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Biotreatment of Hydrogen Sulfide- and Ammonia-Containing Waste Gases by Fluidized Bed Bioreactor

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ABSTRACT
Gas mixtures of \( \text{H}_2\text{S} \) and \( \text{NH}_3 \) are the focus of this study of research concerning gases generated from animal husbandry and treatments of anaerobic wastewater lagoons. A heterotrophic microflora (a mixture of \textit{Pseudomonas putida} for \( \text{H}_2\text{S} \) and \textit{Arthrobacter oxydans} for \( \text{NH}_3 \)) was immobilized with Ca-alginate and packed into a fluidized bed reactor to simultaneously decompose \( \text{H}_2\text{S} \) and \( \text{NH}_3 \). This bioreactor was continuously supplied with \( \text{H}_2\text{S} \) and \( \text{NH}_3 \) separately or together at various ratios. The removal efficiency, removal rate, and metabolic product of the bioreactor were studied.

The results showed that the efficiency remained above 95% when the inlet \( \text{H}_2\text{S} \) concentration was below 30 ppm at 36 L/hr. Furthermore, the apparent maximum removal and the apparent half-saturation constant were \( 7.0 \times 10^{-8} \text{ g-S/cell/day} \) and 76.2 ppm, respectively, in this study. The element sulfur as a main product prevented acidification of the biofilter, which maintained the stability of the operation. As for \( \text{NH}_3 \), the greater than 90% removal rate was achieved as long as the inlet concentration was controlled below 100 ppm at a flow rate of 27 L/hr. In the \( \text{NH}_3 \) inlet, the apparent maximum removal and the apparent half-saturation constant were \( 1.88 \times 10^{-6} \text{ g-N/cell/day} \) and 30.5 ppm, respectively. Kinetic analysis showed that 60 ppm of \( \text{NH}_3 \) significantly suppressed the \( \text{H}_2\text{S} \) removal by \textit{Pseudomonas putida}, but \( \text{H}_2\text{S} \) in the range of 5–60 ppm did not affect \( \text{NH}_3 \) removal by \textit{Arthrobacter oxydans}. Results from bioaerosol analysis in the bioreactor suggest that the co-immobilized cell technique applied for gas removal creates less environmental impact.

IMPLICATIONS
A fluidized bed bioreactor was applied for \( \text{H}_2\text{S} \)- and \( \text{NH}_3 \)-containing waste gases. The evidence indicated that the system has the potential to be an effective means of simultaneously removing \( \text{H}_2\text{S} \) and \( \text{NH}_3 \) at low concentrations. Optimal operation criteria were established for widespread use.

INTRODUCTION
\( \text{H}_2\text{S} \) is highly toxic and could cause a safety hazard, while \( \text{NH}_3 \) can produce odor and create visibility problems. The Taiwan Environmental Protection Agency has set the ambient air quality standards of \( \text{NH}_3 \) and \( \text{H}_2\text{S} \) at 1 and 0.1 ppm, respectively. \( \text{NH}_3 \) and \( \text{H}_2\text{S} \) in a wide range of concentrations are released from industrial processes, such as wastewater treatment, landfills for waste disposal, livestock farming, and hog manure. Typical concentrations of \( \text{NH}_3 \) and \( \text{H}_2\text{S} \) emitted from these industries range from 5 to 60 and from 5 to 100 ppm, respectively. The excess amounts of \( \text{NH}_3 \) and \( \text{H}_2\text{S} \) must be removed not only to avoid safety and health hazards, but also to eliminate environmental impacts, such as greenhouse effect, acid rain, and eutrophication. \( \text{H}_2\text{S} \) is usually emitted from the crude oil refining process and often produces environmental problems. The process of oxidation of \( \text{H}_2\text{S} \) to \( \text{H}_2\text{SO}_4 \) can deplete the oxygen of the receiving water, and the acid products can damage the structure of the pipe.

