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Biotreatment of H_2S- and NH_3-containing waste gases by co-immobilized cells biofilter

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Abstract

Gas mixture of H_2S and NH_3 in this study has been the focus in the research area concerning gases generated from the animal husbandry and the anaerobic wastewater lagoons used for their treatment. A specific microflora (mixture of *Thiobacillus thioparus* CH11 for H_2S and *Nitrosomonas europaea* for NH_3) was immobilized with Ca-alginate and packed inside a glass column to decompose H_2S and NH_3. The biofilter packed with co-immobilized cells was continuously supplied with H_2S and NH_3 gas mixtures of various ratios, and the removal efficiency, removal kinetics, and pressure drop in the biofilter was monitored. The results showed that the efficiency remained above 95% regardless of the ratios of H_2S and NH_3 used. The NH_3 concentration has little effect on H_2S removal efficiency, however, both high NH_3 and H_2S concentrations significantly suppress the NH_3 removal. Through product analysis, we found that controlling the inlet ratio of the H_2S/NH_3 could prevent the biofilter from acidification, and, therefore, enhance the operational stability. Conclusions from bioaerosol analysis and pressure drop in the biofilter suggest that the immobilized cell technique creates less environmental impact and improves pure culture operational stability. The criteria for the biofilter operation to meet the current H_2S and NH_3 emission standards were also established. To reach Taiwan's current ambient air standards of H_2S and NH_3 (0.1 and 1 ppm, respectively), the maximum inlet concentrations should not exceed 58 ppm for H_2S and 164 ppm for NH_3, and the residence time be kept at 72 s.

Keywords: Hydrogen sulfide; Ammonia; Biofilter; *Thiobacillus thioparus* CH11; *Nitrosomonas europaea*

1. Introduction

Recently, people in Taiwan have encountered numerous air pollution problems, especially those associated with bad odor. It has been reported that more than 300 substances can cause bad odor (Ikeda et al., 1980). Among these substances, NH_3 (ammonia) and H_2S (hydrogen sulfide) are the ones that often exist in our surroundings. Ammonia is colorless, but irritant and smelly, while H_2S is corrosive, extremely toxic, and also smelly. Large amounts of NH_3 and H_2S are generated and released from industrial processes, such as petrochemical refining, metallurgy, food preparation, wastewater treatment, and treatment of fuels (Eikum and Storhang, 1986; Ryer-Power, 1991; Yang and Allen, 1994). Excess amounts of NH_3 and H_2S have to be removed for the sake of safety and health (Buchnan and Gibbons, 1974; Prosser, 1989) and also for the reduction of environmental impacts, such as greenhouse effect, acid rain, and eutrophication. Currently, the Taiwan EPA sets the ambient air standards at 1 and 0.1 ppm for

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NH_3 and H_2S, respectively. Based on the cost for the equipment and operation, biological treatment is believed to be the most economical option for treating NH_3 and H_2S. Recently, immobilized-cell technology has been employed in wastewater treatment. Its advantages include high microorganism content, prevention of the microbial loss, high environmental endurance, and high operational stability. In the study of exhaust gas treatment, Chung et al. (1996a,b) proved the value of the immobilized-cell technology because of its high removal efficiency, high removal potential, and high operational stability.

In treating exhaust gas, the selection of the right kind of bacteria is very important. Although activated sludge is often inoculated into the bioreactor, it requires at least three weeks for acclimation (Ottengraf and Van Den Oever, 1983). The use of pure culture has drawn great attention, because it can shorten the start-up time and increase the removal efficiencies of the reactor (Sublette and Sylvester, 1987).

