Biological elimination of H_2S and NH_3 from wastegases by biofilter packed with immobilized heterotrophic bacteria

Ying-Chien Chung [a], Chihpin Huang [b,*], Ching-Ping Tseng [c]

[a] *Science & Technology Information Center, National Science Council, Taipei, Taiwan, ROC*
[b] *Institute of Environmental Engineering, National Chiao Tung University, 75 Po-ai Street, Hsinchu 3009, Taiwan, ROC*
[c] *Institute of Biological Science and Technology, National Chiao Tung University, Hsinchu, Taiwan, ROC*

Abstract

Biotreatment of various ratios of H_2S and NH_3 gas mixtures was studied using the biofilters, packed with co-immobilized cells (*Arthrobacter oxydans* CH8 for NH_3 and *Pseudomonas putida* CH11 for H_2S). Extensive tests to determine removal characteristics, removal efficiency, removal kinetics, and pressure drops of the biofilters were performed. To estimate the largest allowable inlet concentration, a prediction model was also employed. Greater than 95% and 90% removal efficiencies were observed for NH_3 and H_2S, respectively, irrespective of the ratios of H_2S and NH_3 gas mixtures. The results showed that H_2S removal of the biofilter was significantly affected by high inlet concentrations of H_2S and NH_3. As high H_2S concentration was an inhibitory substrate for the growth of heterotrophic sulfur-oxidizing bacteria, the activity of H_2S oxidation was thus inhibited. In the case of high NH_3 concentration, the poor H_2S removal efficiency might be attributed to the acidification of the biofilter. The phenomenon was caused by acidic metabolite accumulation of NH_3. Through kinetic analysis, the presence of NH_3 did not hinder the NH_3 removal, but a high H_2S concentration would result in low removal efficiency. Conversely, H_2S of adequate concentrations would favor the removal of incoming NH_3. The results also indicated that maximum inlet concentrations (model-estimated) agreed well with the experimental values for space velocities of 50–150 h^{-1}. Hence, the results would be used as the guideline for the design and operation of biofilters. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Hydrogen sulfide; Ammonia; Biofilter; *Arthrobacter oxydans* CH8; *Pseudomonas putida* CH11

1. Introduction

H_2S and NH_3 are irritating, smelly substances with very low odor thresholds: 1.1 ppb for H_2S and 37 ppb for NH_3 (Henry and Gehr, 1980). These two unwanted gases are usually liberated in industrial processes, including food preparation, livestock farming, leather manufacturing and wastewater treatment (Eikum and Storhang, 1986; Ryer-Power, 1991; Yang and Allen, 1994; Chung et al., 1996a). Many technologies have been used to treat malodorous compounds from contaminated air. As regulatory measures move toward more stringent control of malodorous compounds, the demand for cost-efficient air pollution control technology will increase. Currently, biotreatments have drawn great attention, especially biofiltration, because they cost less than conventional methods and have comparable removal efficiency (Leson and Winer, 1991).

In treating exhaust gas, the selection of packing materials and inoculated microorganisms has a decisive effect on the biofilter operation. Recently, utilizing

* Corresponding author. Tel.: +886-3-572-6463; fax: +886-3-572-5958.
E-mail address: cphuang@green.ev.nctu.edu.tw (C. Huang).

immobilized cells as packing materials for wastegas removal has been proved to be very promising (Chung et al., 1996b, 1998). In the case of H_2S removal, heterotrophic bacteria *Pseudomonas putida* CH11 performed better than autotrophic bacteria *Thiobacillus thioparus* CH11, while operating at low inlet H_2S concentration (<20 ppm) over a long-term period (Chung et al., 1996c, d). For NH_3 removal, heterotrophic bacteria *Arthrobacter oxydans* CH8, isolated from piggery wastewater, performs better than autotrophic bacteria *Nitrosomonas europaea*, especially for treating high concentrations of NH_3 (Chung et al., 1997; Chung and Huang, 1998). However, there have been no studies on the biological treatment of H_2S and NH_3 in an air stream simultaneously. The Taiwan EPA sets the ambient air standard at 0.1 and 1 ppm for H_2S and NH_3, respectively. To reach the current H_2S/NH_3 emission standards and forthcoming higher standards in the future, the outlet exhaust must satisfy the current legal standards. Thus, critical operating parameters of the biofilter need to be established as soon as possible.