Biotreatments for \( \text{NH}_3 \) and \( \text{H}_2\text{S} \) removal have drawn great attention, because they are more economical than the conventional methods of comparable removal efficiency. Application of a fluidized bed reactor in waste gas treatment is very rare. Some researchers have tried to use the fluidized bed reactor packed with \textit{Nitrosomonas} sp. to remove \( \text{NH}_3 \) from wastewater. Recently, some immobilized-cell technologies have been successfully applied in waste gas treatment. Although Chung et al. demonstrated the high removal efficiency and high operational stability of the immobilized-cell technology in the packed reactor, the advantage of the fluidized-bed system over the traditional packed system for the removal of \( \text{H}_2\text{S} \) and \( \text{NH}_3 \) has not yet been demonstrated from the operational standpoint. Generally, the fluidized bed can provide better mass transfer and less loss of packing material with lower pressure drop.

Until now, few studies have presented the chemical or biochemical relationship between \( \text{NH}_3 \) and \( \text{H}_2\text{S} \). It was discovered that excess sulfide reduced nitrification by 50–100%, which led to the reduction in \( \text{NH}_3 \) removal. However, Sato...
et al. found that the inhibition of nitrification increased with NH₃ concentrations. Hence, in treating waste gas containing more than one component, the inlet gas ratio must be appropriately controlled. In previous studies, we found that *Arthrobacter oxydans* is very effective in removing NH₃ and *Pseudomonas putida* is effective in eliminating H₂S. These two species were, therefore, chosen in our fluidized bed system. In this study, the fluidized bed reactor was fed with NH₃/H₂S gas mixtures at various ratios, and the detailed removal characteristics and inhibitory phenomena between the co-exit gases are discussed.

**MATERIALS AND METHODS**

**Microorganisms and Cultivation**

*Pseudomonas putida* and *Arthrobacter oxydans* were obtained from FIDRI in Taiwan. Stock cultures were grown in plate count broth at 26 °C with 120 strokes/min. The broth contained 5 g/L yeast extract, 10 g/L tryptone, and 2 g/L dextrose. In all consecutive experiments, inflow media (cycling solutions) were used. The compositions of various inflow media are listed in Table 1. The final pH of the medium was adjusted to neutral with 2 N NaOH or HCl.

**Preparation of Immobilized Cells**

*Pseudomonas putida* and *Arthrobacter oxydans* were grown in 100-mL plate count broth separately, harvested by centrifugation (7500 × g, 10 min), and then washed 5 times with sterile distilled water. The cultures were mixed together in sterile 4% (w/w) Na-alginate solution. The Na-alginate solution, containing the mixture of cells, was dropped into a 4% CaCl₂ solution from a syringe. The droplets immediately formed 4-mm-diameter co-immobilized beads. Flushing with sterile buffer solution for 5 hr activated these beads. The immobilized beads were packed into the fluidized bed bioreactor.

<table>
<thead>
<tr>
<th>M1 Medium for <em>Pseudomonas putida</em></th>
<th>M2 Medium for <em>Arthrobacter oxydans</em></th>
<th>M3 Medium for Mix Culture</th>
</tr>
</thead>
<tbody>
<tr>
<td>KH₂PO₄</td>
<td>0.035</td>
<td>0.035</td>
</tr>
<tr>
<td>K₂HPO₄</td>
<td>0.06</td>
<td>0.06</td>
</tr>
<tr>
<td>NH₄Cl</td>
<td>0.1</td>
<td>–</td>
</tr>
<tr>
<td>MgCl₂·6H₂O</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>MgSO₄·7H₂O</td>
<td>–</td>
<td>0.25</td>
</tr>
<tr>
<td>FeCl₂·4H₂O</td>
<td>–</td>
<td>0.0025</td>
</tr>
<tr>
<td>CuSO₄·5H₂O</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>CaCl₂</td>
<td>0.00074</td>
<td>0.00074</td>
</tr>
<tr>
<td>H₂O</td>
<td>1000 mL</td>
<td>1000 mL</td>
</tr>
<tr>
<td>Glucose</td>
<td>0.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Note: Units are g/1000 mL H₂O except where noted.

