

Control of H₂S waste gas emissions with a biological activated carbon filter

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Abstract: The removal of high concentrations of H₂S from waste gases containing mixtures of H₂S and NH₃ was studied using the pilot-scale biofilter. Granular activated carbon (GAC), selected as support material in this study, demonstrated its high adsorption capacity for H₂S and good gas distribution. Extensive tests to determine removal characteristics, removal efficiency, and removal capacity of high H₂S levels and coexisting NH₃ in the system were performed. In seeking the appropriate operating conditions, the response surface methodology (RSM) was employed. H₂S removal capacities were evaluated by the inoculated bacteria (biological conversion) and BDST (Bed Depth Service Time) methods (physical adsorption). An average 98% removal efficiency for 0.083–0.167 mg dm⁻³ of H₂S and 0.004–0.021 mg dm⁻³ of NH₃ gases was achieved during the operational period because of rapid physical adsorption by GAC and subsequently an effective biological regeneration of GAC by inoculated *Pseudomonas putida* CH11 and *Arthrobacter oxydans* CH8. The results showed that H₂S removal efficiency for the system was not affected by inlet NH₃ concentrations. In addition, no acidification was observed in the BAC biofilter. High buffer capacity and low moisture demand were also advantages of this system. The maximal inlet loading and critical loading for the system were 18.9 and 7.7 g-H₂S m⁻³ h⁻¹, respectively. The results of this study could be used as a guide for the further design and operation of industrial-scale systems.

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Keywords: biofilter; hydrogen sulfide; ammonia; granular activated carbon; waste gases

1 INTRODUCTION

Hydrogen sulfide (H₂S) is a highly toxic chemical while ammonia (NH₃) can produce strong odors and create visibility problems. H₂S and NH₃ often coexist and are released in large quantities from agricultural activities and industrial processes such as livestock farming, food and rubber processing, leather manufacturing, wastewater treatment, landfills for waste disposal, and hog manure.^{1–4} Typical concentrations of NH₃ and H₂S emitted from these activities and industries, including compost plants, livestock farming and fish processing, range from 0.004 to 0.042 mg dm⁻³ and from 0.007 to 0.138 mg dm⁻³, respectively.^{1,5,6} However, high concentrations of H₂S and low concentrations of NH₃ are often simultaneously emitted from rubber processing, leather manufacturing, and some wastewater treatment.⁷ In these mixed gases, control of H₂S emission is difficult, yet important, because of its high toxicity and low threshold exposure limits.⁸ Some past studies have focused on H₂S removal alone and have achieved a high removal efficiency,^{9–11} but few studies present removal characteristics and treatment data for high concentrations

of H₂S in the coexistence of NH₃ gas in a long-term treatment.

Physical and chemical methods that have been developed to purify gas pollutants from waste gases and wastewater include activated carbon adsorption, ozone oxidation, and incineration.^{12–14} Considering treatment concentrations, physiochemical properties and cost of treating the pollutant, biological treatment is the best available control technology (BACT) for odor treatment.¹⁵ The kinds of microbes and carriers present are crucial considerations in the development of waste gas treatment.¹⁵ Although appropriate and promising species, including *Thiobacillus* sp, *Xanthomonas* sp and *Pseudomonas* sp have been applied to H₂S removal,^{10,16,17} our previous studies have shown that *Pseudomonas putida* CH11 has great potential to remove H₂S gas effectively because of its fast oxidation rate, low acid production, and high removal capacity.^{7,10} Widely used carriers such as soil, compost, peat, activated carbon, or mixtures of different materials have been examined. The results indicate that soils have limited effectiveness since they are prone to short-circuiting and clogging,¹⁸

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and compost suffers from aging effects that decrease the effectiveness of the biological system.¹⁹ While peat is a good carrier, it may produce pressure loss and water control problems in a long-term treatment.²⁰ Granular activated carbon (GAC), which is inoculated with mixed cells as a packing material, has become popular^{21,22} because it provides a more uniform surface area and good resistance to crushing, allowing better operational control in areas such as gas distribution, pressure drop and pH.²³ In addition, GAC provides higher pollutant adsorption capacity than other inert carriers.²² Since biological oxidation occurs immediately after physical adsorption of activated carbon, minimizing the treatment cost, the biological regeneration of GAC gives the utilization of activated carbon a new direction.

In previous studies, we found that *Pseudomonas putida* CH11 is effective in eliminating H₂S and that *Arthrobacter oxydans* CH8 is effective in removing NH₃.^{10,24} In an effort to improve H₂S removal efficiency (90%) from a mixture with high levels of H₂S and NH₃ using Ca-alginate as a carrier,⁷ these two species were simultaneously inoculated into the biofilter using activated carbon as a carrier. In this study, the pilot-scale BAC biofilter was fed with various high H₂S concentrations and constant NH₃ loadings for a 172-day operating period. The removal characteristics and appropriate operating conditions for H₂S removal in the presence of NH₃ were investigated, and good control of H₂S and NH₃ emissions was achieved.

