Iran. J. Environ. Health. Sci. Eng., 2011, Vol. 8, No. 3, pp.

PERFORMANCE OF A BIOTRICKLING FILTER EMPLOYING *THIOBACILLUS THIOPARUS* IMMOBILIZED ON POLYURETHANE FOAM FOR HYDROGEN SULFIDE REMOVAL

[1]N. Abdehagh, [1]M. Taghinejad Namini, *[2]S. M. Heydarian, [1,3]B. Bonakdarpour, [2]D. Zare

[1] Department of Chemical Engineering, Amirkabir University of Technology, Tehran, Iran
[2] Department of Biotechnology, Iranian Research Organization for Science and Technology (IROST), Tehran, Iran
[3] Food Process Engineering and Biotechnology Research Center, Amirkabir University of Technology, Tehran, Iran

Received 20 January 2010; revised 20 June 2011; accepted 14 August 2011

ABSTRACT

The removal of hydrogen sulfide (H_2S) from contaminated airstream was studied in a biotrickling filter (BTF) packed with open-pore polyurethane foam as a carrier of *Thiobacillus thioparus* (DSMZ5368) with counter current gas/liquid flows. The effect of operating parameters on BTF performance was studied. Experiments were performed at different Empty Bed Residence Times (EBRT) from 9 to 45 seconds, and different initial H_2S concentration from 25 to 85 ppm. The results showed reasonable performance of the BTF, in H_2S removal from the synthetic gas stream. However, the performance was somewhat lower than other studies in BTF in which either *Thiobacillus thioparus* with other packings or polyurethane foam with other microbial cultures were used. The effect of liquid recirculation rate (LRR) in the range of 175-525 ml/min (0.46-1.34 m/h) on BTF performance was also studied. Results showed that increasing LRR from 175 to 350 mL/min resulted in significant enhancement of H_2S removal efficiency, but further increase in LRR up to 525 mL/min had an insignificant effect. H_2S elimination at different heights of the bed was studied and it was found that decrease in EBRT results in more homogeneous removal of the pollutant in BTF. Determination of microbial species in the BTF after 100 days performance showed that during BTF operation the only H_2S degrading specie was *Thiobacillus thioparus*.

Key words: Biotrickling filter; Hydrogen sulfide; Polyurethane foam; *Thiobacillus thioparus*

INTRODUCTION

Hydrogen sulfide (H_2S) is a toxic, colorless and flammable gas heavier than air, with an odor threshold of about 0.47 ppb and a typical smell of rotten egg. It is released to the atmosphere as a by-product of industrial processes including sour gas flaring, petroleum refining, wastewater treatment, food processing, pulp and paper manufacturing and the treatment of fuels (Chung *et al.*, 1996; Hendrickson *et al.*, 2004). The removal of H_2S from waste gases is necessary because it is harmful for living organisms and its maximal allowed concentration (MAC) is 10 ppm. At concentration of 50 ppm, body develops symptoms; at 150 ppm, the smell disappears and at around 500 ppm it causes rapid knock down of exposed person (Naminі*et al.,* 2007). Therefore, strict regulations are necessary for controlling the emission levels.

A number of methods including absorption, combustion, masking and scrubbing have been used to remove this dangerous contaminant from industrial waste gas streams. But the physicochemical methods have high energy requirements, high chemical and disposal

*Corresponding author: E-mail: heydarian@irost.org
Tel: +98 912 148 36 85, Fax: +98 228 227 66 30

costs and result in the production of secondary pollutants (Namini *et al.*, 2007). Compared to these physical and chemical processes, biofiltration is considered economical, cleaner and greener because of its low operation costs, absence of secondary pollutants and emission of lower amounts of environmentally unfriendly gases compared to other methods (Jin *et al.*, 2005a; Shahmansouri *et al.*, 2005).

Two types of biofiltration reactors have shown most promise as alternatives to physical and chemical treatments: biofilters and biotrickling filters (Delhomenie and Heitz, 2005; Duan*et al*., 2005; Qi *et al*., 2005). In biotrickling filters, polluted air is passed, together with a recycled liquid, through a packed bed on which a pollutant-degrading biofilm develops. As a result of the liquid recirculation flow, the reaction products are washed out of the medium and acidification, especially in eliminating Volatile Organic Compounds (VOCs), can thus be avoided; for this reason, biotrickling filters offer superior performance over biofilters. In addition, the presence of the free trickling liquid phase in the biotrickling system allows better control of operational conditions (Duan *et al.*, 2005; Nikpay*et al.,* 2006).