The objective of this study was to determine the effectiveness of co-immobilized biofiltration technology on gas mixtures of H_2S and NH_3. In this study, a *P. putida* CH11 and *A. oxydans* CH8 co-immobilized biofilter were used to remove a H_2S and NH_3 gas mixture, where *P. putida* CH11 is effective in removing only H_2S and *A. oxydans* CH8 is effective in eliminating only NH_3 (Chung et al., 1996c,d). Various ratios of inlet H_2S/NH_3 gas mixtures were introduced into the biological system to investigate the removal efficiency, mechanism, metabolized products and kinetic parameters of the biofilter. In addition, enzyme kinetic theory was used to develop a model to estimate the maximum inlet concentration for practical application.

2. Materials and methods

2.1. Organism cultivation and medium preparation

The original pure-culture strains of heterotrophic ammonia oxidizer, *A. oxydans* CH8 and heterotrophic sulfur oxidizer *P. putida* CH11 were isolated from swine wastewater (Chung et al., 1996c, 1997). Stock cultures were both grown in nutrient broth at 30°C. The nutrient broth contained yeast extract 5 g/l, tryptone 10 g/l, and dextrose 2 g/l. In all continuous experiments, the inflow medium was used and stored in the nutrient tank. The inflow medium contained glucose 0.2 g/l, KH_2PO_4 1.2 g/l, K_2HPO_4 1.2 g/l, NH_4Cl 0.4 g/l, $MgCl_2 \cdot 6H_2O$ 0.2 g/l, and Fe(III)-citrate 0.01 g/l (C:N = 4:5). The final pH of the medium was adjusted to neutral using 2 N NaOH or HCl.

2.2. Immobilization procedure

A. oxydans CH8 and *P. putida* CH11 were each grown in 100 ml nutrient broth, harvested by centrifugation ($8000 \times g$ for 10 min), and then washed three times with sterile distilled water. The cultures were mixed together with a sterile 4% Na-alginate solution. Then the Na-alginate solution containing the mixture of cells was introduced into a 4% $CaCl_2$ solution using a syringe, which immediately formed 3-mm diameter co-immobilized beads. Flushing with sterile buffer solution for 5 h activated these beads.

2.3. Apparatus and H_2S/NH_3 removal for continuous operation

A schematic of the experimental setup of the lab scale biofilter is shown in Fig. 1. Glass columns (6 cm $\varnothing \times$ 25 cm of working height) were packed with cell-laden Ca-alginate beads on top of a perforated sieve plate fitted at the bottom of the column to ensure the uniform distribution of the inlet gas. The packed volume, dry weight of beads and number of cells initially packed in each column were 0.7 l, 0.28 kg, and 10^{10} cells/g-dry bead, respectively. The column wall contained two sampling ports, 12.5 cm apart, for measuring H_2S and NH_3 concentrations during the experiments. The pressure drop across the reactor was measured using a u-tube water manometer. The $H_2S_{(g)}$ and $NH_{3(g)}$, supplied from separate gas cylinders, were first diluted with compressed air and flowed upwards through the bottom of the biofilter. An inflow medium (see medium preparation) was intermittently re-circulated every 2 h by a peristaltic pump at 25 ml/min to maintain the moisture of the biofilter and supply nutrient to the co-immobilized cells. The peristaltic pump was connected to a spray nozzle to uniformly spray the medium on the surface of filter bed in a counter-flow direction with the influent gas.

In the continuous experiment, the simulated H_2S- and NH_3-containing wastegas was prepared at 1:1 (60 ppm:60 ppm), 1:2 (60 ppm:120 ppm), and 2:1 (120 ppm:60 ppm) by volume/volume. These mixtures were sequentially supplied to the biofilter at 36 l/h (residence time = 72 s) and the operating temperature was controlled at 30°C. The products resulting from the biofilter were also measured during the continuous experiment.