Immobilized *Thiobacillus thioparus* CH11 biofilter has been successfully applied on removing H_2S alone (Chung et al., 1996a,b). It can treat H_2S gas from high concentration (100 ppm) to low concentration (10 ppm). On the other hand, immobilized *Nitrosomonas europaea* was discovered capable of treating NH_3 gas of various concentrations (10–100 ppm), and exceeding 97.5% removal after a 4-day operation (Chung and Huang, 1998). This high removal efficiency can last for three months if the pH value is adequately controlled. As NH_3 and H_2S often coexist in real situation, high concentration of these two gases may damage *N. europaea* and thus decrease the NH_3 removal efficiency (Prakasam and Loehr, 1972; Hunik et al., 1992; Joye and Holibaugh, 1995). Hence, how to optimize the operation of the immobilized-cell biofilter to simultaneously remove NH_3 and H_2S is the key to the success of this technology.

In our previous studies, we have discovered that *N. europaea* is very effective in removing NH_3 alone and *T. thioparus* CH11 is very good at eliminating H_2S alone (Chung et al., 1996a,b; Chung and Huang, 1998). Therefore, we adopted *N. europaea* and *T. thioparus* CH11 co-immobilized biofilter in this study. By supplying NH_3/H_2S gas mixtures of various ratios, we examined the removal efficiency, removal mechanisms, metabolized products, and kinetic parameters of the co-immobilized biofilter. In addition, the likely encountered operational problems such as pressure drop and contamination by other bacteria in field applications, as well as the design of optimal operation conditions were also discussed.

2. Materials and methods

2.1. Organism cultivation and medium preparation

The original pure-culture strain of autotrophic ammonia oxidizer, *N. europaea* ATCC 19718 was obtained from the American Type Culture Collection. Their stock cultures were grown in the ammonia medium in the dark at 30°C. Autotrophic sulfur-oxidizing *T. thioparus* CH11 was isolated from the swine wastewater (Chung et al., 1996b), and the stock cultures were grown in the thiosulfate medium at 30°C. For all continuous experiments, the inflow medium was drawn directly from the nutrient tank. The basic media compositions are listed in Table 1.

2.2. Immobilization procedure

Both *N. europaea* grown in 100 ml ammonia medium and *T. thioparus* CH11 grown in 100 ml thiosulfate medium were harvested by centrifugation (7500×g for 10 min), and then washed three times with sterile distilled water. These organisms were mixed together with a sterile 4% (w/v) Na-alginate solution. With a syringe, this Na-alginate solution was dropped into a 4% (w/v) $CaCl_2$ solution and immediately formed 3 mm-diameter beads. These co-immobilized beads were activated by flushing with sterile buffer solution for 5 h. The initial biomass concentrations of each specie in the beads were 10^5 cells/g-bead.

Table 1
Composition of basic media for cultivation of microorganisms and continuous experiments

Medium	Ammonia medium		Thiosulfate medium		Inflow medium	
Composition g l^{-1}	$(NH_4)_2SO_4$	3.3	NH_4Cl	0.4	NH_4Cl	0.1
	$MgSO_4$	0.25	$MgCl_2 \cdot 6H_2O$	0.2	$MgCl_2 \cdot 6H_2O$	0.2
	NaH_2PO_4	0.78	NaH_2PO_4	1.2	NaH_2PO_4	0.78
	Na_2HPO_4	0.89	Na_2HPO_4	1.2	Na_2HPO_4	0.89
	$CaCl_2$[a]	0.74	$Na_2S_2O_3 \cdot 5H_2O$	8.0	$CaCl_2$[a]	0.74
	$FeSO_4 \cdot 7H_2O$[a]	2.5	$FeSO_4 \cdot 7H_2O$	0.01	$FeSO_4 \cdot 7H_2O$	0.01
	$CuSO_4$[a]	0.08			$CuSO_4$[a]	0.08
pH		7.5		7.0		7.5
Temp. (°C)		30		30		30

[a] Unit: mg l^{-1}.