Packing materials used up to date in biotrickling filters include either natural or synthetic materials such as ceramic saddles, polyethylene pall rings, polyurethane foam, activated carbon, and extruded diatomaceous earth pellets. The use of synthetic packing media has advantages, such as low head losses due to larger interstices between packing granules or pieces, larger specific surface areas, and the possibility of solid phase adsorption of contaminants (Moe and Irvine, 2000; Chitwood and Devinny; 2001, Duan*et al.,* 2005; Ramirez *et al.*, 2009; Mehrdadi *et al.*, 2010).

Polyurethane foam (PUF) is a synthetic packing media with widespread usage in biotrickling filters. Few cases of the use of PUF in biofiltration appear in the literature, and accounts generally do not contain specific information about the media properties. Shareefdeen *et al*. (1993) reported the use of PUF plugs and shredded polyurethane foam mixed with peat, perilte, and vermiculite as a support medium but no specific information is given in this paper about the media properties. There are extensive reports of the use of PUF as synthetic bed in biotrickling filters for immobilization of activated sludge, pure and mixed fungi and *Brevibacillus* sp. (Philip and Deshusses, 2003; Deshusseset *al.,* 2004; Gabriel *et al.,* 2004; Kim and Deshusses, 2005). However, the use of this packing with autotrophic bacteria in BTF has not been reported before.

The aim of the present study was to evaluate the performance of a BTF containing PUF as synthetic bed immobilized with the autotrophic bacteria, Thiobacillus *thioparus,* for the removal of H_2S from a synthetic gas stream containing moderate concentrations of H_2S. Moreover the effect of Liquid Recirculatin Rate (LRR) on the removal efficiency was examined at different amounts of H_2S concentration and Empty Bed Residence Times (EBRT). Finally, the removal efficiency as a function of distance along the bed of BTF was studied.

MATERIALS AND METHODS

Microorganism and media

Thiobacillus thioparus (DSMZ5368) was obtained from Persian Type Culture Collection (PTCC). The strain was maintained on agar slant (medium DSM 486) containing (in g/L): KH_2PO_4, 2.00; K_2HPO_4, 2.00; NH_4Cl, 0.4; Na_2CO_3, 0.4; $MgCl_2.6H_2O$, 0.2; $Na_2S_2O_3.5H_2O$, 5; plus vitamin and trace metals solutions. The inoculum for the biotrickling filter was prepared in liquid culture using a rotating shaker at 150 rpm and 30ºC. The medium used (DSM 36) had the following composition: (g/L) $(NH_4)_2SO_4$, 0.10; KH_2PO_4, 4.00; K_2HPO_4, 4.00; $MgSO_4.7H_2O$, 0.10; $CaCl_2$, 0.10; $FeCl_3.6H_2O$, 0.02; $MnSO_4.H_2O$, 0.02; $Na_2S_2O_3.5H_2O$, 10.00.

Immobilization and acclimation experiments

Both immobilization and acclimation were carried out in the BTF according to the procedure previously described by Naminiet *al.*(2007). Immobilization of BTF was started by recirculating 2.5 liters of *T. thioparus* solution. Contrary to some previous works (Wu *et al.*, 2001; Deshusses and Gabriel, 2005) sodium thiosulfate was used in preparation of the inocula, and this substrate, instead of H_2S, was supplied to the bacterial culture in the BTF during the immobilization

stage. The consumption of thiosulfate and pH changes were monitored and controlled during the immobilization stage. pH was adjusted to 7 every day and 500 mL of liquid solution was replaced with 500 mL of fresh nutrient solution every 24 hours. The highest concentration of sulfate measured during these experiments was 1.7 g/L. Immobilization was continued until the consumption of sodium sulfate in the recirculating liquid approached steady state conditions. The complete and steady consumption of 8 g/L.day of thiosulfate occurred from the eighth day onwards. After 12 days the acclimation stage started with introduction of air stream containing 60 ppm of H_2S at EBRT of 45 seconds. It took 6 days to reach steady state conditions in the outlet.