**Apparatus and Hydrogen Sulfide/Ammonia Removal for Continuous Operation**

A schematic set-up of the fluidized bed bioreactor is shown in Figure 1. Glass columns (60 mm φ × 60 cm of working height) were packed with cell-laden Ca-alginate beads, and a perforated sieve plate was fitted at the bottom of the column to ensure the uniform distribution of the inlet gas. The volumes of bead and inflow medium in the glass column were 600 and 520 mL, respectively. The initial number of cells in the column was determined to be 10⁶ CFU/g-bead. H₂S(g) or NH₃(g), supplied from separate gas cylinders, was diluted with compressed air passed through an air filter and flowed upward from the bottom of the biofilter. The gas flow rate was controlled to make the beads in the glass fluidized. The beads carrying the microorganisms were suspended in the solution, which the air was bubbled through. An inflow medium (composition shown in Table 1) was recirculated by a peristaltic pump at a flow rate of 25 mL/min to supply nutrient to the co-immobilized cells and modify the pH value.

In the continuous flow experiment, the H₂S- or NH₃-containing waste gas was introduced into the fluidized bed reactor. Gas concentrations in the range of 10–100 ppm were supplied at various flow rates (18, 27, 36, 54, and 72 L/hr). The empty bed detention times were 3.73, 2.49, 1.87, 1.24, and 0.92 min, respectively. The removal efficiency and the metabolic products of the biosystem were determined. In the co-immobilized system, the simulated H₂S- and NH₃-containing waste gases were prepared at ratios of 1:1 (30:30), 1:2 (30:60), 2:1 (60:30), and 2:2 (60:60), respectively, by concentration (ppm/ppm). These mixtures were supplied to the bioreactor at 36 L/hr, and the operating temperature was controlled at 28 °C. To evaluate the adaptability of the fluidized bed reactor to upset conditions, H₂S was sequentially fed into the reactor from low to high concentration (20, 40, 60, 80, and 110 ppm), and then the opposite method was used.

**Bioaerosol Analysis**

Microorganisms liberated from the bioreactor were collected by liquid impingement. The air escaped from the top of the bioreactor was forced through a 250-mL flask containing 100 mL of aseptically distilled water at 150 L/hr. One mL of the collected solution was inoculated to the medium, and the cell counts were determined by the serial dilution method. PDA, NA, thiosulfate, and modified Waksman media were used for fungi, heterotrophic bacteria, non-acidophilic *Thiobacillus*, and acidophilic *Thiobacillus*, respectively. The counts were reported as colony forming units in air (CFU/m³).
Concentrations of inlet and outlet H$_2$S/NH$_3$ gases of the reactor 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 0.5 to 120 ppm. In all consecutive experiments, the variation in H$_2$S/NH$_3$ concentration at steady state was within ±5%. Therefore, the 12 values obtained at steady states were averaged as the H$_2$S/NH$_3$ outlet concentrations. The chemical composition in the cycling solution was also determined.

**RESULTS AND DISCUSSIONS**

**Hydrogen Sulfide Removal**

A fluidized bed bioreactor packed with immobilized *Pseudomonas putida* bead was used to remove H$_2$S. This experiment was scheduled to continuously process for one month at 36 L/hr, while the inlet H$_2$S concentration was varied from 15 to 100 ppm. The removal efficiencies for various inlet concentrations at different periods are shown in Figure 2. When the inlet H$_2$S concentration was below 60 ppm, this system achieved greater than 92% removal efficiency. The average removal efficiency was greater than 95% for 15 ppm of inlet H$_2$S concentration. On the 15th day, a fresh nutrient solution was introduced into this system to start a new cycle. It was found that high removal efficiency was still maintained. As shown in Figure 2, the system achieved steady state in a short period of time. Therefore, the time to reach its optimum operation condition for H$_2$S removal was very short. It was also noted that the variation in pH was 6.3–7.5, showing very little acidification during the entire operation period.

In order to establish design criteria for the scale-up of the fluidized bed bioreactor, the correlation between inlet load and outlet load (removal capacity) needs to be studied. As shown in Figure 3, the dotted line represents the removal capacity at 100% removal efficiency, while the solid line represents 90% removal efficiency. When the inlet load was below 70 g·S/m$^3$·hr, the inlet load equaled the removal capacity. This point is called the critical loading state, since beyond this point, the system can no longer reach 100% removal. As inlet load exceeds the critical loading, the corresponding line curves downward to the right and deviates from the 100% coequal line. In addition, this system has a maximum inlet load of ~83.
Figure 2. H₂S removal efficiency of a fluidized bed bioreactor at 36 L/hr in continuous operation.