2.3. Apparatus and H_2S/NH_3 removal for continuous operation

A schematic of the experimental set-up of the lab scale biofilter is shown in Fig. 1. Glass columns (60 mm $\phi \times 25$ 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 numbers of cells initially packed in each column were 0.7 l, 0.28 kg, and 10^{10} cells/g-bead, respectively. Four ports, 12.5 cm in spacing, were drilled along the column for sampling. The flow meter and valve were used for monitoring and controlling the gas flow through the reactor. The pressure drop across the reactor was measured with a u-tube water manometer. The $H_2S_{(g)}$ and $NH_{3(g)}$, supplied from separate gas cylinders, were first diluted with compressed air, passed a air filter (pore size 0.2 μm, LIDA 3000-06, Made in USA) and flowed upwards through the bottom of the biofilter. An inflow medium (composition shown in Table 1) was re-circulated by a peristaltic pump at a flow rate of 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 the filter bed. During the entire experiments, aluminum foil was wrapped around the column to prevent photo-inhibition.

In the continuous experiment, the simulated H_2S- and NH_3-containing waste gas was prepared at 1:1 (60:60), 1:2 (60:120), and 2:1 (120:60) by concentration (ppm/ppm). These mixtures were supplied to the biofilter at a flow rate of 36 l/h (residence time = 72 s) and the operating temperature was controlled at 30°C.

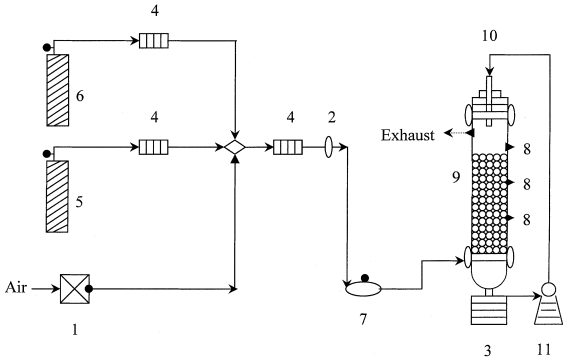

Fig. 1. Schematic of the lab scale biofilter: (1) air compressor; (2) air filter; (3) nutrient tank; (4) flow meter; (5) H_2S gas cylinder; (6) NH_3 gas cylinder; (7) inlet chamber; (8) sampling port; (9) glass column; (10) spray nozzle; (11) peristaltic pump.

2.4. Bioaerosol analysis

Microorganisms released from the biofilter were collected by liquid impingement. The air that escaped from the top of the biofilter was forced through a 250 ml flask containing 80 ml aseptically distilled water at 45 l/min for 10 h. One ml of the collected solution was inoculated to different media and the numbers of cells were determined by the serial dilution method. The PDA medium was used for fungi, the nutrient broth medium for heterotrophic bacteria, the thiosulfate medium for non-acidophilic *Thiobacilli*, and the modified Waksman medium 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 in air (CFU/m^3).

2.5. Kinetic analysis

The H_2S and NH_3 removal rate in the immobilized-cell biofilter were 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-bead) – apparent removal rate; $C_{ln}(\text{ppm}) = (C - C_e)/\ln(C_o/C_e)$, logarithmic mean concentration of H_2S or NH_3 at the inlet and outlet of the biofilter; V_m (g-S or g-N/day/kg-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 slope and intercept. In this experiment, the flow rates were controlled in the range of 36–72 l/h to minimize the mass-transfer limitation.

When the NH_3 oxidation was inhibited due to high NH_3 concentration (>120 ppm), 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 (5–65 ppm), the Eq. (2) can be simplified back to Eq. (1). However, at high inlet concentration (120–200 ppm), Eq. (2) becomes Eq. (3):

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

2.6. Design criteria of scale-up biofilte

The concentrations of H_2S and NH_3 at the biofilter outlet were targeted at 0.1 ppm and 1 ppm. The maximum inlet concentrations and critical H_2S and NH_3

loads to meet this effluent concentration were determined at various space velocity (or residence time) according to the following equation (Chung et al., 1998).