2.4. Bioaerosol analysis

Microorganisms liberated from the biofilter were collected by liquid impingement. The exhaust air evacuated at the top of the biofilter was forced through a 250-ml flask containing 100 ml aseptically distilled water at 72 l/min for 5 h. One ml of the collected solution was inoculated to different media and the cell numbers were
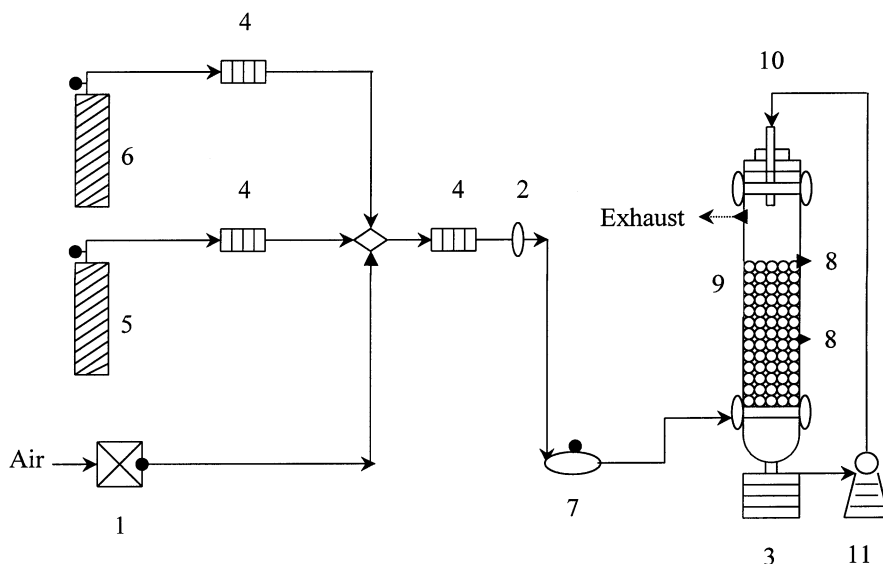
Fig. 1. Schematic of the lab scale biofilter: (1) air compressor; (2) air filter; (3) nutrient tank; (4) flow meter; (5) H_2S gas cylinder; (6) NH_3 gas cylinder; (7) inlet chamber; (8) sampling port; (9) glass column; (10) spray nozzle; (11) peristaltic pump.

determined by plate count method. Potato dextrose agar (PDA) was used to culture fungi, nutrient agar for heterotrophic bacteria, the thiosulfate agar for non-acidophilic *Thiobacilli*, and the modified Waksman agar for acidophilic *Thiobacilli* (Cho et al., 1991). The cell counts of autotrophic ammonia oxidizer were determined by the amount of nitrite produced (Sato et al., 1985). The counts were reported as colony forming units per unit of air (CFU/m^3).

2.5. Kinetic analysis

The H_2S or NH_3 removal rate in the immobilized-cell biofilter was calculated using the following equation derived from the Michaelis–Menten equation (Hirai et al., 1990):

$$\frac{1}{R} = \frac{K_s}{V_m} \times \frac{1}{C_{ln}} + \frac{1}{V_m}, \tag{1}$$

where R (g-S or g-N/day/kg-dry bead) is apparent removal rate; C_{ln} (ppm) equals to $(C_o - C_e)/\ln(C_o/C_e)$, logarithmic means concentration of H_2S or NH_3 in the biofilm; C_o (ppm) the concentration of H_2S or NH_3 in the biofilm at the inlet; C_e (ppm) the concentration of H_2S or NH_3 in the biofilm at the outlet; V_m (g-S or g-N/ day/kg-dry bead) = maximum apparent removal rate, and K_s (ppm) = apparent half-saturation constant. From the linear relationship between $1/C_{ln}$ and $1/R$, V_m and K_s were calculated from the intercept and slope, respectively. In this experiment, the mass-transfer limitation was minimized by restricting the flow rates in the range of 36–72 l/h (residence time: 36–72 s).

When the H_2S oxidation was inhibited due to high H_2S concentration, an inhibition constant K_i, must be incorporated into Eq. (1) as

$$\frac{1}{R} = \frac{K_s}{V_m} \times \frac{1}{C_{ln}} + \frac{1}{V_m} + \frac{C_{ln}}{V_m \times K_i}. \tag{2}$$

At low inlet concentration, Eq. (2) can be simplified back to Eq. (1). However, at high inlet concentration, Eq. (2) becomes

$$\frac{1}{R} = \frac{1}{V_m} + \frac{C_{ln}}{V_m \times K_i}. \tag{3}$$

2.6. Model prediction of maximum inlet concentration

If desired effluent concentrations of H_2S and NH_3 at the biofilter outlet were presumed to be 0.1 and 1 ppm, respectively, then the maximum inlet concentrations of different pollutants (e.g., H_2S and NH_3) needed to attain the desired effluent concentrations could be calculated by the following kinetic analysis. Assuming the wastegas into the biofilter was plug flow, the following equation was applied (Tiwaree et al., 1992):

$$\frac{-dC}{dl} = \left(\frac{V_m \times C}{K_s + C} \right) \times \left(\frac{S_a}{F} \right) \times \alpha, \tag{4}$$

$$\frac{-dC}{dl} = \left(\frac{V_m \times C}{K_s + C} \right) \times \left(\frac{1}{L \times SV} \right) \times \alpha, \tag{5}$$

where C (ppm) is the concentration of H_2S or NH_3 in the biofilm, l (m) the column length, SV (d^{-1}) the space

velocity $F (S_a L)^{-1}$, F (m^3 d^{-1}) the gas flow rate, S_a (m^2) the column cross-section, L (m) the packing height, α is the conversion coefficient (kg-dry bead ppm/g-S or g-N).