Figure 3. The relationship between the inlet load and the outlet load for H₂S.
g-S/m³-hr when the removal efficiency of the system is required to be 90% or more. Based on this graph, an operation model for H₂S removal can be established. If 100% H₂S removal is required, then the inlet load has to be controlled within 70 g-S/m³-hr. Similarly, if 90% H₂S removal is needed, then an inlet load within 83 g-S/m³-hr should be maintained. Since the inlet load of the system depends on inlet concentration and flow rate, any specific removal efficiency (e.g., 100% removal) can be achieved by adjusting the flow rate or the inlet concentration in compliance with emission limits.

Since the inlet concentration in the field operation is not constant, dramatic fluctuation can happen during the course of treatment. Therefore, it is necessary to demonstrate the adaptability of the system to dramatic change in inlet loading. A field operation was simulated by gradually increasing the inlet H₂S concentration from 20 to 110 ppm and then returning to the normal operation concentration (40 ppm). The correlation of inlet loading and outlet concentration in conjunction with the removal efficiency are shown in Figure 4. Greater than 96% removal efficiency was maintained in the first 32 hr of operation, as the inlet loading was below 75 g-S/m³-hr. However, when the inlet concentration was suddenly raised to 110 ppm at the 32nd hour, the outlet concentration increased to 13 ppm, a 7% drop in the removal efficiency. The system resumed its original removal efficiency (96%) when the H₂S concentration was returned to lower concentration after the 40th hour. This result indicates that our bioreactor system is able to cope with the fluctuation of inlet concentration during field operation. On the whole, this bioreactor system can achieve a good removal efficiency (>96%) if the inlet loading is controlled within 75 g-S/m³-hr.

### Ammonia Removal

A fluidized bed bioreactor packed with immobilized *Arthrobacter oxydans* bead was used to remove NH₃ ranging from 10 to 100 ppm under various flow rates (18, 27, 54, and 72 L/hr). The effect of the flow rate on removal efficiency at different inlet concentrations is shown in Figure 5. The removal efficiency decreased with the increasing inlet concentration and flow rate. Meanwhile, it was discovered that the impact of flow rate on removal efficiency within the concentration range of 10–100 ppm was the same, as shown by the same slopes. For a typical NH₃ concentration (60 ppm) emitted from various types of industries in Taiwan, this system can accomplish greater than 93% removal efficiency when the flow rate is kept at less than 54 L/hr, which confirmed the applicability of this system in reducing the NH₃ emission.

To establish the operation criteria in removing NH₃, we studied the correlation between inlet load and outlet concentration.

![Figure 4. The correlation of inlet loading and outlet concentration in conjunction with the removal efficiency by an abrupted condition.](image-url)
load (removal capacity). As shown in Figure 6, the critical loading was determined to be 3.7 g-N/m³-hr. To reach 90% removal efficiency, its maximum inlet load was ~7.5 g-N/m³-hr. The extrapolated correlation line suggested a maximum removal capacity of 9.0 g-N/m³-hr. There, acceptable NH₃ removal can be accomplished through adjusting the flow rate and the inlet concentration based on the correlation presented in Figure 6.

**Simultaneous Removal of Hydrogen Sulfide/Ammonia**

A fluidized bed bioreactor packed with co-immobilized *Pseudomonas putida* and *Arthrobacter oxydans* was used to simultaneously remove H₂S and NH₃. The removal efficiencies and pH variations for various ratios of H₂S/NH₃ gas mixtures (e.g., 1:1, 1:2, 2:2, and 2:1) at different periods are illustrated in Figure 7. During the initial 8 days (stage 1), an H₂S/NH₃ gas ratio of 1:1 (e.g., 30:30 ppm) was prepared as the inlet gas mixture. A gas ratio of 1:2 was operated from the 9th to the 16th day (stage 2). A gas ratio of 2:2 was run from the 17th to the 24th day (stage 3). During the final 8 days (stage 4), a gas mixture of 2:1 was used.