$$\mathrm{SV} = \frac{\alpha}{(C_o - C_e)} \times V_m \times \frac{C_{ln}}{(K_s + C_{ln})}$$

or

$$\frac{(C_o - C_e)}{\theta} = \alpha \frac{V_m \times C_{ln}}{(K_s + C_{ln})}, \quad (4)$$

where SV is the $(d^{-1}) = F \cdot (S_a \cdot L)^{-1}$; F the gas flow rate (m^3/day); S_a the column cross-section (m^2); L the packing height (m); θ the residence time (s); C_o the inlet concentration (ppm); C_e the outlet concentration (ppm); and α is the conversion coefficient (kg-bead ppm /g-S or g-N). Let C_e be 0.1 or 1 ppm in Eq (4), and the maximum C_o can be estimated at various residence times. The loading rate (g-S or g-N/m^3-h) of the biofilter can be obtained from Eq (5).

$$\mathrm{Load} = \frac{\mathrm{SV} \times C_o}{\alpha}. \quad (5)$$

2.7. Analytical methods

Inlet and outlet H_2S 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 60 ppm. Inlet and outlet NH_3 gas concentrations were measured either continuously by a single 0 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_2S/NH_3 concentration was recorded as the variation of H_2S/NH_3 concentration was within ±5% in 2 h. The total 12 data were recorded and then averaged to be the H_2S or NH_3 outlet concentration. Samples were taken 48 times per day for the periodic measurement with the gas detector tubes. The chemical composition of the cycling solution was also determined. Nitrate, nitrite and sulfate concentrations in the solution were measured by ion chromatography (Dionex 4500i). Ammonium was determined using an ion-specific electrode. Sulfite was determined by titration using a standard potassium iodide-iodate titrant and a starch indicator (APHA, 1992). Sulfide was determined using an ion-specific electrode. Elemental sulfur was determined by reaction with cyanide to produce thiocyanate, which was quantitated as $Fe(SCN)_6^{3-}$ (Schedel and Truper, 1980). The pH value in the circulating solution was measured 96 times per day. The data were obtained from two or more duplicate tests.

3. Results and discussion

3.1. H_2S/NH_3 removal efficiency in continuous operation

The removal efficiencies for different ratios (e.g., 1:1, 1:2, and 2:1) of H_2S/NH_3 gas mixtures at various times are illustrated in Figs. 2A and B. When H_2S and NH_3 were mixed in a ratio of 1:1, the optimum set of removal efficiency of H_2S and NH_3 occurred at around the seventh day (e.g., 98% for H_2S and 99% for NH_3) and then slightly decreased toward the end of the experiment. The removal efficiencies for H_2S and NH_3 still remained above 95% at the end of the two-month treatment (data not shown). The reduction in efficiency may be caused by the conversion of H_2S to $SO_4^{=}$ by *T. thioparus* CH11 and the conversion of NH_3 to NO_2^- by *N. europaea*. Both $SO_4^{=}$ and NO_2^- cause the acidic condition of the system, resulting in the decrease in the removal efficiencies of the biofilter. During the entire period, the fluctuation of pH was between 7.5 and 6.9. When H_2S and NH_3 were mixed in a ratio of 1:2, high NH_3 concentration (120 ppm), surprisingly, did not inhibit the H_2S metabolism by *T. thioparus* CH11. On the contrary, it enhanced the