2 MATERIALS AND METHODS

2.1 Organism, cultivation and medium preparation

The original pure-culture strains of heterotrophic sulfur-oxidizer *P putida* CH11 and heterotrophic ammonia-oxidizer *A oxydans* CH8 were isolated from swine wastewater.^{10,24} Stock cultures were both grown in nutrient broth at 30 °C. The nutrient broth contained yeast extracts (5 g dm⁻³), tryptone (10 g dm⁻³) and dextrose (2 g dm⁻³). In continuous-treatment experiments, the inflow medium was supplied and stored in the nutrient tank. The inflow medium contained glucose (10 g dm⁻³), KH₂PO₄ (4.08 g dm⁻³), K₂HPO₄ (5.22 g dm⁻³), NH₄Cl (0.4 g dm⁻³), MgCl₂·6H₂O (0.2 g dm⁻³) and Fe(III)-citrate (0.01 g dm⁻³) (C:N = 40:1). The final pH of the medium was adjusted to neutral using 2 mol dm⁻³ NaOH or HCl. The buffer capacity in the inflow medium was calculated as 0.033 (mol dm⁻³ pH⁻¹).

2.2 Immobilization procedure

Granular activated carbon (extruded 4.5 mm particles, from Cherng Tay Corporation Ltd in Taiwan) was used as the support material for the biomass. The main characteristics of the support material—bulking density, specific surface area, and pH value—were

0.48 g cm⁻³, 1250 BET-m² g⁻¹ and 9, respectively. *P putida* CH11 or *A oxydans* CH8 were each grown in 1000 cm³ nutrient broth for 1 day and then harvested by centrifugation (8000 × *g* for 10 min). The precipitate was put into a 10 dm³ PVC tank containing 7 dm³ nutrient broth. Prior to initiating the biofilter experiments, 2.7 kg of the granular activated carbon (GAC) were separately mixed with the above solution for microbial attachment. During the cultivation period, nutrient broth was added every 3 days until the cell numbers of *P putida* CH11 and *A oxydans* CH8 reached 8.0 × 10¹⁰ and 6.0 × 10¹⁰ cfu(g dry GAC)⁻¹, respectively. After 28 days the GAC in the tank was then transferred into the biofilter.

2.3 Apparatus and gas removal for continuous operation

To investigate the capacity of GAC to adsorb H₂S, three glass columns (12 cm φ × 3 cm working length) connected in series were packed with GAC without microbes to perform the BDST (Bed Depth Service Time) experiment. H₂S, 0.083 mg dm⁻³, was continuously supplied to these glass columns at 500 dm³ h⁻¹. The desired concentration of H₂S at the breakthrough was defined as 0.0033 mg dm⁻³ (ie: C_e/C₀ = 0.04).

A set-up of the pilot-scale experimental BAC biofilter is shown in Fig 1. Two glass columns (12 cm φ × 40 cm working height) connected in series were packed with cell-laden GAC supported by a perforated sieve plate at the bottom of the column to allow the circulating liquid to flow out. The packed volume and GAC dry weight in the biofilter were 9.05 dm³ and 4.34 kg, respectively. Each glass column

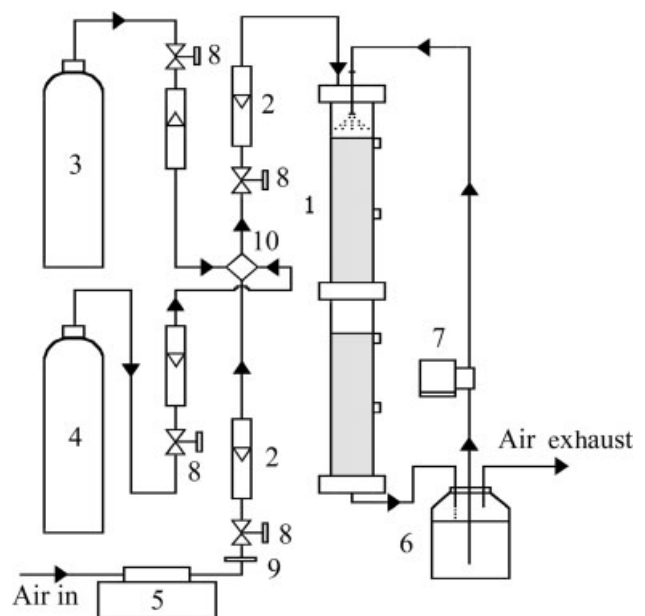


Figure 1. Schematic of the pilot scale BAC biofilter. 1, Glass column; 2, flow meter; 3, NH₃ gas cylinder; 4, H₂S gas cylinder; 5, air compressor; 6, nutrient tank; 7, pump; 8, regulator; 9, air filter; 10, four-way connector.

with two gas sampling ports, which spaced 20 cm apart, were drilled along the column for measuring gas concentrations. The flow meters and valves were used for monitoring and controlling the gas flow through the reactor. The pure H₂S and NH₃ gases, supplied from separate gas cylinders with flow rates ranging from 1.4 to 10.8 dm³ h⁻¹, were first diluted with compressed air, which passed an air filter (pore size 0.2 μm, LIDA 3000-06, made in USA) and then flowed downward through the biofilter at the top. The 7 dm³ of inflow medium (see Section 2.1) stored in the nutrient tank was recirculated by a peristaltic pump at 10 dm³ min⁻¹ for 6 min, once every 4 h to maintain the moisture of the biofilter and supply nutrient to the attached cells. The peristaltic pump was connected to a spray nozzle (spray angle 115°; connecting thread 3/8"; Chang Der Co, Taiwan) to uniformly spray the medium on the filter bed's surface. Glucose of 1% was added every 2 weeks. In the 172-day treatment experiment, different H₂S concentrations (0.083–0.167 mg dm⁻³) under constant inlet NH₃ loading (1.37 g-N m⁻³ h⁻¹) were introduced to the BAC biofilter at various flow rates (180–1080 dm³ h⁻¹) to evaluate the system's performance.