Experimental setup

The laboratory scale BTF used in the present study had a total height of 90 cm with diameter of 8.8 cm. The active height was 60 cm, which resulted in an active volume of 3.64 liters. The schematic of BTF is shown in Fig.1. Open-pore polyurethane foam was used as support for the microbial population. The initial porosity and density of PUF were determined as 97% and 7.445 kg/m^3, respectively. Volume of recycled liquid in the vessel was 2.5 liters. The countercurrent mode of gas/liquid flow was the chosen mode of operation. A solution of 4M NaOH was used to control the pH of recycled liquid with a peristaltic pump. During the operation of the BTF, pH was kept constant at 7. Three sampling ports were located at 20, 40 and 60 cm of effective bed height. 1 liter of recycled medium was replaced with concentrated fresh aqueous mineral medium every 24 hours.

Analytical methods

The hydrogen sulfide concentration was determined using a Drager Sensor (Drager Sensor Micropac Plus, Drager Safety, Germany). Thiosulfate and sulfate analysis were carried out idometrically and gravimetrically, respectively (Mendham, 2006). Microbial counts were conducted by plating biofilm samples on various media. Determination of population density of *T. thioparus* was done using colony counting procedure employing DSM-486 agar medium. For the study of system contamination samples of biofilm were cultured in both nutrient agar medium (for bacterial contamination) and potato dextrose agar (PDA) (for fungal contamination). To assess the performance of the biotrickling filter, H_2S removal efficiency (RE) and elimination capacity (EC) were determined at different EBRTs, and pollutant loadings (L). These parameters are defined according to the following equations:

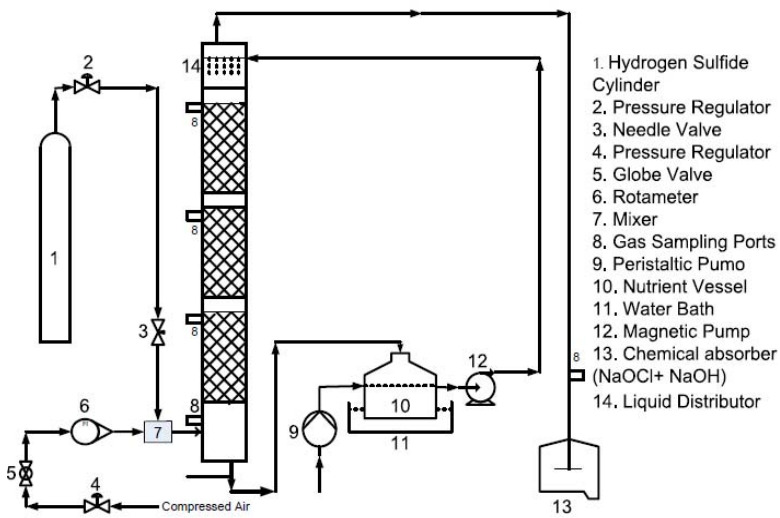

Fig.1: Schematic of Biotrickling filter

$$RE(\%)=\frac{C_{in}-C_{out}}{C_{in}} \quad (1)$$

$$EBRT(s)=\frac{V}{Q} \quad (2)$$

$$EC\left(\frac{g}{M^s\,h}\right)=\frac{C_{in}-C_{out}}{V}\times Q \quad (3)$$

$$L\left(\frac{g}{M^s\,h}\right)=\frac{C_{in}}{V}\times Q \quad (4)$$

Where the C_{in} and C_{out} are inlet and outlet pollutant concentrations; V is volume of the packed bed (m^3) and Q is gas flow rate (m^3/h).

RESULTS

Effect of loading on H_2S *removal*

The H_2S loading to the BTF was increased by either increasing the inlet concentration of pollutant or decreasing EBRT (Namini *et al.*, 2007). Fig.2 shows the effect of increase in the loading rate, by decreasing EBRT, on H_2S elimination capacity at constant H_2S concentration.

As it can be seen all of the curves can be divided into three regions: in the first region (first order regime) the elimination capacity and loading of H_2S are the same and the removal is complete. In the second region, the breakthrough of H_2S occurs. In this region, the elimination capacity of H_2S increases to a lesser extent than the loading and the removal efficiency starts to decease below 100%. In the third region (zero order regime), the BTF reaches its maximum elimination capacity (EC_{max}) and an increase in loading does not lead to increase in elimination capacity.