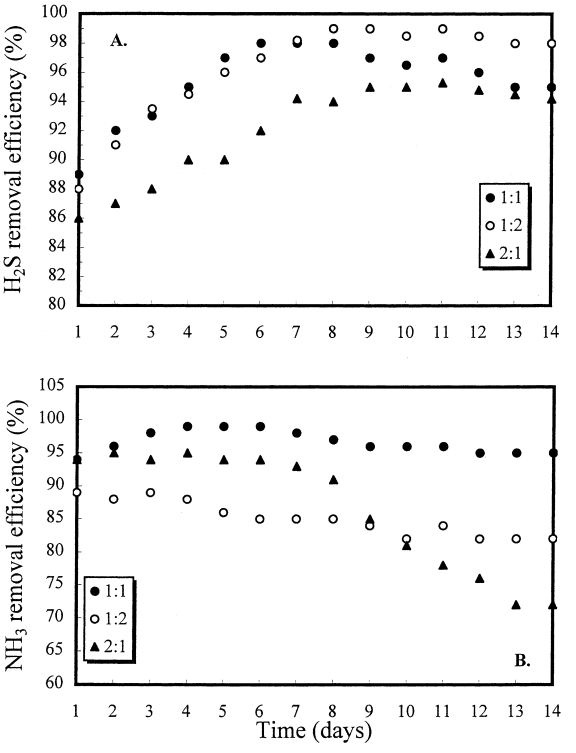

Fig. 2. Relationships between the removal efficiencies and operating time at different ratios (1:1, 1:2, 2:1) of H_2S/NH_3 gas mixtures. (A) H_2S removal efficiency (B) NH_3 removal efficiency. Conditions: 1:1 (60:60), 1:2 (60:120), and 2:1 (120:60) by concentration ppm/ppm; Superficial gas velocity = 36 l/h.

H_2S metabolism when compared to the result of 1:1 ratio, as shown in Fig. 2A. The H_2S removal efficiency increased with operating time and reached as high as 99%. The reason for this enhancement may be that the ample supply of nitrogen promotes the metabolic activity of *T. thioparus* CH11 (Starkey, 1934). By measuring the pH of the biofilter, we found that the decrease in pH was insignificant regardless of the generation of acid products (pH ranging 7.5–7.2). Thus, the high H_2S removal efficiency maintained in the biofilter could be due to the neutralization reaction between $NH_{3(aq)}$ and $SO_4^=$, which prevents the occurrence of acidification. In the case of mixing H_2S and NH_3 with a ratio of 1:2, the removal efficiency of NH_3 is only about 85%. The lower removal efficiency may be caused by the inhibition of the *N. europaea* at higher NH_3 concentration (e.g., 120 ppm) (Hunik et al., 1992). When H_2S and NH_3 were mixed in a ratio of 2:1, the H_2S removal efficiency increased during the operation period and reached a maximum value of 95%. However, at such high H_2S concentration (120 ppm), the NH_3 removal was significantly suppressed and decreased down to 72%. This dramatic reduction in NH_3 removal (Fig. 2B) can be explained by two reasons. The high concentrations of H_2S and the low pH of the biofilter, which dropped to 6.5 after a 9-day operation, hinder the nitrification of *N. europaea* (Joye and Holibaugh, 1995).

3.2. Product analysis

Mass balances of sulfur and nitrogen in the biofilter at different ratios of H_2S/NH_3 supply are listed in Tables 2 and 3. As indicated in Table 2, when 60 ppm of H_2S (in cases of 1:1 and 1:2) was supplied for 14 days, product varieties and their conversion ratios remained unchanged regardless of the NH_3 concentration. However, when the inlet H_2S concentration was increased to 120 ppm, the production ratio of $S^=$ (8.0%) increased, which suggested the incomplete H_2S metabolism of *T. thioparus* CH11 at high H_2S concentration (Table 2). As a result, the residual $S^=$ might suppress the nitrification of *N. europaea* (Joye and Holibaugh, 1995). This is confirmed by the significantly lower NH_3 removal efficiency in the case of a H_2S/NH_3 mixing ratio of 2:1, (Fig. 2B). Table 3 shows that the main product of NH_3 oxidation by the biofilter was NO_2^-. Because only NO_2^- was produced from the *Nitrosomonas*, the small amount of NO_2^- detected might result from either the contamination of *Nitrobacter* or the chemical oxidation. Since no *Nitrobacter* was detected in the experiment, NO_3^- was likely the oxidation product of NO_2^-. When the biofilter was supplied with high NH_3 (e.g., 120 ppm), 5% of residual NH_4^+/NH_3 was found in the reactor. This residual NH_4^+/NH_3 may neutralize the acid product of H_2S (i.e. $SO_4^=$) and thus maintain the pH of the operation. This is the reason why high H_2S removal efficiency was found in the case of 1:2 mixing ratio (Fig. 2A). In this case, the NO_2^- concentration was about 396 mg l^{-1}. Hence, the low NO_2^- concentration would not result in toxicity to *Nitrosomonas* (Sato et al., 1988).