2.4 Experimental design

Response surface methodology (RSM), described first by Box *et al.*,²⁵ is an experimental strategy for seeking the appropriate operating conditions for a multivariable system. The combined effect of inlet concentration and gas flow rate on H₂S removal was investigated using this methodology. In the present work, a central composite design (CCD) for two variables was used to study the response pattern. The full-factorial design consisting of a two-factor-two-level pattern with 11 design points (nine combinations with three replications of the center points) was used. A multiple regression analysis was performed to obtain the coefficients, and these equations were used to predict the response using the STATISTICA program.

2.5 Analytical methods

Inlet H₂S gas concentrations in the reactor were periodically measured by gas detector tubes (Kitagawa, Japan) in the range of 0.00138–0.207 mg dm⁻³. Outlet concentrations were continuously measured using a Single Point Monitor (MDA Scientific, USA) in the range of 7 × 10⁻⁵–2.1 × 10⁻⁴ mg dm⁻³ or periodically measured by gas detector tubes (Kitagawa, Japan) in the range of 0.00138–0.083 mg dm⁻³. Inlet NH₃ gas concentrations in the reactor were periodically measured by gas detector tubes (Kitagawa, Japan) in the range of 0.0035–0.069 mg dm⁻³. Outlet concentrations were continuously measured using a Single Point Monitor (MDA Scientific, USA) in the range of 7 × 10⁻⁵–7 × 10⁻⁴ mg dm⁻³. To measure the pH, 0.5 g GAC was withdrawn from the biofilter through the appropriate sampling port and mixed with 5 cm³ of distilled water. The sample was vortexed for 3 min, and the pH was then determined using a pH meter.

Circulating liquid (20 cm³) was withdrawn from the nutrient tank at regular intervals to measure the tank's pH value. To determine the moisture content in the GAC, about 5 g GAC were withdrawn, weighed, and dried over a 24-h period at 103 ± 0.5 °C, and the measurement was conducted 20 min after spraying the aqueous medium. For cell number estimation, 0.5 g GAC was separately taken from sampling ports at the different depths and mixed with 5 cm³ sterile water. These samples were vortexed for 3 min, and the cell numbers in the filter were enumerated by traditional plate-counting methods. In this case, the LB (Luria–Bertani) medium for heterotrophic microbial cultivation, the Cetrimide selective medium for *P. putida* CH11,²⁶ and the Hagedorn & Holt selective medium for *A. oxydans* spp were used.²⁷ The *N*-cetyl-*N,N,N*-trimethylammonium bromide in the Cetrimide selective medium was able to largely inhibit the growth of the accompanying microbial flora except for *Pseudomonas* sp²⁶ and the cycloheximide in the Hagedorn & Holt selective medium was a selective agent for *Arthrobacter* sp.²⁷ The inoculated plates grew for 2 days in an incubator at 26 °C. The 16S rDNA sequence was applied after the selective medium was used to ensure they were *P. putida* CH11 and *A. oxydans* CH8.

3 RESULTS AND DISCUSSION

3.1 H₂S adsorption by GAC

GAC is a good carrier for bacterial growth, but it is also effective for direct adsorption of H₂S. The physical adsorption capacity of GAC to remove H₂S is shown in Fig 2. The adsorption process is described by Eqn (1):

$$t = \left(\frac{N_0}{C_0 V} \right) X - \frac{1}{KC_0} \ln \left(\frac{C_0}{C_e} - 1 \right) \quad (1)$$

where C_0 and C_e are the inlet concentration and the desired concentration of gas at breakthrough (mg dm⁻³), respectively; V , t and X are hydraulic loading (cm h⁻¹), service time (h⁻¹) and bed depth (cm); N_0 and K are adsorption capacity (mg dm⁻³) and adsorption rate constant (mg⁻¹ dm³ h⁻¹).

Equation constants were obtained from the slope and the intercept of Fig 2 by a regression method. The values of slope and intercept were 13.72 and 0.069, respectively. The adsorption capacity (N_0) and the adsorption rate constant (K) were 5057 mg dm⁻³ and 70.29 mg⁻¹ dm³ h⁻¹, respectively. The adsorption capacity expressed as dry weight of GAC was 11.78 mg-H₂S(g dry GAC)⁻¹. This indicates that GAC has much greater adsorption capacity than Calcium alginate (6.8 × 10⁻⁴ mg-H₂S(g dry bead)⁻¹).²⁸ The data of H₂S adsorption capacity by pure GAC provide a reference value for the removal capacity of cell-laden GAC.