Integrating Eq. (5) under the condition of $C = C_o$ at $l = 0$, $C = C_e$ at $l = 0$, $C = C_e$ at $l = L$ Eq. (6) was obtained.

$$\frac{\alpha}{SV \times (C_o - C_e)} = \left(\frac{K_s}{V_m}\right) \times \left(\frac{1/(C_o - C_e)}{\ln(C_o/C_e)}\right) + \frac{1}{V_m}. \quad (6)$$

Setting $C_{ln} = (C_o - C_e)/\ln(C_o - C_e)$, Eq. (6) was transferred as follows:

$$SV = \frac{\alpha \times V_m}{(C_o - C_e)} \times \left(\frac{C_{ln}}{C_{ln} + K_s}\right). \quad (7)$$

Setting C_e at 0.1 ppm for H$_2$S concentration or 1 ppm for NH$_3$ concentration in Eq. (7), the maximum inlet C_o can be estimated at various space velocities.

As the Michaelis–Menten equation considered gas concentration in the biofilm rather than in the gas phase, the concentrations of H$_2$S and NH$_3$ in the biofilm were obtained by Henry's law. Henry's law constants of H$_2$S and NH$_3$, determined by the method presented by Shinabe et al. (1995), depended strongly on the pH of the liquid medium. The concentration of undissociated H$_2$S in the liquid can be calculated from pH and their acidity constants as below

$$H_2S_{biofilm} = \frac{H_2S_T \times [H^+]^2}{[H^+]^2 + [H^+]K_1 + K_1K_2},$$

where H$_2$S$_T$ (ppm) is the total H$_2$S concentration in the biofilm and K_1, K_2 are the dissociation constants for H$_2$S.

Also the concentration of undissociated NH$_3$ in the liquid can also be obtained from the following equation:

$$NH_{3biofilm} = \frac{NH_{3T} \times K_a}{[H^+] + K_a},$$

where NH$_{3T}$ (ppm) is the total NH$_3$ concentration in the biofilm and K_a is the dissociation constant for NH$_3$.

2.7. Analytical methods

Inlet and outlet H$_2$S gas concentrations of the biofilter were measured either continuously by a single point monitor (MDA Scientific) ranging from 50 to 1500 ppb, or periodically by gas detector tubes (GASTEC) ranging from 1 to 100 ppm. Inlet and outlet NH$_3$ gas concentrations in the biofilter were measured either continuously by a single point monitor (MDA Scientific) in the range of 0.1–10 ppm, or periodically by gas detector tubes (GASTEC) in the range of 5–100 ppm. In all continuous experiments, H$_2$S/NH$_3$ concentration recorded as the variation of H$_2$S/NH$_3$ concentration was within ±5% in 2 h. Totally 12 data were recorded and

then averaged to be the H$_2$S or NH$_3$ outlet concentration. Samples were taken 48 times per day for the periodic measurement with the gas detector tubes. When the pseudo-steady-state was reached, samples were then taken 6 times per hour. The chemical composition of circulation solution was also determined. Nitrate, nitrite and sulfate concentrations in the solution were measured by ion chromatography (Dionex 4500i). Ammonium and sulfide ion concentrations were determined using an ion-specific electrode. Sulfite was determined by titration using a standard potassium iodide–iodate titrant and a starch indicator (APHA, 1992). Elemental sulfur was determined by reaction with cyanide to produce thiocyanate, which was quantitated as Fe(SCN)$_6^{3-}$ (Schedel and Truper, 1980).