3.3. Bioaerosol analysis

Applying biofiltration on deodorized process has been proved to be very promising (Leson and Winer, 1991). However, because biofilters contain tremendous amounts of microorganisms, it is necessary to assess the environmental risk associated with the bacteria released from the biofilter when large quantities of waste gases are treated.

Table 4 shows the number of microorganisms in the outlet exhaust when the biofilter was conducted continuously for two months. Apparently, as microorganisms were immobilized in Ca-alginate, the exhaust contained only small amounts of nitrify bacteria (3 CFU/m^3) and neutrophic *Thiobacillus* spp. (6–15 CFU/m^3) in any cases. Because only *Thiobacillus* and *Nitrosomonas* were found in the outlet exhaust, it is reasonable to believe that the seeded species remained dominant. Since the immobilized cell biofilter was free from the contamination by heterotrophic bacteria (Table 4), this system can be considered safe if placed close to populated areas.

3.4. Pressure drop

The relationship between pressure drop and superficial gas velocity is an important parameter in determining the operational cost. The influence of superficial gas velocity on pressure drop is shown in Fig. 3. In this experiment, the superficial gas velocity was raised gradually from 18 to 180 h^{-1} and the temperature was

Table 2
Sulfur mass balances in the biofilter at different ratios of H_2S/NH_3 supply

Mixture ratio (ppm/ppm)	H_2S Removed (g-S/kg-bead)	$SO_4^=$ 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]	3.21	0.81 (25.2)	2.32 (72.3)	0.065 (2.0)	0.016 (0.5)
1:2	3.17	0.78 (24.6)	2.28 (72.0)	0.082 (2.6)	0.026 (0.8)
2:1	6.19	1.12 (18.0)	4.45 (72.0)	0.125 (2.0)	0.499 (8.0)

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

Table 3
Nitrogen mass balances in the biofilter at different ratios of H_2S/NH_3 supply

Mixture ratio (ppm/ppm)	NH_3 Removed (g-N/kg-bead)	NH_4^+/NH_3 Amount (g-N/kg-bead) (%)	NO_2^- Produced (g-N/kg-bead) (%)	NO_3^- Produced (g-N/kg-bead) (%)
1:1[a]	1.67	0.017 (1.0)	1.62 (97.0)	0.03 (2.0)
1:2	2.98	0.151 (5.0)	2.83 (95.0)	–
2:1	1.49	0.020 (1.4)	1.44 (96.6)	0.03 (2.0)

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

Table 4
Bioaerosol analysis in the outlet exhaust of the autotrophic biofilter

Mixture ratio	Medium type				
	Nutrient	PDA	Thiosulfate	Modified Waksman	Nitrifying
1:1	ND	ND	9	ND	3
1:2	ND	ND	6	ND	ND
2:1	ND	ND	15	ND	ND[a]

[a] ND < 3 CFU/m^3, Unit: CFU/m^3.

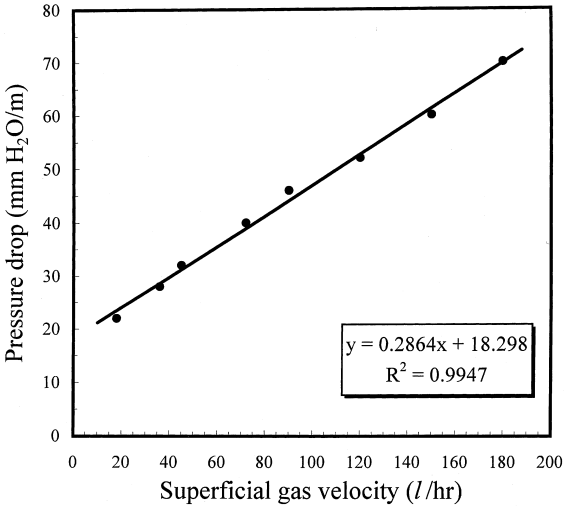

Fig. 3. Profile of pressure drop vs. superficial gas velocity for the autotrophic biofilter.

maintained 30°C. When the operation reached the steady state (about 3 days), a new superficial gas velocity was selected. Inspection of the figure reveals that pressure drop of the biofilter increases with increasing superficial gas velocity and in a good linear behavior. The possible reason is due to no significant biomass accumulation from which would slow the growth rate of the autotrophic bacteria. Consequently, the biofilter exhibited excellent dispersion characteristics.