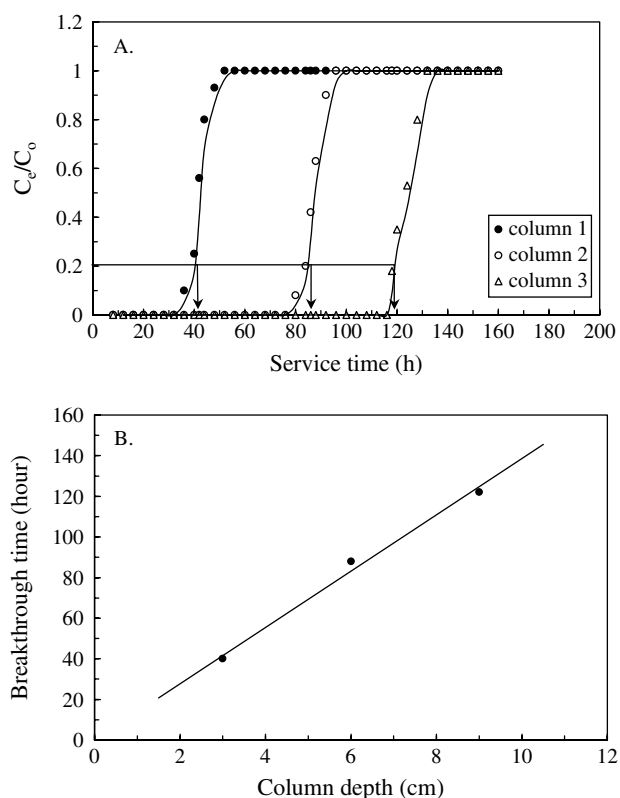


Figure 2. Adsorption capacity of GAC to H₂S evaluated by three identical GAC columns connected in series. (A) breakthrough curve by BDST method and (B) linear correlation between packing height and breakthrough time according to the equation using breakthrough data.

3.2 Removal of high H₂S concentrations from mixed waste gases in continuous operation

Because mixtures of H₂S at high concentrations and NH₃ at low concentrations are often simultaneously emitted from various industries or manufacturing processes, it is necessary to examine the removal efficiencies or characteristics of a long-term treatment.⁷ The H₂S removal efficiency profile in the BAC biofilter during 172 days of operation is indicated in Fig 3(A). When the system operated to pseudo steady state (ie the rate of physical adsorption equals that of biological oxidation) on about the 22nd day, different inlet H₂S loadings under constant inlet NH₃ loading (1.37 g-N m⁻³ h⁻¹) were introduced into the bioreactor to test the H₂S removal performance of the system. Throughout all the operating periods, 100% NH₃ removal was achieved. The complete breakthrough time of H₂S concentration in this system was about 44 days as estimated by the adsorption equation. Hence, physical adsorption of GAC was theoretically responsible for 100% H₂S removal in the first 44 days. After that, the inoculated bacteria apparently contributed to complete removal (100%) from the 44th to the 59th day. After the 119th day, intermittent shock loadings were conducted to test the response of the system. By day 119, the H₂S loading had been tripled (from 6.64 g-H₂S m⁻³ h⁻¹ to 19.92 g-H₂S m⁻³ h⁻¹) for 4 days. Although removal efficiency (>98%) was down to 55%, once the inlet

H₂S loading returned to its original level, 80% removal efficiency (RE) was achieved within 2 days, and 98% removal efficiency was achieved in 5 days. Besides, at the load used in this experiment, RE > 95% were obtained at EBRT > 45 s and the RE dropped to about 60% at an EBRT of 30 s. During 6 months of study, an average of 98% H₂S removal was demonstrated. In comparison to other systems (compost filter, three-phase fluidized bed bioreactor, or Calginate biofilter) under similar operating conditions, the BAC biofilter exhibited much higher efficiency than other biofilter systems,^{9,10,29} though it was inferior to the parallel dual column filters packed with compost.⁴ Additionally, the BAC biofilter also exhibited a high buffer capacity for perturbed H₂S inlet. Fast physical adsorption and subsequently effective biological oxidation resulted in successful H₂S and NH₃ removal. Simultaneous bioregeneration of the activated carbon was an additional economic advantage to these systems. To understand the microbial activity and distribution in the bioreactor, changes in the cell numbers were examined. During the 172-day experiment, variations in cell number ranged from 10⁹ and 10¹⁰ cfu(g dry GAC)⁻¹ at the different sampling ports (data not shown). The profile of the cell numbers did not show gradient change along the column length. Hence, H₂S and NH₃ gases accumulated uniformly in the GAC or were distributed over the bioreactor and were subsequently degraded by the inoculated microbial population within the matrix of carbon particles. The data of Table 1 show the cell numbers and distribution ratios of the inoculated cells (*P putida* and *A oxydans*) and other heterotrophic bacteria in the middle zone of the BAC biofilter. The results indicated that sulfur-oxidizer *P putida* was still a dominant microorganism in the bacterial population and accounted for 92%, 94% and 92% of bacteria on the 60th, 120th, and 172nd days, respectively. The ammonia-oxidizer *A oxydans* was second and accounted for 8%, 6%, and 8%. The other heterotrophic bacteria were relatively few.

Although the *P putida* and *A oxydans* cell numbers were almost equal at the beginning, the gas mixtures with a high concentration of H₂S and low concentration of NH₃ would be the main reason for the change in the microbial population. In this system, we found that *P putida* maintained a high distribution ratio and cell number in the bacterial community during the process of intermittent shock loading, and it brought a stable and high removal efficiency. In addition, low concentrations of NH₃, in the range of 0.004–0.021 mg dm⁻³, could be also effectively removed by inoculated *A oxydans*. To establish operating criteria for scale-up BAC biofilter, the relationship between inlet loading and removal capacity for H₂S gas is indicated in Fig 3(B). As shown in Fig 3(B), the relationship curve first rose and then leveled off to a maximum level. The critical loading (ie complete removal capacity) was