3. Results and discussion

3.1. H$_2$S/NH$_3$ removal efficiency in continuous operation

The removal efficiencies for different ratios (e.g., 1:1, 1:2, and 2:1) of H$_2$S/NH$_3$ gas mixtures at various time are illustrated in Fig. 2. A ratio of 1:1 for inlet H$_2$S/NH$_3$ was used during the first 7-day period, then a ratio of 1:2 was used for the following 7-day period, and a ratio of 2:1 was used for the last 7-day period. During the operating period, the circulation solution with fresh medium was replaced at day 14. When H$_2$S and NH$_3$ were mixed in a ratio of 1:1, both the removal efficiencies for H$_2$S and NH$_3$ increased with operating time. Moreover, the NH$_3$ removal efficiency reached a maximum of 98.5%. As pointed out in the literature, the NH$_3$ removal by *Arthrobacter* sp. was enhanced if other heterotrophic bacteria existed (Prosser, 1989). Thus, co-immobilized cells performed better on NH$_3$ removal than *A. oxydans* CH8 alone did at a similar condition (Chung et al.,
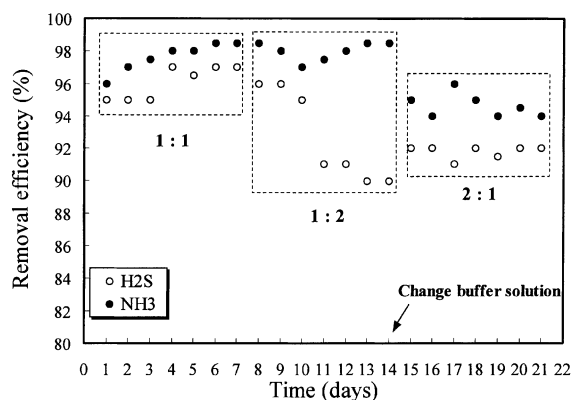

Fig. 2. Relationship between the removal efficiency and operating time at different ratios (1:1, 1:2, 2:1) of H$_2$S/NH$_3$ gas mixtures.

1997). When H_2S and NH_3 were mixed in a ratio of 1:2, the high NH_3 concentration (120 ppm) inhibited the H_2S metabolism of *P. putida* CH11 and the removal efficiency dropped to a value of 90% on the 14th day. Analyzing the pH in the biofilter, we found slight acidification of the biofilter: the pH value was 5.8. The activity of the *P. putida* CH11 was thus reduced, so the metabolic capacity for H_2S was reduced. In the case of mixing H_2S and NH_3 with a ratio of 2:1, high H_2S concentrations (120 ppm) apparently inhibited H_2S metabolism by *P. putida* CH11 in comparison to the case of 1.1, and only 92% removal efficiency was achieved at the end of the operation. The H_2S metabolism by heterotrophic sulfur-oxidizing bacteria was a detoxicant process (Chung et al., 1996c), and input of high H_2S concentration would significantly affect the H_2S removal efficiency. Similarly, NH_3 removal efficiency also decreased from 98% to 94% with a higher H_2S concentration. These results indicate that the effective range of H_2S concentration for treating H_2S/NH_3 gas mixtures is limited to medium inlet concentration less than 120 ppm. The long-term experiment (about 100 days) was also conducted to remove H_2S and NH_3 with a ratio of 1:1 and similarly high efficiency was obtained (data not shown).

3.2. Product analysis

Products from various ratios of the H_2S/NH_3 gas mixtures are indicated in Tables 1 and 2. The metabolic products for the organic N and S were omitted in the experiments. As indicated in Table 1, the conversion ratio of $SO_3^=$ increased remarkably (from 10.0% to 20.9%) as inlet NH_3 concentration increased. As mentioned above, the slight acidification of the biofilter was found when higher NH_3 concentration was introduced. The drop in pH of the biofilter resulted in lower microbial degradation potentials and the accumulation of incompletely oxidized product ($SO_3^=$). When H_2S and NH_3 were mixed at a 2:1 ratio, high H_2S concentration (0.16 mg/l in aqueous phase) inhibited the activity of the *P. putida* CH11 which resulted in the increase in the ratio of $S^=$ (from 9.4% to 20%). As *P. putida* CH11 was poisoned by high H_2S concentration, excess residual $S^=$ accumulated (Chung et al., 1996c). The accumulation of $S^=$ might further have suppressed the nitrification of the ammonia-oxidizing bacteria (Joye and Holibaugh, 1995) and therefore caused the reduction in NH_3 removal efficiency (Fig. 2). Table 2 shows that the products and their conversion ratios were unchanged while the inlet NH_3 concentration was 60 ppm (in the cases of 1:1 and 2:1). However, high NO_2^- concentration was found in the biofilter when the inlet NH_3 concentration was raised to 120 ppm (0.069 mg/l in aqueous phase). Similarly, the acidic product resulted in slight acidification of the biofilter and a decrease in the activity of the sulfur-oxidizing bacteria, and finally caused the decrease in the H_2S removal efficiency (Fig. 2).

3.3. Bioaerosol analysis

As biofilters contain tremendous numbers of microorganisms, it is necessary to assess the environmental risk associated with the bacteria released from the biofilter when large quantities of wastegases are treated.