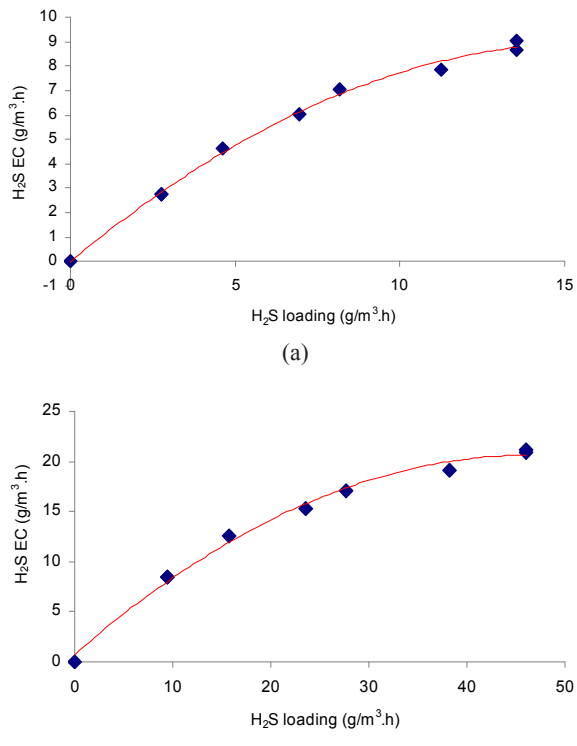

Fig. 2: Effect of decrease in EBRT (at constant H2S inlet concentration) on EC in the BTF at a)Cgi=25 ppm b) Cgi=85 ppm (0.1 g/m^{-3} corresponds to 73 ppmv)

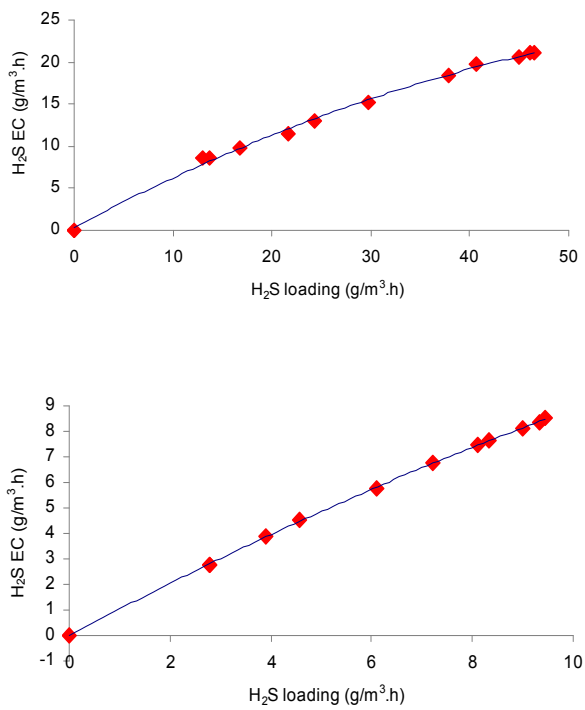

Fig. 3: Effect of increase in H2S inlet concentration at constant EBRTs on EC in the BTF at a) EBRT= 45 s, b) EBRT=9s

Fig.2 shows that EC_{max} increases from 7.2 to 20.6 g/m^{3}h with increase in H_2S concentration from 25 ppm to 85 ppm. The EC_{max} observed at H_2S inlet concentration in the range 40-75 ppm was between these two values and also increased with increasing H_2S concentration (results not shown).

Effect of increasing loading rate (by increasing H_2S inlet concentration) on H_2S elimination capacity at constant EBRT is presented in Fig.3. The results show the EC_{max} increases from 7.5 to 20.6 g/m^3.h with decrease in EBRT from 45 s to 9 s (Fig. 3). The EC_{max} observed at EBRT in the range of 11-27 seconds was between these two values and also increased with decreasing EBRT (results not shown). These results are in line with findings of previous studies (Devinny*et al.*, 1999; Martin *et al,* 2004; Deshusses, 2005).

The results of removal efficiency of H_2S as a function of loading at different H_2S concentrations show that the removal efficiency decreases more rapidly at low H_2S concentration (Fig.4). For example, at loading rate of 10 g H_2S/m^3.h the removal efficiency for 20 ppm (EBRT=9 s) and 85 ppm (EBRT=45 s), were 81% and 90%, respectively.