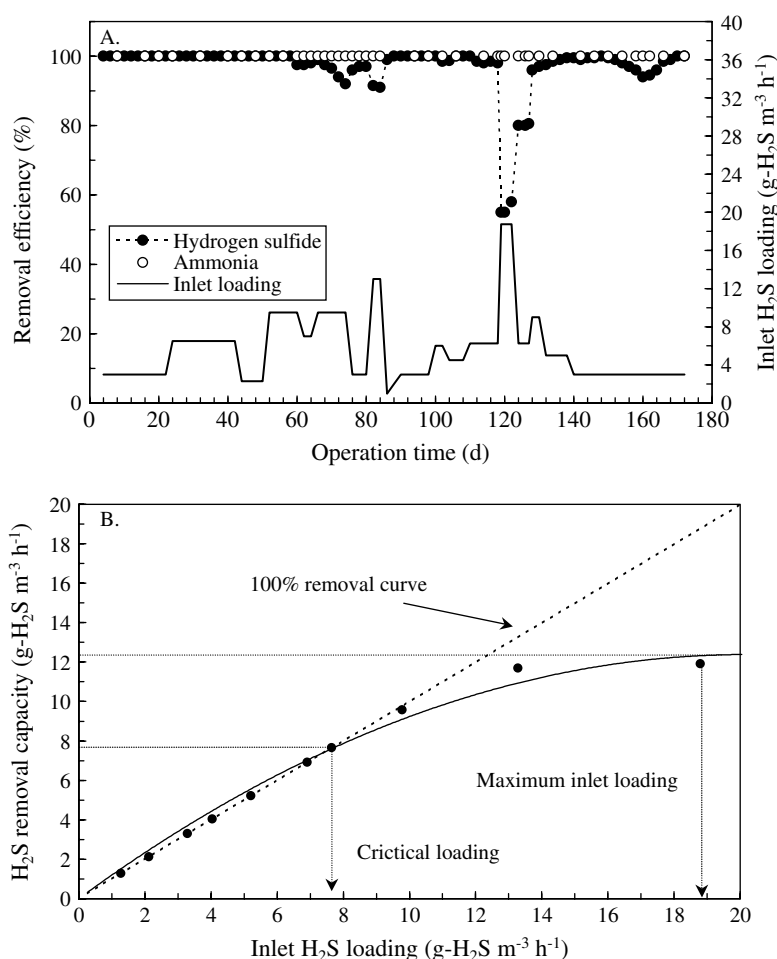


Figure 3. (A) H₂S, NH₃ removal efficiency in the BAC biofilter under constant inlet NH₃ loading (1.37 g-N m⁻³ h⁻¹) during 172-day operation at room temperature and (B) relationship between inlet loading and removal capacity for H₂S gas.

Table 1. Cell numbers and distribution ratios of inoculated cells and other heterotrophic bacteria in the middle zone of BAC biofilter

Strain ^a	Day			
	0	60	120	172
<i>P. putida</i>	8.0 × 10 ¹⁰ (57%)	7.4 × 10 ⁹ (92%)	5.7 × 10 ⁹ (94%)	6.3 × 10 ⁹ (92%)
<i>A. oxydans</i>	6.0 × 10 ¹⁰ (43%)	6.3 × 10 ⁸ (8%)	3.8 × 10 ⁸ (6%)	5.4 × 10 ⁸ (8%)
Other	0 (0%) ^b	7.4 × 10 ^{5b}	3.6 × 10 ^{5b}	5.2 × 10 ^{5b}

^a Indicates the unit as cfu (g dry GAC)⁻¹.

^b indicates the distribution ratio's lower than 0.01%.

determined as 7.7 g-H₂S m⁻³ h⁻¹. The extrapolated correlation line suggested a maximum inlet loading of 18.9 g-H₂S m⁻³ h⁻¹ (0.85 g-H₂S kg⁻¹ day⁻¹). Compared with biological systems with peat support, values of which were 0.102, 0.403, and 0.468 g-H₂S kg⁻¹ day⁻¹,³⁰⁻³² the maximum inlet loading of the BAC biofilter was better. Thus, efficient and acceptable H₂S removal can be accomplished through adjusting the flow rate and inlet concentration based on the correlation presented in Fig 3(B).

3.3 Effect of temperature on H₂S removal

Temperature is an important factor that will affect physical absorption, biological oxidation, and especially microbial growth. To estimate the system

response to temperature variation, the H₂S removal efficiencies with temperature changes at gas retention time of 45 s are shown in Fig 4. The experiment was conducted on the same biofilter after a 172-day operating period. H₂S loading was controlled at 9.64 g-H₂S m⁻³ h⁻¹, and NH₃ loadings were in the range of 0.389–2.04 g-NH₃ m⁻³ h⁻¹. Since the optimum temperature for *P. putida* CH11 growth is 26 °C,¹⁰ the removal efficiency slightly decreased from 99% to 96% when temperature rose from 26 °C to 45 °C. Surprisingly, 96% removal of H₂S was still maintained even with the temperature controlled at 45 °C for another 6 days. When the temperature of the system was returned to 26 °C, the removal efficiency was 99% within 4 days. These results suggest that

a temporary biological deactivation did not result in complete loss of removal capacity of the BAC biofilter because the adsorption process might remove H₂S gas. Therefore, the BAC biofilter possessed high adaptability to temperature shock, and this property would favor its application for H₂S removal in the field.

