams and nutrition within the medium bed in BTF 1 than in BTF 2. Therefore, BTF 1 posed a better performance of re-startup and recovery.
2.3 Effects of gas EBCT on BTF performance

In the second run, BTF 1 and BTF 2 were used to evaluate the effect of gas EBCT on toluene removal at toluene loading rate of 16 g/(m$^3$·hr), and the results are presented in Fig. 3. When gas EBCT decreased from 30 to 5 sec, the average toluene removal efficiency at pseudo steady state decreased from 98.8% to 64.3% for BTF 1, and dropped from 98.4% to 74.1% for BTF 2. It can be seen from Fig. 3 that toluene removal efficiency decreased while EBCT decreased at a constant toluene loading rate. Sorial et al. (1997) and Yang et al. (2008b) have reported similar results. The following reasons could contribute to the result. First, the shorter gas EBCT was, the fewer contact between toluene and biofilm. Secondly, the intermittent nutrient solution supply led to significant variation of moisture content in the biofilm phase, especially at shorter gas EBCT. Thirdly, the incomplete humidification of influent gas might lead to the drying of the packing materials and high salinity at the inlet of a BTF (Qi et al., 2005), which was also consistent with the fact that we observed in the operation.

However, both BTFs could maintain high removal efficiencies even if at a short gas EBCT. Figure 3 shows that both BTFs achieved 64.3% and 74.1% respectively at an EBCT of 5 sec. It was worth notice that BTF 2 achieved higher removal efficiency. For this result, various shapes of packing materials played an important role in biofilter performance (Leslous et al., 2004; Sakuma et al., 2006). The polyurethane sponge cubes in BTF 2 provided more space for biomass accumulation, while the structured sponge plugs in BTF 1 provided more uniformly distributed pores. It is anticipated that the effective contact area of biomass with waste gas streams in BTF 1 was larger than that in BTF 2 initially. The interstices among the sponge cubes in BTF 2 decreased with operation time by a much less rate than the void volume within the pores of sponges. Therefore, the effective contact area in BTF 1 decreased more quickly than that in BTF 2, which led BTF 2 achieved higher toluene removal efficiency than BTF 1 under similar conditions.

Pollutant degradation performance of biofilters can also be expressed in terms of elimination capacity (EC) (Devinnny et al., 1999). The EC decreased rapidly as gas EBCT decreased from 15 to 7.5 sec during stage B, C and D (see Table 1) at a constant toluene loading rate of 16 g/(m$^3$·hr). In stage A at gas EBCT of 30 sec, toluene in the incoming gas stream was almost removed completely, and the EC achieved 15.2 g/(m$^3$·hr). From stage B to D, the gas EBCT was set at 15, 10 and 7.5 sec, and the corresponding EC decreased to 14.1, 9.5 and 8.2 g/(m$^3$·hr). During stage F when the EBCT was further decreased to 5 sec, the EC increased to 14.3 g/(m$^3$·hr) which is higher than expected. The too high gas flow rate for this system was considered to be very important reason for this phenomenon. At high gas flowrate, it is very difficult to precisely control the flow rate, and influent toluene loading rate averaged 19.5 g/(m$^3$·hr) which was also higher than the target value of 16.0 g/(m$^3$·hr). Similar results were observed in BTF 2. Most of the feed loading rate of both BTFs was removed after the gas streams passed through the two uppermost layers of the sponge media under the various EBCTs. In stage E, mass of biofilms within the middle and bottom layer media increased in different degrees, and the ECs of both BTFs in stage F increased consequently. These results achieved in both BTFs at low EBCT displayed that both BTFs showed better performances than those in previous studies under similar condition. The highest EC value reached 9.8 g/(m$^3$·hr) at inlet concentration of toluene of 100 mg/m$^3$ and EBCT of 13 sec in a trickle bed reactor packed with polypropylene high flow rings (Misiaczek et al., 2007).

In this run, the pressure drop of the both BTFs was also investigated. From stage A to G, the pressure drop values for both BTFs increased with an increased air flow rate. The average pressure drop values ranged from 95.5 to 986 Pa and from 52.9 to 402 Pa for BTF 1 and BTF 2, respectively, and BTF 2 showed a lower pressure drop value than BTF 1 under similar operating conditions. It was obvious that the transfer resistance of structured sponges was higher than cubic sponges. The pressure drop value at a gas EBCT higher than 7.5 sec for BTF 1 or higher than 5 sec for BTF 2 was lower than 205.8 Pa, the recommended value of pressure drop for practical application of biofilters (Ergas et al., 1995). Lower pressure drops resulted from a high porosity of polyurethane sponges.