Effect of LRR on H_2S *removal*

To study the effect of LRR on H_2S elimination, three levels of LRR (175, 350 and 525 mL/min) were selected and the performance of BTF was studied at different conditions. Performance of BTF was almost the same at LRR of 350 and 525 mL/min, but there was a significant difference between the performance of BTF at LRR of 175 and 350 mL/min. This is illustrated by the

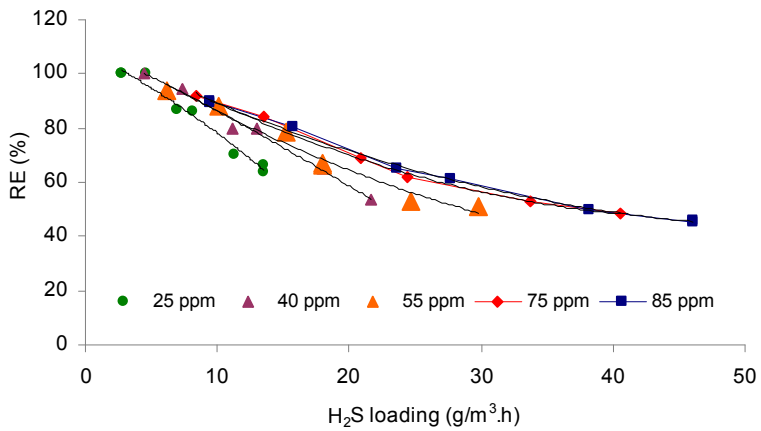

Fig. 4: Removal efficiency of H2S as a function of H2S loading rate in the BTF

relationship between H_2S elimination capacity (Fig.5a) and removal efficiency (Fig.5b) versus H_2S loading at two different LRRs at C_{in}=40 mg/L. For runs at other H_2S concentrations, similar effect of LLR on the performance of the BTF was observed (results not shown).

Effect of EBRT and C_{in} *on the elimination of* H_2S *at different heights of bed*

Choosing the bed height is one of the most important issues in designing a biotrickling filter. The height and cross sectional area of bed determines the volume of the bed and consequently the empty bed residence time (Namini *et al,* 2007). In this study the removal of H_2S at different heights of BTF was determined. Fig.6 shows the removal efficiency versus height of the bed at three different EBRTs in the range 9-45 s when the inlet concentration of H_2S was 25 ppm. It can be seen that decreasing EBRT leads to slight increase in homogenous elimination of H_2S at different heights of the BTF. For example, at EBRT of 45s, 80% of elimination occurred in 67% of bed height, but when EBRT decreased to 9 s, only 60% of elimination occurred in the same height of the bed.

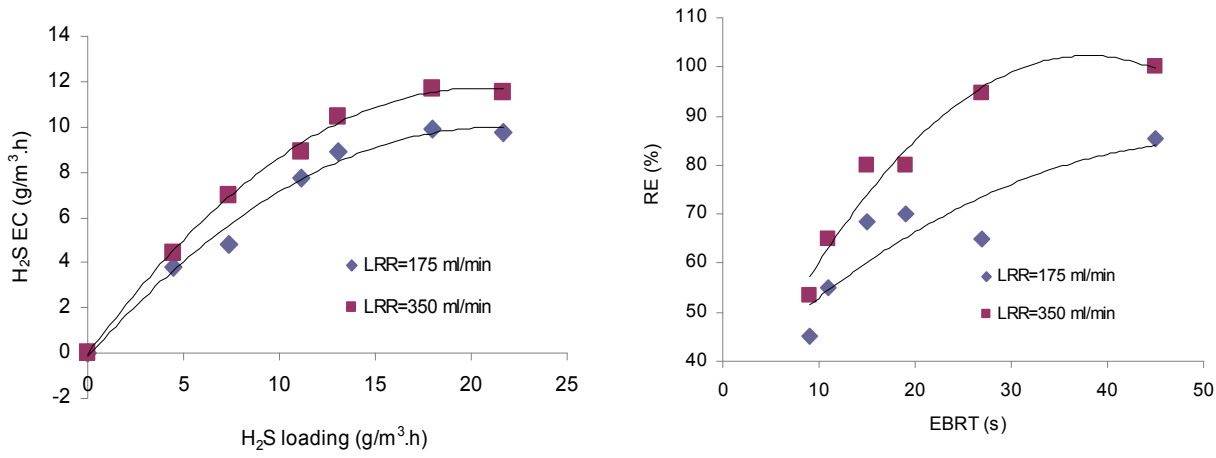

Fig. 5:(a) Effect of LRR on the H2S elimination capacity at Cgi=40 ppm and different EBRTs (b) Effect of LRR on the removal efficiency at Cgi=40 ppm and different EBRTs