3.4 pH and moisture change in the BAC biofilter

Acidification has often been an obstacle to traditional biofilter methods for acid gas treatment.⁴ Hence, pH control at a constant level in the bioreactor and leachate was very important for biofilter function. During the 172-day treatment in this study, the pH values in the filter bed and leachate were determined every 4 days, and the results are shown in Fig 5. The increasing acid gas H₂S at constant inlet NH₃ loading resulted in a decrease of pH in the leachate. The lowest pH value of 4.5 was observed in the leachate on the 120th day because the shock loading of inlet H₂S was conducted on the 120th day, and then 0.1 mol dm⁻³ NaOH solution was added into the nutrient tank to restore the pH to neutral. The acidification did not occur in the filter because the changes in pH value were between 6.0 and 8.5. Since the pH of the activated carbon was 9, it was able to stabilize the pH of the filter bed and provide the system with high alkalinity following the addition of acidic H₂S gas. The presence of ammonia in the waste gas provided another neutralizing effect. Therefore, the profile of the pH showed a gradient change in the axial direction of the filter bed. These results were due to accumulation of more acid products on the lower layer (data not shown). This phenomenon was attributed to the combined effect of liquid flow direction and gravity.

Microbial activity and mass-transfer rate often depend on the moisture content in or around the support material. High moisture could lead to a serious problem, such as the formation of stagnant zones with diffusion limitations and possible anaerobic conditions or increased pressure drop.^{1,33} Thus, an appropriate

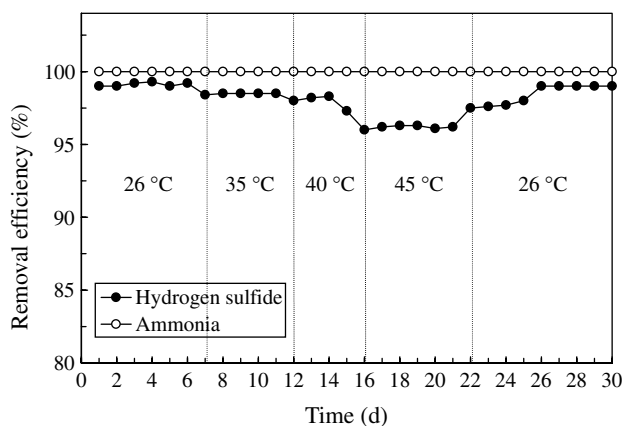


Figure 4. Effect of temperature on H₂S and NH₃ removal efficiency. H₂S concentration was controlled at 0.138 mg dm⁻³. NH₃ concentrations were in the range of 0.004–0.021 mg dm⁻³, and 100% NH₃ removal efficiency was achieved.

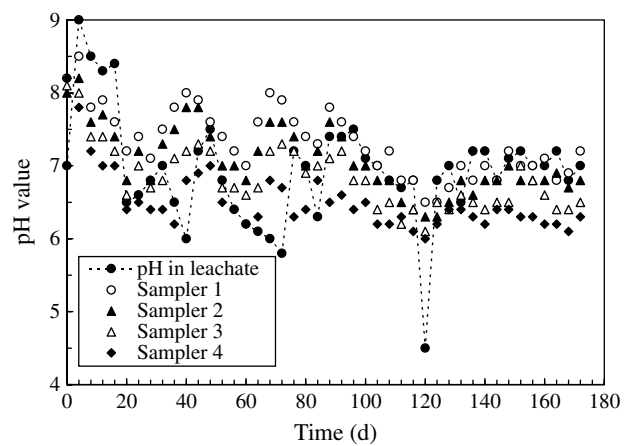


Figure 5. Change of pH values in the leachate and the filter operated with BAC biofilter for 172 days.

moisture content in GAC was required to efficiently remove waste gas. The moisture content of GAC at the different filter depths was determined every 5 days, and the results are shown in Fig 6. Moisture content varied from 34% to 42% with the average at 38%. By comparison, the transitional biofilter required a 40–60% moisture content to maintain biological activity.³⁴ The BAC biofilter is energy-saving and highly efficient.

3.5 Effect of NH₃ on H₂S removal

To understand the effect of NH₃ coexistence on H₂S removal from mixed waste gases in the BAC biofilter, different NH₃ concentrations (0.007–0.084 mg dm⁻³) were introduced at 360 dm³ h⁻¹ with 0.138 mg dm⁻³ H₂S. The study was a separate experiment and was conducted after a 172-day operating period. Compared with the previous studies, the H₂S removal efficiency gradually fell to 88% in the Ca-alginate immobilization system because of slight alkalization from NH₃ coexistence.⁷ By contrast, H₂S removal by the BAC biofilter was not influenced by NH₃ concentrations under these operating conditions. A removal efficiency of 99% was achieved in all operating

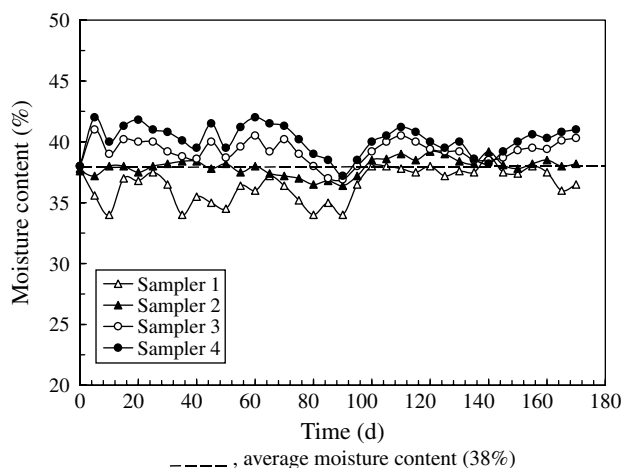


Figure 6. Change of moisture content at different sampling ports in the BAC biofilter.

conditions. Thus, the results of this study suggest that using activated carbon as a packing material or a temporary gas storage tank would facilitate high removal efficiencies.