During stage A, E, and G when both BTFs were operated at the reference conditions in the second run, the pressure drop of both BTFs increased slowly due to the gradual accumulation of biomass within the media. During stage G, both BTFs were operated again at the reference condition to ensure the reproducibility of the BTFs. Unfortunately, the pressure drops for BTF 1 and BTF 2 increased gradually in stage G which reached 1370 and 490 Pa, respectively, on day 120, and were higher than that at stage A. Visual observation showed that a layer of water film existed on the uppermost layer of the media during stage G, which, very likely, resulted from the excessive accumulation of biomass within the uppermost layer of the media and the consequent clogging of the media. The excess biomass was removed by squeezing the media and was wasted from the biofilers on day 124, and the pressure drop for BTF 1 and BTF 2 decreased to 181 and 50.6 Pa, respectively.

2.4 Effects of toluene loading rate on BTF performances

Organic loading rate is another very important parameter in the evaluation of a reactor’s performance. In the third run (from day 130 to day 206), the toluene loading rates evaluated were 35, 70, and 140 g/(m$^3$·hr), successively, and the corresponding toluene concentrations in the incoming gas streams were 295, 590, and 1180 mg/m$^3$. The gas flow rates were 280 L/hr which leads to a gas EBCT of 30 sec. All other operation conditions remained unchanged. The performances of BTF 1 and BTF 2 at the various influent toluene concentrations are illustrated in Fig. 4. During the period from day 130 to day 139, the influ-
ent toluene concentration averaged 295 mg/m$^3$, and the toluene removal efficiency exceeded 99% stably. From day 140 to day 155, the influent average toluene concentration to both BTFs was kept at 590 mg/m$^3$, resulting in a loading rate of 70 g/(m$^3$·hr). The average removal efficiency during this period immediately dropped to 78.1% and 75.7% for BTF 1 and BTF 2, respectively. Then the loading rate was resumed to the reference condition for the next 19 days, and over 99% removal efficiency was achieved within 1 day for BTF 1 and within 11 days for BTF 2. From day 176 to day 187, the average influent toluene concentration to both BTFs was set at 1180 mg/m$^3$ (140 g/(m$^3$·hr)), and the corresponding removal efficiency for BTF 1 and BTF 2 stabilized at about 77.4% and 80.4%, respectively. In general, VOC removal efficiency of a BTF decreased when a toluene feed loading rate was increased under a constant EBCT. Yang et al. (2006) reported that toluene removal efficiency declined from 99.8% to 82.2% and 74.1% at an EBCT of 30 sec in a rotating drum biofilter when corresponding VOC feed concentration was increased from 221 to 442 and 884 mg/m$^3$. In this study, however, the removal efficiency slightly increased when influent loading rate was increased from 70 to 140 g/(m$^3$·hr) in BTF 2. This is closely related to the efficient contact area.

Toluene loading rate was then decreased to 295 mg/m$^3$ on day 187, the reference loading rate, to check the reproducibility of both BTFs. Toluene removal efficiencies of BTF 1 and BTF 2 stabilized at 99.0% and 90.2%, respectively. The removal efficiency of BTF 2 is lower than 99.0% removal efficiency at the earlier reference condition from day 156 to day 175. Excess accumulation of biomass within the media and consequent clogging among the medium cubes in BTF 2 were considered to be the dominant factor. Moreover, a relatively low quantity of biocatalyst after long term operation would also contribute to the effect (Misiaczek et al., 2007). The excess biomass was then removed and wasted from the BTF systems on day 197, and stable removal performance was reached after 6 and 7 days for BTF 1 and BTF 2, respectively.

Although toluene removal efficiency decreased, toluene EC of both BTFs increased with an increased influent toluene loading rate at the gas EBCT of 30 sec from day 130 to day 206. EC of BTF 1 achieved 34.0 g/(m$^3$·hr) at influent loading rate of 34.4 g/(m$^3$·hr) from day 130 to day 139. Then, EC increased to 49.2 and 113.0 g/(m$^3$·hr) when influent toluene loadings rate were 70 and 140 g/(m$^3$·hr), respectively. In BTF 2, the elimination capacity increased from 32.9 to 113.6 g/(m$^3$·hr) when the toluene loading rates increased from 35 to 140 g/(m$^3$·hr) at a gas EBCT of 30 sec. The maximum value of EC obtained was 133.9 g/(m$^3$·hr) when the feed loading rate was maximized at 161.0 g/(m$^3$·hr) for BTF 2.