Table 3 shows the numbers of viable microorganisms in the outlet exhaust when the biofilter was utilized continuously for 7, 14, and 21 days with different ratios of H_2S/NH_3 gas mixtures. Microorganisms investigated included fungi, heterotrophic bacteria, neutrophic

Table 1
Metabolic products of the H_2S at different ratios of H_2S/NH_3 supply

Mixture ratio (ppm/ppm)	$SO_4^=$ produced (g-S/kg-bead)	S^0 produced (g-S/kg-bead)	$SO_3^=$ produced (g-S/kg-bead)	$S^=$ produced (g-S/kg-bead)
1:1[a]	0.29 (20.9%)	0.83 (59.7%)	0.14 (10.0%)	0.13 (9.4%)
1:2	0.16 (11.9%)	0.75 (56.0%)	0.28 (20.9%)	0.15 (11.2%)
2:1	0.37 (12.5%)	1.49 (50.2%)	0.51 (17.2%)	0.60 (20.0%)

[a] 1:1 equals 60:60 (ppm/ppm).

Table 2
Metabolic products of the NH_3 at different ratios of H_2S/NH_3 supply

Mixture ratio (ppm/ppm)	NH_4^+ produced (g-N/kg-bead)	NO_2^- produced (g-N/kg-bead)	NO_3^- produced (g-N/kg-bead)
1:1[a]	0.02 (2.3%)	0.78 (90.7%)	0.06 (7.0%)
1:2	0.05 (3.0%)	1.50 (89.3%)	0.13 (7.7%)
2:1	0.02 (2.4%)	0.75 (90.4%)	0.06 (7.2%)

[a] 1:1 equals 60:60 (ppm/ppm).

Table 3
Bioaerosol analysis in the outlet exhaust of the biofilter

Mixture ratio	Type of microorganism				
	Heterotrophic bacteria	Fungi	Neutrophic sulfur oxidizer	Acidophilic sulfur oxidizer	Autotrophic nitrifying bacteria
1:1[a]	<ND	<ND	<ND	<ND	<ND
1:2	14	<ND	<ND	<ND	<ND
2:1	18	5	<ND	<ND	<ND

[a] ND < 5 CFU/m^3.

sulfur-oxidizing bacteria, acidophilic sulfur-oxidizing bacteria, and chemoautotrophic nitrifying bacteria. Apparently, as microorganisms were immobilized in Ca-alginate, the exhaust contained only small amounts of bacteria (less than 19 CFU/m^3 in all cases). This indicates that the microorganisms were well immobilized in Ca-alginate. These bioaerosol concentrations were far smaller than those released from a peat biofilter (Hartikainen and Martikainen, 1996). In other words, the environmental risk of bioaerosol released through immobilized technology is minimal and this system can be considered safe if placed close to populated areas.

3.4. Pressure drop

The influence of surface load on pressure drop is shown in Fig. 3. In this experiment, the flow rate was raised gradually from 36 to 180 l/h and the temperature was maintained at 30°C. When the variation of outlet H$_2$S/NH$_3$ concentration was within ±5%, a new flow rate was selected. The experiment was initiated after three-month acclimation. The pressure drop ranged from 0.75 to 1.8 cm of H$_2$O. The data correspond well with other biofilters study that utilized compost, pine bark, and a bulking agent as the packing media (Leson and Winer, 1991; Lackey et al., 1998). Inspection of the
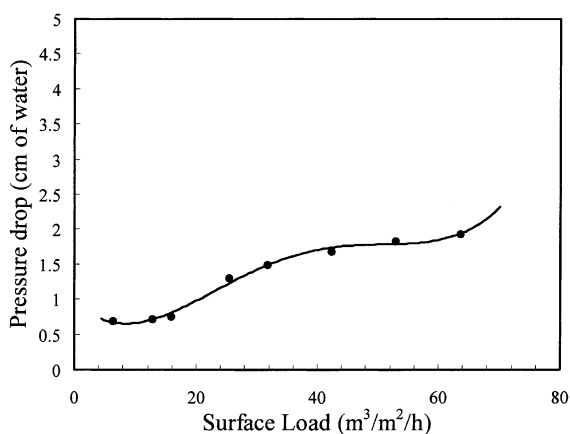
figure reveals that pressure drop of the biofilter increases with increasing flow rate. However, the pressure drop across the biofilter increased in a non-linear manner. This may be attributed to abundant biomass or sulfur (product) accumulation. Therefore, the heterotrophic biofilter exhibits no excellent dispersion characteristics compared with the autotrophic biofilter (Chung et al., 2000).