3.6 Comprehensive evaluation of concentration and flow rate for H₂S removal

Because the most important physical factors affecting H₂S removal are inlet H₂S concentrations and gas flow rates, experiments were performed at different inlet concentrations and flow rate combinations, and suitable combinations for emission limits were determined when NH₃ concentration fluctuated within 0.004–0.021 mg dm⁻³. Using the results of the experiments, the following equation giving the relative H₂S removal efficiency as the function of inlet concentration and flow rate was obtained:

$$y = 101.3621 - 0.00102x_1 - 0.0054x_2 - 0.000016x_1x_2 \quad (2)$$

where x_1 = H₂S concentration and x_2 = gas flow rate.

According to the above equation, all the factors have negative effects. The equation also shows that x_2 (gas flow rate) is the most significant factor, with its coefficient effect being most pronounced. The contour plot of Fig 7 explains the behavior of the system. If 100% removal efficiency was achieved at 0.07 mg dm⁻³ of inlet H₂S, the gas flow rate should be controlled below 225 dm³ h⁻¹. In addition, 99% removal was obtained when the gas flow rate was controlled below 375 dm³ h⁻¹. By the developed equation or contour plot, a set of suitable operating combinations was determined and could be further applied in the field with gases containing high concentrations of H₂S and coexistent NH₃.

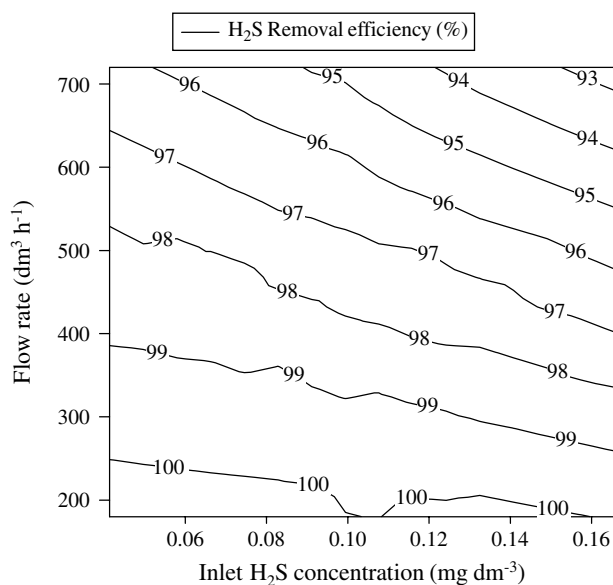


Figure 7. Contour plot showing H₂S removal efficiency in response to varying concentration and flow rate.

4 CONCLUSIONS

Control of high concentrations of H₂S together with low concentrations of NH₃ emitted from industrial waste gas treatments is an important process because H₂S has high toxicity and a low threshold limit. In this study, we demonstrate that the BAC biofilter can effectively remove a high concentration of H₂S gas from mixed waste gases containing H₂S and NH₃. The system achieved an average 98% removal efficiency of H₂S during a 172-day operating period when NH₃ concentrations were between 0.004 and 0.021 mg dm⁻³. Additionally, the system shows the potential for high H₂S removal efficiency even with temperatures as high as 45 °C. No significant acidification phenomenon occurred in this system during H₂S treatment. The low moisture demand of the BAC biofilter was another advantage. A set of the operating combinations was further established for application in the field. Therefore, the results of this study suggested that the BAC biofilter has significant potential to treat H₂S gas from mixed waste gases containing NH₃.

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REFERENCES

- Langenhove HV, Wuyts E and Schamp N, Elimination of hydrogen sulphide from odorous air by a wood bark biofilter. *Water Res* 20:1471–1476 (1986).
- Eikum AS and Storhang R, Odour problems related to waste water and sludge treatment in. *Odour Prevention and Control of Organic Sludge and Livestock Farming*, ed by Neilsen VC, Voorburg JH and Hermite PL. Elsevier Applied Science Publishers, London, pp 12–18 (1986).
- Devinyin JS, Deshusses MA and Webster TS, *Biofiltration for Air Pollutant Control*. Lewis Publishers, New York, pp 7–8 (1999).
- Yang Y and Allen ER, Biofiltration control of hydrogen sulphide 1. Design and operational parameters. *J Air Waste Manage* 44:863–868 (1994).
- Cho KS, Hirai M and Shoda M, Enhanced removal efficiency of malodorous gases in a pilot-scale peat biofilter inoculated with *Thiobacillus thioiparus* DW44. *J Ferment Bioeng* 73:46–50 (1992).
- Chung YC, Huang C and Tseng CP, Reduction of H₂S/NH₃ production from pig feces by controlling environmental conditions. *J Environ Sci Health Part A* 31:139–155 (1996).
- Chung YC, Huang C, Tseng CP and Pan JR, Biotreatment of H₂S- and NH₃- containing waste gases by co-immobilized cells biofilter. *Chemosphere* 41:329–336 (2000).
- Buchman RE and Gibbons NE, *Bergey's Manual of Determinative Bacteriology*. Williams and Wilkins Co, Baltimore, MD, pp 52–54 (1974).
- Rands MB, Cooper DE, Woo CP, Fletcher GC and Rolfe KA, Compost filters for H₂S removal from anaerobic digestion and rendering exhausts. *J Water Pollut Control Feder* 53:185–189 (1981).
- Chung YC, Huang C and Tseng CP, Biodegradation of hydrogen sulphide by a laboratory-scale immobilized *Pseudomonas putida* CH11 biofilter. *Biotechnol Prog* 12:773–778 (1996).
- Oh KJ, Kim D and Lee IH, Development of effective hydrogen sulphide removing equipment using *Thiobacillus* sp IW. *Environ Pollut* 99:87–92 (1998).