The maximum EC value obtained in BTF 2 was higher than that in previous studies. A BTF packed with open-pore polyurethane foam reached toluene EC of 22 g/(m$^3$·hr) at gas EBCT of 27 sec and influent concentration of 860 mg/m$^3$ (Sakuma et al., 2006). The overall toluene EC reached a maximum of 125 g/(m$^3$·hr) in the recycle liquid of BTFs at toluene concentration of 1.8–2.2 g/m$^3$ within 2 days after reactor start up (Cox et al., 2000). ECs of 77 and 55 g/(m$^3$·hr) were reached with *E. oligosperma* and *Paecilomyces variotii*, respectively in fungal biofilters when the toluene load was 100 g/(m$^3$·hr) (Estévez et al., 2005). Yang et al. (2008b) reported that the toluene EC increased from 20.4 to 26.7 g/(m$^3$·hr) when the feed concentrations in the incoming gas streams were 442, 221, 110, 55.2, and 36.8 mg/m$^3$ for multi-layer rotating drum biofilters, and the corresponding gas EBCTs were 60, 30, 15, 7.5, and 5.0 sec, respectively. The reason that high ECs achieved in this study should attribute to higher specific surface area of polyurethane sponges than the media used in other BTFs. There was also more space for microorganisms to grow in the sponge media due to a higher porosity, more biomass could be kept within the medium bed and enhanced the efficiency of mass transport between the gas/liquid phase and biofilm phase. Another important characteristic of most inert carriers is that they present a much more regular shape than natural ones, allowing a uniform air distribution (Kennes and Veiga, 2002). More uniform distribution of pores of the sponge could also reduce the channeling of gas streams within the media. Therefore, high removal efficiency and EC could be achieved.

In this study, BTFs packed with either structured sponge plugs or cubes performed rather stably under dynamic mass loadings when the influent organic loading ranged from 30.2 to 150 g/(m$^3$·hr). The stable response to dynamic mass loadings was mainly due to the even distribution of pores and the consequent even distribution of biofilms within the sponge media. The biofilms in the whole medium bed could play an important role in the degradation of toluene at the high toluene loadings, while only the biofilms close to the gas inlet port could significantly contribute to toluene degradation at the low toluene loadings. Biomass was evenly distributed on the uppermost surface of the sponge media in biofilters, and the uppermost surface was also considered to contribute significantly to VOC EC for a biofilter due to the accumulation of more biomass in this region (Devinn et al., 1999; Sakuma et al., 2006). The mass of microorganisms within the middle and bottom layers of media would increase with time, which would enhance the EC. The profile of biomass accumulation rate along the different layers of the media in the BTFs supported this conclusion. In the second run, 14.4 and 2.0 g VSS were yielded within the upper (included the top and middle layer) and the bottom layers of the media in BTF 1. In the third run, the corresponding mass of biomass were 18.8 and 4.0 g VSS in BTF 1. However, biomass yielded 11.9 and 4.2 g VSS within the upper and the bottom layer in second run in BTF 2, and the corresponding mass of biomass were 27.7 and 10.7 g VSS in the third run. These results demonstrated that most biomass accumulated in the layer close to the gas inlet. Sakuma et al. (2006) reported that the higher toluene degradation performance was related to the total number of active microorganisms in the packing material.

The growth of microorganisms could also be reflected by pressure drop. The pressure drop ranged from 140 to
330 Pa for BTF 1 and from 30 to 150 Pa for BTF 2 in the third run (Fig. 5). The main reason was that the volume of the interstices among the sponge cubes was bigger than the volume of the voids in the structured sponge plugs within the medium beds. Furthermore, compared to other media, the open-pore reticulated polyurethane sponges could limit excess accumulation of biomass to some extent in a local region which would reduce clogging and channeling efficiently. In this study, BTF 2 which was packed with sponge cubes was considered more economical for toluene removal due to the lower pressure drop. However, the pressure drop always increased slowly but steadily with the duration of operation, which could eventually lead to clogging of the medium bed and consequently lead to performance deterioration. Therefore, the pressure drop should be monitored closely to prevent the accumulation of excess biomass within a medium bed.

3 Conclusions

Two identical BTFs packed with either structured plugs or cubes of polyurethane sponges were evaluated under various EBCTs and toluene loading rates for toluene removal from waste gas streams. At a constant toluene loading rate of 16 g/(m$^3$.hr), VOC removal efficiency increased with increased gas EBCT value for both BTFs. At a gas EBCT of 30 sec, the elimination capacity of both BTFs increased with increase in inlet loading rate. BTF 1 and BTF 2 could start up successfully and exceeded 99% in removal efficiency after 19 and 27 days, respectively, and recovered in 3–7 days after excessive biomass was removed from the BTFs by washing. Both BTFs could resist efficiently the shock loading. This study demonstrated that the BTFs packed with polyurethane sponges could effectively remove toluene from waste gas streams at various VOC loading rates and gas EBCTs. However, BTF 2 displayed higher removal efficiency even under shorter EBCT or higher loading rate than BTF1 when other operation conditions were similar, while showed lower pressure drop than BTF 1 during the whole period of operation. Therefore it is concluded that proper shape selection of packing materials in BTF systems could affect performance of BTFs for VOC removal significantly.

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