3.5. Kinetic analysis

The apparent kinetic parameters of the maximum removal rate and half-saturation constant to degrade H_2S (5–120 ppm) under the presence of NH_3 (60 or 120 ppm) are calculated by the Lineweaver-Burk method and the results are shown in Fig. 4. Inspection of the related coefficient of the regression equation (i.e., 0.998) shown in Fig. 4 indicates that NH_3 does not intervene the metabolism of H_2S by *T. thioparus* CH11. Furthermore, the apparent maximum removal (1.11 g-S/day/kg-bead) and the apparent half-saturation constant (34.6 ppm) are similar to those reported by Chung et al. (1997) under a NH_3-free atmosphere.

Fig. 5A illustrates the kinetic analysis of NH_3 removal by *N. europaea* in the range of 5–65 ppm under various H_2S concentrations. Compared to the result of the H_2S-free inlet, the influence of 60 ppm H_2S in the inlet on NH_3 removal of gas mixture was not obvious. However, once the concentration of H_2S was raised to

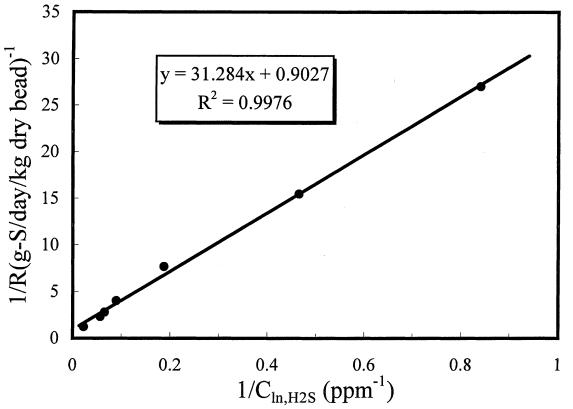

Fig. 4. Relationship between $1/R$ and $1/C_{ln,H_2S}$ of H_2S degradation in the biofilter. Conditions: H_2S (5–120 ppm), NH_3 (60 or 120 ppm), and superficial gas velocity (36–72 l/h).

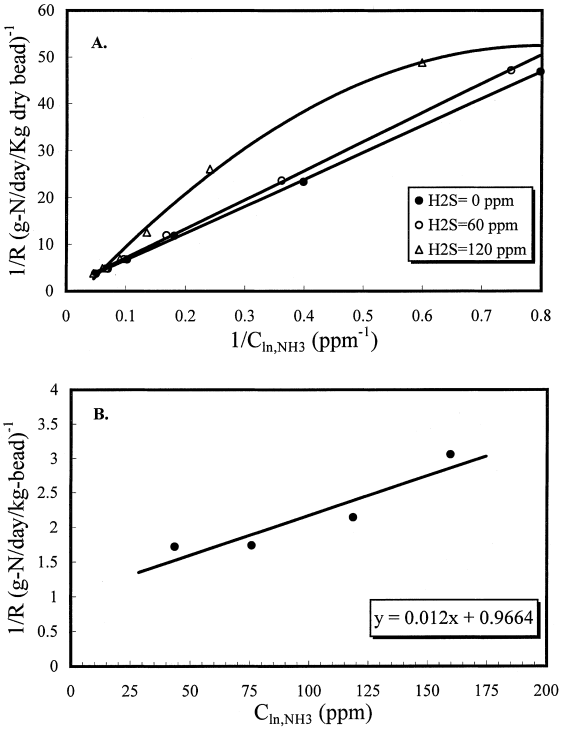

Fig. 5. Relationship between $1/R$ and $1/C_{ln,NH_3}$ of NH_3 degradation in the biofilters. (A) Effect of H_2S concentrations on NH_3 removal by *N. europaea* at NH_3 (5–65 ppm) and H_2S (0, 60 or 120 ppm). (B) Effect of NH_3 concentrations on NH_3 removal by *N. europaea* at NH_3 (120–200 ppm) and H_2S 60 ppm. The superficial gas velocity ranges from 36 to 72 l/h.