At stage 1, removal efficiencies of H₂S and NH₃ increased with time and reached the maximum values for the entire experiment, which are 97 and 95.5%, respectively. During the second period, the removal efficiency for H₂S in the gas mixture dropped from 97 to 93.5%, while the removal of NH₃ was about the same. Therefore, it was suggested that higher NH₃ concentration might affect the H₂S removal. Furthermore, the removal efficiency for H₂S dropped significantly (to 84%) in stage 3 at the inlet gas mixture of 2:2, a more significant drop than the 1:2 gas ratio. On the other hand, NH₃ was maintained at ~93%. The system can be used for at least 3 months, provided the cycling solution is routinely refreshed.

According to the results of this experiment, higher H₂S and NH₃ concentrations in the gas mixture can affect H₂S removal, especially the gas component of H₂S. However, the NH₃ removal is not affected by the concentration of the gas component in the mixture. Because the pH value of the system dropped to ~5, the decrease in H₂S removal could be attributed to the more acidic environment caused by the release of the H⁺ when H₂S and NH₃ were partially oxidized to SO₄²⁻ and NO₂⁻. During stages 3 and 4, the removal efficiency of H₂S was maintained at roughly 83% for an inlet gas mixture ratio of 2:1, and the removal efficiency of NH₃ was increased slightly. At the end of the experiment, the immobilized cells were sliced and observed under a scanning electronic microscope (SEM) to study the composition of these cluster cells. The SEM photograph (not shown) revealed that the spherical *Arthrobacter oxydans* outnumbered the rod-like *Pseudomonas putida*.

**Figure 5.** Effect of flow rate on NH₃ removal efficiency by the fluidized bed bioreactor.
Figure 6. The relationship between the inlet load and the outlet load for NH₃.

Figure 7. Relationships between the removal efficiency and operating time at different ratios (30:30, 30:60, 60:60, and 60:30) of H₂S/NH₃ gas mixtures.
The head loss was an important parameter for the operational cost. In this experiment, the gas velocity was raised gradually from 18 to 72 L/hr. When the operation reached the steady state, gas velocity was changed for the test. The averaged 64-cm H2O of head loss in the system (data not shown) was higher than a traditional biofilter (~10 cm). This might be a major disadvantage with respect to the conventional biofilter.

Kinetics Analysis
During the process of removing H2S/NH3, high concentrations of these compounds may inhibit the enzyme activity of Pseudomonas putida and Arthrobacter oxydans (mentioned in the section of simultaneous removal of H2S/NH3). Hence, inhibition in the system caused by high H2S and NH3 concentrations should be presented by analyzing the apparent kinetic parameters, which have successfully applied for the system.14,19 In the kinetics analysis, the gas flow rate was controlled to less than 36 L/hr to minimize the effect of mass-transfer limitation. The apparent kinetic parameters of the removal of single- and mixed-gas under various operating conditions are presented in Table 2. Generally speaking, the smaller the saturation constant (Ks), the higher the affinity of the substrate onto the bacteria cells and the better the effect of removal. In a co-immobilized system, the Ks of NH3 removal was 31 ppm when the inlet H2S/NH3 concentrations were between 5 and 60 ppm, which was very close to the Ks generated from the system of single gas removal (5–30 ppm of inlet NH3), namely 30.5. It is evident that the co-existence of H2S and NH3 did not affect the enzyme activity of Arthrobacter oxydans. When the inlet H2S/NH3 concentrations were between 5 and 30 ppm, the Ks of H2S was 75.8 ppm, which is close to the Ks of 76.2 ppm obtained from the single-gas system. This indicates that “low” NH3 concentration has no effect on H2S removal. When the inlet NH3 concentration in the gas mixture was increased to 60 ppm, the apparent kinetic parameter (Ks) for H2S was 106.5 ppm, which is higher than the Ks (76.2 ppm) obtained from a single H2S system.

This indicates that the enzyme activity of Pseudomonas putida is inhibited by 60 ppm NH3. When the inlet H2S concentration was increased to 60 ppm along with the addition of 5–30 ppm NH3, a saturation constant Ks of 186.3 ppm for H2S was obtained, which was much higher than the Ks of 106.5 ppm obtained from a gas mixture with a high NH3 concentration (60 ppm). Figure 7 also shows that the impact of high H2S concentration was more significant on H2S removal than a high NH3 concentration was. Therefore, if the inlet H2S/NH3 concentrations can be controlled to within 60 ppm, this system can be applied to remove the H2S- and NH3-containing waste gases effectively and simultaneously.