3.5. Kinetic analysis

Fig. 4(a) indicates that a low NH$_3$ concentration (60 ppm) does not effect the metabolism of H$_2$S (5–65 ppm) by *P. putida* CH11, but a high NH$_3$ concentration (120 ppm) results in a negative effect. The K_s and V_m values
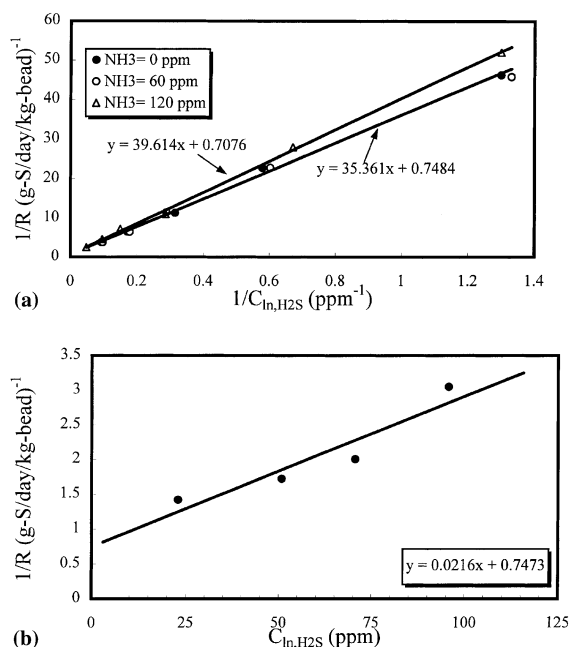
(a)

(b)

Fig. 4. Relationship between $1/R$ and $1/C_{\mathrm{ln,H_2S}}$ of H$_2$S degradation in the biofilters: (a) effect of NH$_3$ concentrations on H$_2$S removal by *P. putida* CH11 at H$_2$S (5–65 ppm) and NH$_3$ (0, 60 or 120 ppm); (b) effect of high H$_2$S concentrations on H$_2$S removal by *P. putida* CH11 at H$_2$S (120–200 ppm) and NH$_3$ 60 ppm.

Fig. 3. Profile of pressure drop vs surface load for the heterotrophic biofilter.

were calculated to be 47.2 ppm and 1.33 g-S/day/kg-dry bead, respectively, at the NH_3 concentration 0 or 60 ppm. In addition, the K_s and V_m values were calculated to be 56.0 ppm and 1.41 g-S/day/kg-dry bead, respectively, at the NH_3 concentration 120 ppm. Generally, if we inferred a physical meaning for K_s analogous to enzymatic kinetics, a higher of K_s value indicated a lower enzymatic affinity for H_2S. Thus, high NH_3 concentrations affect H_2S removal by the biofilter. As mentioned in earlier sections, we have found that the biofilter caused acidification in this case (120 ppm of NH_3).

In the case of treating high H_2S concentration (120–200 ppm), irrespective of the fluctuating influent NH_3 concentration of the biofilter, the removal efficiency of H_2S was far from ideal (data not shown). This is possibly due to the poisoning of *P. putida* CH11, which was responsible for the metabolism of H_2S. Hence, by using Eq. (3) and plotting the logarithmic mean concentration of H_2S (C_{ln,H_2S}) vs the reciprocal of the removal rate (1/R) the inhibition constant (K_i) could be obtained. According to the regression equation obtained, the K_i is 34.6 ppm. Moreover, the maximum removal rate (1.34 g-S/day/kg-dry bead) is similar to the value (1.33 g-S/day/kg-dry bead) obtained from Fig. 4(a).

Fig. 5 illustrates the kinetic analysis of the NH_3 removal by *A. oxydans* CH8 in the range of 5–65 ppm NH_3 under various H_2S concentrations. Interestingly, adequate H_2S concentration (60 ppm) favored the metabolism of NH_3 by *A. oxydans* CH8 compared with the H_2S-free inlet. In contrast, excess H_2S concentration (120 ppm) decreased NH_3 removal. According to the regression analysis, the saturation constants (K_s) of the NH_3 metabolism by the biofilter under different H_2S concentrations were 70.9, 62.4 and 72.8 ppm (at $H_2S = 0$, 60, 120 ppm). Here, low saturation constant indicates higher affinity to the substrate (NH_3).