- 12 Eby HJ and Wilson GB, Poultry house dust, odour and their mechanical removal, in *Agricultural Waste Management Proceedings, Cornell University Conference on Agricultural Waste Management*, Syracuse, NY, pp 303–308 (1969).
- 13 Barth CL, Elliott FL and Melvin SW, Using odour control technology to support animal agriculture. *Trans ASAE* 27:859–864 (1984).
- 14 Mannebeck H, Covering manure storing tanks to control odour, in *Odour Prevention and Control of Organic Sludge and Livestock Farming*, ed by Neilsen VC, Voorburg JH and Hermite PL. Elsevier Applied Science Publishers, London, pp 188–192 (1986).
- 15 Leson G and Winer AM, Biofiltration: an innovative air pollution control technology for VOC emission. *J Air Waste Manage* 41:1045–1054 (1991).
- 16 Wainwright M, Sulfur oxidation in soil. *Adv Agron* 37:350–396 (1984).
- 17 Cho KS, Hirai M and Shoda M, Degradation characteristics of hydrogen sulphide, methanethiol, dimethyl sulphide and dimethyl disulphide by *Thiobacillus thioparus* DW44 isolated from peat biofilter. *J Ferment Bioeng* 71:384–389 (1991).
- 18 Carlson DA and Leisner CP, Soil beds for the control of sewage odors. *J Water Pollut Control Feder* 38:829–834 (1966).
- 19 Langenhove HV, Bendinger B, Oberthur R and Schamp N, Organic sulfur compounds: persistent odorants in biological treatment of complex waste gases, in *Biotechniques for Air Pollution Abatement and Odour Control Policies*, ed by Dragt AJ and Ham JV. Elsevier Science Publishers, Amsterdam, pp 177–182 (1992).
- 20 Tanji Y, Kanagawa T and Mikami E, Removal of dimethyl sulphide, methyl mercaptan, and hydrogen sulphide by immobilized *Thiobacillus thioparus* TK-m. *J Ferment Bioeng* 67:280–285 (1989).
- 21 Weber FJ and Hartmans S, Use of activated carbon as a buffer in biofiltration of waste gases with fluctuating concentration of toluene. *Appl Microbiol Biotechnol* 43:365–372 (1995).
- 22 Graham JR, GAC based gas phase biofiltration, in *Proceedings of the 1996 Conference on Biofiltration*, ed by Reynolds FE. The Reynolds Group, Tustin, CA, pp 85–90 (1996).
- 23 Medina VF, Webster T and Devinny JS, Treatment of gasoline residuals by granular activated carbon based biological filtration. *J Environ Sci Health Part A* 30:407–412 (1995).
- 24 Chung YC, Huang C and Tseng CP, Biotreatment of ammonia from air by an immobilized *Arthrobacter oxydans* CH8 biofilter. *Biotechnol Prog* 13:794–798 (1997).
- 25 Box GEP, Hunter WG and Hunter JS, *Statistics for Experiments*. John Wiley and Sons, New York, pp 374–418 (1978).
- 26 Mossel DAA and Indacochea L, A new cetrimide medium for the detection of *Pseudomonas aeruginosa*. *J Medical Microbiol* 4:380–382 (1971).
- 27 Hagedorn C and Holt JG, A nutritional and taxonomic survey of *Arthrobacter* soil isolates. *Can J Microbiol* 21:353–361 (1975).
- 28 Chung YC, Huang C and Tseng CP, Removal of hydrogen sulphide by immobilized *Thiobacillus* sp strain CH11 in a biofilter. *J Chem Technol Biotechnol* 68:58–62 (1997).
- 29 Kim KR, Oh KJ, Park KY and Kim D, Removal of hydrogen sulphide and methylmercaptan using *Thiobacillus* in a three phase fluidized bed bioreactor. *J Microbiol Biotechnol* 9:265–270 (1999).
- 30 Furusawa N, Togashi I, Hirai M, Shoda M and Kubota H, Removal of hydrogen sulphide by a biofilter with fibrous peat. *J Ferment Technol* 62:589–593 (1984).
- 31 Wada A, Shoda M, Kubota H, Kobayashi T, Yoko KF and Kuraishi H, Characteristics of H₂S oxidizing bacteria inhabiting a peat biofilter. *J Ferment Technol* 64:161–168 (1986).
- 32 Cho KS, Hirai M and Shoda M, Degradation of hydrogen sulphide by *Xanthomonas* sp strain DY44 isolated from peat. *Appl Environ Microbiol* 58:1183–1189 (1992).
- 33 Ottengraf SPP and Van den Oever AHC, Kinetics of organic compound removal from waste gases with a biological filter. *Biotechnol Bioeng* 25:3089–3102 (1983).
- 34 Sabo F, Development and testing of high-efficiency biofilters, in *Proceedings of the 86th Annual Meeting of the Air and Waste Management Association*, Denver, Colorado (1993).