Product Analysis
To understand the mechanism of NH3/H2S removal, various concentrations of the gases were introduced into a bioreactor at 36 L/hr for 7 consecutive days. Then the recycling solution was collected for product analysis. Tables 3 and 4 present the results of this analysis. Since the concentrations of metabolic products in the immobilized bed are far less than those in the recycling solution (data not shown), only the data regarding the recycling solution are given here.

As seen in Table 3, the amounts of S, SO4, SO3, and S2– are 162.12, 5.88, 33.76, and 0.02 (mg-S), respectively, at an inlet H2S concentration of 30 ppm. Among these metabolic products, elemental sulfur attained the highest percentage (80.3%), whereas the sulfide ions attained the least, only 0.1%. When the inlet H2S concentration was changed to 60 ppm, the neutral sulfur remained the major metabolic product, at 84.5%. Obviously, these neutral sulfur products will not bring acidity to the system, and sulfur with economical value can be recycled. However, the higher concentration of sulfide indicated that part of the H2S was not metabolized by sulfur-oxidizing bacteria. Fortunately, these sulfide ions made up a very low ratio, with an average of 0.15%. This suggested that most H2S was converted into metabolic products and stored in the recycling solution. Hence, the

Table 2. Apparent kinetic analysis for H2S and NH3 removal by a fluidized bed bioreactor.

<table>
<thead>
<tr>
<th></th>
<th>Single Inlet</th>
<th>Mixture Inlet</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NH3 = 0 ppm</td>
<td>NH3 = 0 ppm</td>
</tr>
<tr>
<td>Ks</td>
<td>76.2×10^-8</td>
<td>75.8</td>
</tr>
<tr>
<td>Vm</td>
<td>7.0×10^-8</td>
<td>7.2×10^-4</td>
</tr>
<tr>
<td>NH3</td>
<td>—</td>
<td>1.88×10^-8</td>
</tr>
</tbody>
</table>

ppm; 1g-S/cell/day; 2g-N/cell/day.
sulfur balance (average value) in Table 3 can be described as follows:

\[
H_2S \rightarrow 0.149 SO_4^{2-} + 0.824 S + 0.026 SO_3^{2-} + 0.0015 S_2^- 
\]

As shown in Table 4, the compositions of metabolic products are very similar regardless of the inlet NH\(_3\) concentration. Among these metabolic products, NO\(_2^-\)N is the main product, taking up an average of 76.8%, followed by organic nitrogen, at 14.7% (as an average). Literature also shows that the main metabolic products from NH\(_3\) through *Arthrobacter oxydans* are NO\(_2^-\) and organic nitrogen.\(^{19}\) Therefore, the presence of NH\(_3\) indicates that some of it was not oxidized. At the same time, NH\(_3\) in the recycling solution accumulates significantly as the inlet concentration increases. NO\(_3^-\) was not produced by *Arthrobacter oxydans*. It is likely that NO\(_3^-\) was chemically oxidized to NO\(_3^-\) after the sample was removed. The nitrogen balance (average value) in Table 4 can be described as follows:

\[
NH_3 \rightarrow 0.768 NO_2^- + 0.039 NO_3^- + 0.072 NH_4^+ + 0.147 N
\]

### Table 3. Metabolic products of H\(_2\)S by a fluidized bed bioreactor.

<table>
<thead>
<tr>
<th>Inlet H(_2)S Concentration (ppm)</th>
<th>SO(_4^{2-}) Produced (mg-S)</th>
<th>SO(_3^{2-}) Produced (mg-S)</th>
<th>NO(_2^-)-N Produced (mg-N)</th>
<th>NO(_3^-)-N Produced (mg-N)</th>
<th>Organic N Produced (mg-N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>162.12 (80.3%)</td>
<td>5.88 (2.9%)</td>
<td>52.2 (76.2%)</td>
<td>2.7 (3.9%)</td>
<td>9.1 (13.3%)</td>
</tr>
<tr>
<td>60</td>
<td>376.12 (84.5%)</td>
<td>10.4 (2.3%)</td>
<td>159.5 (78.5%)</td>
<td>ND</td>
<td>29.2 (14.3%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>314.5 (75.8%)</td>
<td>ND</td>
<td>67.9 (16.4%)</td>
</tr>
</tbody>
</table>

Note: ND indicates experimental data above MDL. The MDL of NO\(_3^-\)-N is 0.0137 mg-N.