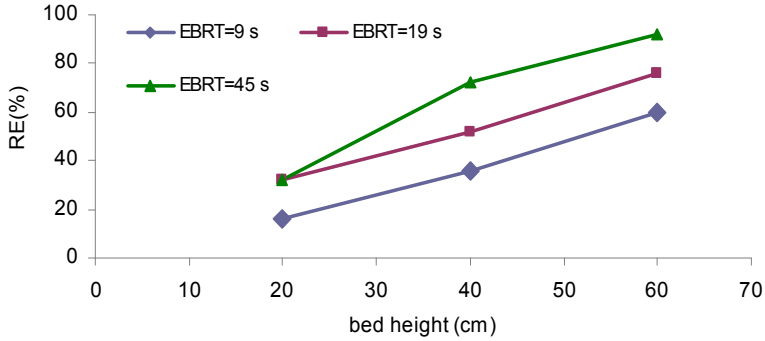

Fig. 6: Removal efficiency versus effective bed height of BTF at constant Cgi and different EBRTs

Investigation of the system contamination

After 100 days of operation, the contamination in the BTF was investigated. The results confirmed existence and dominance of *T. thioparus*. The plate count of samples of biofilm on the appropriate media confirmed the existence of three other unidentified species of bacteria (Figs.7 a, b) and one unidentified species of moulds (Fig.7c). However, when the bacterial and fungal contaminant were cultivated in flasks with H_2S feeding, no removal of H_2S took place suggesting that the contaminating species were incapable of H_2S removal (results not shown).

DISCUSSION

The BTF using polyurethane foam as a synthetic packing for immobilization of autotrophic *T. thioparus* showed reasonable performance in the removal of H_2S from synthetic air streams containing H_2S concentrations in the range of 25-85 ppm, but lower than those reported in other studies in which either polyurethane foam with other microorganisms or *T. thioparus* with other packings were employed. This can be attributed to unsatisfactory immobilization of *T. thioparus*on polyurethane foam.

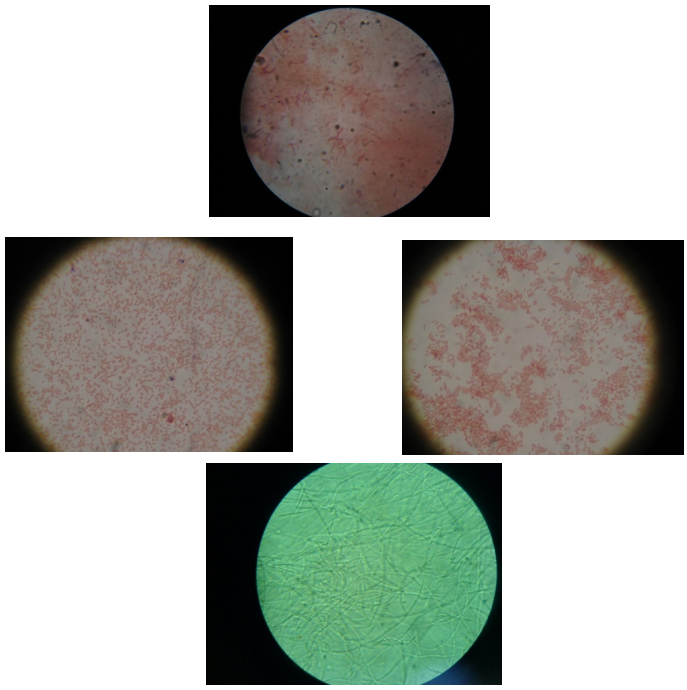

Fig. 7: (a *T.thioparus* (b) Bacterial contaminants of the BTF (c) Fungal contaminants of the BTF

Results of immobilization and acclimation experiments showed that the length of immobilization and acclimation stages (12 and 6 days) were reasonable compared to previous reports (Martin *et al*, 2004; Deshusses, 2005; Deshusses and Gabriel, 2005; Namini*et al,* 2007). It has been previously shown that concentration of sulfate > 7 g/L can inhibit the elimination of H_2S in a BTF (Kim and Deshusses, 2005). However, in the present study, since the recirculating liquid flow was refreshed daily, the highest concentration of sulfate measured was 1.7 g/L. Therefore, it can be concluded that inhibitory effect of sulfate did not occur during the immobilization stage of BTF operation.