120 ppm, a concave-down curve occurred, suggesting that high H_2S concentration inhibited the metabolism of NH_3 by *N. europaea* (Julitte et al., 1993). It is difficult to categorize H_2S as a competitive or non-competitive inhibitor from the existing experimental evidence. Fig. 5B shows the effect of high NH_3 concentration (120–200 ppm) on the NH_3 removal by *N. europaea* when the inlet H_2S concentration in the mixture was maintained at 60 ppm. As mentioned in Fig. 2B, high NH_3 concentration may inhibit the nitrification of *N. europaea.* Therefore, further research on the inhibition kinetic analysis at high NH_3 concentration is essential. The inhibition coefficient (K_i) was calculated as 80.5 ppm from Eq. (3) using linear regression.

3.6. Criteria for design of scale-up biofilters

A complete removal for H_2S and NH_3 can be achieved only at less than critical loading rate. The loading rate of the system is defined as the amount of inlet gas per unit of time, per volume of packing material (g-S or-N/m^3-h). Thus, inlet gas concentrations play an important role in the design of a scale-up biofilter when constant packing material volume and space velocity are used. Finding the maximum inlet concentration and the optimal loading rate, therefore, is crucial for the biofilter operation. The relationship between the maximum inlet concentration and residence time (space velocity) for H_2S and NH_3 removal is shown in Fig. 6. From the operational point of view, to reach the ambient air standards of H_2S and NH_3 (0.1 and 1 ppm, respectively), the maximum inlet concentrations were 58 ppm for H_2S and 164 ppm for NH_3, as specified in the figure, at the residence time of 72 s. Under such condition, the maximum loading rate was determined as 3.8 g-S/m^3-h for H_2S and 5.6 g-N/m^3-h for NH_3, respectively (data not shown).

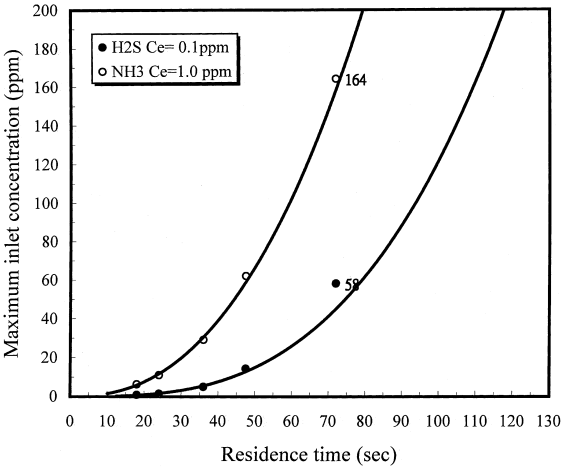

Fig. 6. Relationship between maximum inlet concentration and residence time for H_2S and NH_3 removal by the autotrophic biofilters.

4. Conclusions

It is demonstrated that the removal efficiency remains above 95% when treating H_2S and NH_3 gas mixtures of various ratios with a co-immobilized cell biofilter. Hydrogen sulfide appears to be an inhibitory substrate for NH_3 removal, while NH_3 concentration has little effect on H_2S removal. From our studies, appropriate inlet ratios of H_2S/NH_3 (e.g., <2) and excellent dispersion characteristics can maintain the operational stability of the biofilter. Maximum inlet concentrations to reach the ambient air standards are 58 ppm for H_2S and 164 ppm for NH_3 at residence time of 72 s. These inlet concentrations are in the range of gas emission from the livestock farming and the accompanying wastewater treatment.

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