3.6. Model prediction

To establish the operation principle, the enzymatic kinetic theory is utilized to develop a model as well as to predict the maximum H_2S and NH_3 inlet concentrations under different space velocities (residence time). Here, we assume an emission limit of 0.1 and 1.0 ppm for H_2S and NH_3, respectively. We supply the biofilters with various ratios of inlet gas mixtures and progress till the effluent concentrations exceeded our limits. Fig. 6 illustrates the experimental data and model prediction for the profiles of maximum inlet H_2S and NH_3 concentrations. The figure indicates that experimental values are smaller than those predicted by the model when space velocity exceeds 150 h^{-1}. In fact, the experimental values are smaller than the predicted values by 11–25% for H_2S and 14–20% for NH_3, respectively. This suggests that mass transfer under high space velocity limit the removal capacities of the biofilter.

With space velocities ranging between 50 and 150 h^{-1} (residence time: 45–15 s), the allowable maximum inlet concentration increases as space velocity decreases. In addition, the experimental values and the model-estimated values agree well within this range of space velocities. Therefore, this suggests that mass transfer is not the rate-determining step under low space velocities. Note that the maximum inlet H_2S concentration in the biofilm was about 121 ppm when the space velocity was below 50 h^{-1} and the value was very different from that estimated by the model. The possible reason is because high H_2S concentration damages the sulfur-oxidizer bacteria and therefore causes the discrepancy compared to the model estimation.
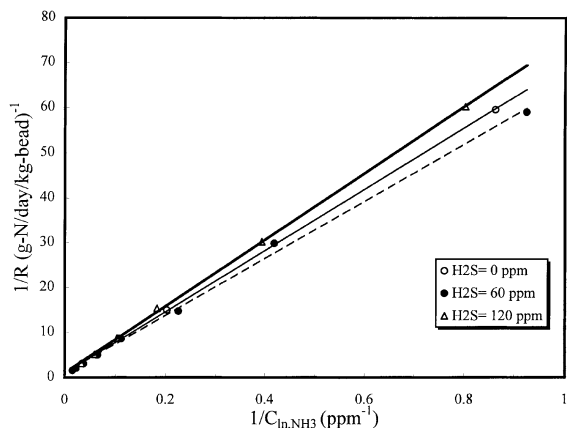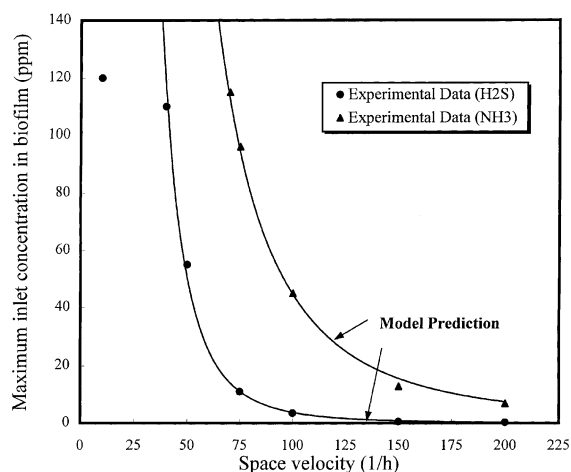

Fig. 5. Relationship between 1/R and $1/C_{ln,NH_3}$ of NH_3 degradation in the biofilter. Conditions: NH_3 (5–65 ppm) and H_2S (0, 60 or 120 ppm).

Fig. 6. Relationship between the maximum inlet concentration and space velocity for H_2S and NH_3 removal by the heterotrophic biofilters for meeting the outlet H_2S 0.1 ppm and NH_3 1 ppm requirements.

4. Conclusions

The results of our experiments have indicated that the biofilter successfully handled the gas mixture of H_2S and NH_3 within the 5–65 ppm range, showing removal efficiencies greater than 96% under these conditions. H_2S and NH_3 at high concentrations were observed to be inhibitory substrates for H_2S removal. H_2S of moderate concentrations favored NH_3 removal. Conversely, high H_2S concentrations resulted in low removal efficiency. The phenomenon was explained by kinetic analysis and the related parameters were determined. Results from the bioaerosol analysis indicated that the immobilized cell biofilter caused low environmental impact and can, therefore, be located close to populated areas. Low-pressure drop hinted the new type of biofilter is a potentially feasible and cost-effective technology. The results also indicated that model-estimated maximum inlet concentrations agreed well with the experimental values, while the space velocity was within the range of 50–150 h^{-1}, and therefore they could be used as the guideline for the operation of the biofilter. Excellent performance ranging from median target wastegas concentration indicates that the heterotrophic biofilter can be feasibly applied to the livestock farming and wastewater treatment.

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