The complete performance assessment for this BTF was done at different inlet H_2S concentrations and EBRTs. These experiments showed that whenH_2S loading is increased, either by increasing the inlet H_2S concentration or decreasing EBRT, EC_{max}increases. Jin *et al*. (2005b) have observed 99% removal efficiency at H_2S inlet concentration of 55 ppm and EBRT of 28 seconds. Namini *et al.* (2007) also reported 96% removal efficiency at H_2S inlet concentration of 57 ppm and EBRT of 28 seconds. In this study the RE under the same conditions (C_{in}= 55 ppm and EBRT= 27 s) was 90%. The lower RE obtained in the present study is related to the type of bed and microorganism used in the BTF system. Jin *et al*. (2005b) immobilized activated sludge on synthetic bed and since activated sludge contains a variety of microbial species, including heterotrophic species, biofilm can form better. Also, some other species such as *Pseudomonas putida*can contribute to H_2S removal and these facts can be the reasons for the higher RE in their work. Namini *et al.* (2007) used Lava rock as the microbial support in BTF and the better attachment of *T.thioparus*to this support can be one of the reasons for the better performance obtained in that work. But it should be noted that changes in microbial population in activated sludge and problems of using natural beds such as clogging and pressure drop, affect the performance of system in the long term.

It was also found that, contrary to work with some other packings in which a part of elimination is the result of the adsorption of contaminant onto the bed material, experiments (results not presented) indicated no adsorption of H_2S on the PUF packing inside the BTF; therefore, the H_2S removal obtained can be solely attributed to biodegradation by the bacterial population inside BTF.

The results of removal efficiency of H_2S as a function of loading at different H_2S concentrations emphasized the effect of EBRT on the elimination of H_2S. This is in line with the results of Chung *et al*, (2004) and Namini *et al*. (2007), who demonstrated that the effect of decreasing EBRT on RE is more pronounced than the effect of increase in inlet H_2S concentration.

Moreover, it was found that increasing LRR had a pronounced positive effect on H_2S removal efficiency. The positive effect of increasing LRR on H_2S removal in this work is probably related to the fact that, with increasing LRR, the better liquid distribution creates more homogenous conditions in the bed for the growth and activity of the biofilm. This result is in line with some previous reports (Diks and Ottengraf, 1991; Hartmas *et al.*, 1991; Kim and Deshusses, 2005) and contrary to some others (Chou and Huang, 1997; Jin *et al.*, 2005b; Namini*et al.*, 2007). The latter can be attributed to the different packings, EBRT and inlet H_2S concentrations used in those studies compared to the present one.

A fairly homogeneous H_2S removal along bed height was obtained with the degree of homogeneity increasing with decrease in EBRT. This result is in line with the result obtained by Namini *et al.* (2007) who used a similar BTF with *T. thioparus*but Lava rock as the microbial carrier. Jin *et al*. (2005a) observed that increasing concentration of pollutant leads to a more homogenous removal along the height of the BTF bed. These results show a relatively better homogeneity of H_2S elimination in BTFs at different heights of the bed compared to studies in biofilters (Shojaosadati and Elyasi, 1999; Elias *et al.*, 2002; Martin *et al.*, 2004). This can be attributed to the more homogenous conditions prevailing in the BTF, due to the existence of a circulating liquid flow. The more homogenous H_2S removal in this work, compared to the results reported in biofilters, can also be related to the use of polyurethane foam as microbial support in the present study, since more homogenous pollutant elimination in a BTF with synthetic supports compared to compost has been reported before (Jones *et al.*, 2004).

Finally it was found that during 100 days operation of the BTF, the only H_2S degrading microbial species was *T. thioparus*. The unidendified species of bacteria and fungi probably used the nutrients resulted from cell lysis or microbial extracellular polymeric substances, but since the concentration of organic nutrients cannot be high, their growth is slow and they cannot become the dominant strains in the biofilm structure. Furthermore, the H_2S removal experiments in flask for the contaminating species showed that they are incapable of H_2S removal, and the biodegradation of H_2S in the BTF system used in the present study can be solely attributed to the activity of *T. thioparus*. Kanagawa and Mikami (1989) and Namini *et al.* (2007) had also observed significant heterotrophic contamination in their biofiltration systems using *T. thioparus* for removal of H_2S under non-sterile conditions.

ACKNOWLEDGEMENTS

The authors express their gratitude to Hooshang Moradi for his technical assistance. This work was supported by a grant from the Iranian Research Organization for Science and Technology.

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