As shown in Table 4, the compositions of metabolic products are very similar regardless of the inlet NH\(_3\) concentration. Among these metabolic products, NO\(_2^-\)-N is the main product, taking up an average of 76.8%, followed by organic nitrogen, at 14.7% (as an average). Literature also shows that the main metabolic products from NH\(_3\) through *Arthrobacter oxydans* are NO\(_2^-\) and organic nitrogen.\(^{19}\) Therefore, the presence of NH\(_3\) indicates that some of it was not oxidized. At the same time, NH\(_3\) in the recycling solution accumulates significantly as the inlet concentration increases. NO\(_3^-\) was not produced by *Arthrobacter oxydans*. It is likely that NO\(_3^-\) was chemically oxidized to NO\(_3^-\) after the sample was removed. The nitrogen balance (average value) in Table 4 can be described as follows:

\[
NH_3 \rightarrow 0.768 NO_2^- + 0.039 NO_3^- + 0.072 NH_4^+ + 0.147 N
\]

### Table 5. Bioaerosol concentrations in the exhaust of a fluidized bed bioreactor.

<table>
<thead>
<tr>
<th>Mixture Ratio (H(_2)S/NH(_3))</th>
<th>Nutrient Agar</th>
<th>Medium Types</th>
<th>Medium Types</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A. oxydans</td>
<td>Thiosulfate</td>
<td>Nitrifying</td>
</tr>
<tr>
<td>1:1</td>
<td>115</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td>1:2</td>
<td>173</td>
<td>57</td>
<td>ND</td>
</tr>
<tr>
<td>2:1</td>
<td>173</td>
<td>ND</td>
<td>ND</td>
</tr>
</tbody>
</table>

Note: ND < 57 CFU/m\(^3\); a1:1 = 30:30 (ppm/ppm).

### Bioaerosol Detection

Biotreatment systems have recently become a major interest in removing malodor. However, as large quantities of gases are treated, the liberation of microbes from the bioreactor has to be considered. Because a large number of microbes are contained in the system, they can have a severe impact on the environment if released accidentally. In this experiment, aseptically distilled water was used to absorb the exhaust air, and the numbers of bioaerosols contained are counted as listed in Table 5. The results indicate that only very few heterotrophic *Arthrobacter oxydans* (115–173 CFU/m\(^3\)), *Pseudomonas putida* (115–173 CFU/m\(^3\)), and fungi (57 CFU/m\(^3\)) were present in the exhaust air, because the microbes were immobilized in the beads of Ca-alginate. The amount contained in these bioaerosols was far less than that released from some conventional biofilters.\(^{23}\) The bioaerosol analysis assures the safety of our system to the environment.

### CONCLUSIONS

The fluidized bed bioreactor, when used to remove H\(_2\)S, needs only a short time to reach its steady state. The metabolic products of the systems will not acidify the reactor because the main product is neutral sulfur. Since the sulfur element can be reclaimed, this makes the bioreactor system even more competitive. Under appropriate operating conditions, the removal efficiency can reach a satisfactory high (95% or higher) when H\(_2\)S or NH\(_3\) is processed individually. In a co-immobilized system, relatively high H\(_2\)S/NH\(_3\) concentrations have no inhibitory effect on the enzyme activity of *Arthrobacter oxydans*. On the other hand, both H\(_2\)S and NH\(_3\) at high concentrations inhibit the enzyme activity of *Pseudomonas putida*. The influence of H\(_2\)S on enzyme activity is far greater than that of NH\(_3\). The bioaerosols released from the reactor were discovered to be quite low, due to the use of an immobilized cell, which ensures its safety concerning environmental impact. It is concluded that this system may be operated near areas with high population density.
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