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Development of a two-phase bioreactor for the biological removal of hydrogen sulfide from biogas

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Abstract

In this study, a two-phase bioreactor, consisting of an anaerobic absorption column and an aerobic biofilter, was constructed and utilized to determine the removal efficiency of high strength hydrogen sulfide from biogas. A microbial strain of sulfur oxidizing bacteria (SOB), *Acidithiobacillus thiooxidans*, isolated from earthworm casts was inoculated in the aerobic biofilter. Initially, an inlet concentration of hydrogen sulfide supplied to the two-phase bioreactor was 180 ppm, and overall removal efficiencies of hydrogen sulfide were 30 to 60% due to low cell density and activity of the SOB in the bioreactor. As bioreactor operation continued, microbial activity and bioreactor performance increased with sharply decreasing pH levels from 6.3 to 1.5. During the same operational period, the optical density (OD_{600}) increased from 0.05 to 0.4, indicating that the SOB was in an experiential growth stage at the low pH condition. The overall removal efficiency of hydrogen sulfide was found to be greater than 97% from day 8, and the high removal efficiency maintained. On day 30, the inlet concentration of hydrogen sulfide was increased to 400 ppm, and the removal efficiency did not decline, showing that the activity of the SOB was high enough to handle the high loading rate of hydrogen sulfide. An interesting finding of the study was that the activity of the SOB strain used in this study was not affected at the extremely low pH values, and no chemical additives were required to maintain the pH. Dissolved oxygen concentrations in the anaerobic column and the aerobic biofilter were maintained at 2 and 8 mg/L, respectively. Consequently, the two-phase bioreactor showed advantages over conventional biofilters for the treatment of high strength hydrogen sulfide from biogas.

Keywords : Biogas, Hydrogen sulfide, Sulfur oxidizing bacteria (SOB), Bioreactor

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1. Introduction

Recently, various attempts have been made to develop renewable and alternative energy sources to minimize global environmental problems and green house gas emissions. As one of the prominent renewable energy sources, biogas produced by anaerobic processes using industrial and domestic wastes has been emerged as a green energy source as well as an alternative to reduce green house gas emissions. At many domestic landfill sites, biogas is being collected and used to generate heat and electrical energy. As a result, the effective use of biogas not only supplies a sustainable energy source but also minimize a release of methane, which is the main component of biogas and its green house effect is approximately 21 times higher than carbon dioxide (Lee et al., 2003).

However, biogas produced from domestic landfill sites commonly contains impurities such as hydrogen sulfide. Hydrogen sulfide can be generated from a variety of sources, and it is a very strong malodorous compound detected even at a low concentration lower than 0.1 ppm. Many investigations have reported that a concentration range of hydrogen sulfide from biogas was found to be as high as 5000 ppm. The impurity of biogas, therefore, needs to be pre-treated to avoid facility corrosion, unnecessary production of byproducts, and possible public exposure and complaints (Lee et al., 2007; Taliwa et al., 2009).

General treatment processes to remove hydrogen sulfide from biogas include physical, chemical and biological methods. Among them, biological processes for the treatment of hydrogen sulfide have been investigated and developed because of their relatively low operating costs and minimal generation of undesirable byproducts (Park et al., 2006). In bioreactor systems, hydrogen sulfide in the gas phase is first dissolved into microbial media containing sulfur oxidizing bacteria (SOB), and the bacteria subsequently oxidize hydrogen sulfide in the presence of oxygen in the liquid phase(Duan et al., 2006; Park et al., 1999). In addition, to apply biological methods to remove high-strength hydrogen sulfide in a biogas stream, bioreactor systems requires high elimination capacity and stability under severe operating conditions. Some studies using packed-bed bioreactors reported the maximum elimination capacity for hydrogen sulfide from 8 to 55 g-H_2S/m^3/hr (Kim et al., 2008; Ramirez et al., 2009; Ryu et al., 2004).

In this study, a two-phase bioreactor, consisting of an anaerobic absorption column and an aerobic biofilter, was constructed and utilized to determine the removal efficiency of high strength hydrogen sulfide from biogas. A microbial strain of SOB, *Acidithiobacillus thiooxidans* isolated from earthworm casts was inoculated in the aerobic biofilter. Since the SOB required an external oxygen supply for microbial respiration and hydrogen sulfide oxidation, the process needed to maintain different oxygen contents in each bioreactor unit. Therefore, in this study, the concentration of dissolved oxygen in the bioreactor was monitored over the operational period.

2. Materials and Methods

2.1. Two-phase bioreactor

In this study, a lab-scale two-phase bioreactor has been constructed as shown in Figure 1. The two-phase bioreactor consisted of the anaerobic absorption column and the aerobic biofilter, and an effective volume of each bioreactor was 2 L. A polyurethane foam material was packed in each reactor with a filled volume of 1.5 L each. The packing material was used to increase the contact surface area between gaseous hydrogen sulfide and microbial media.

In order to mimic the anaerobic (i.e., no oxygen) condition of biogas in the laboratory, the gas stream was mainly made up with nitrogen gas. A controlled volume of 0.5% hydrogen sulfide was combined

with the nitrogen gas stream at a mixing chamber, and the mixed gas stream at a total flow rate of 0.5 L/min was introduced to the bottom of the anaerobic absorption column. Into the aerobic biofilter unit, purified air at an air flow rate of 2.0 L/min was introduced through a fine-bubble sparger to supply oxygen. As shown in Figure 1, gas sampling ports were installed on the inlet and outlet parts to measure the concentration of hydrogen sulfide in the gas phase, and liquid samples were taken from the port located along each unit.

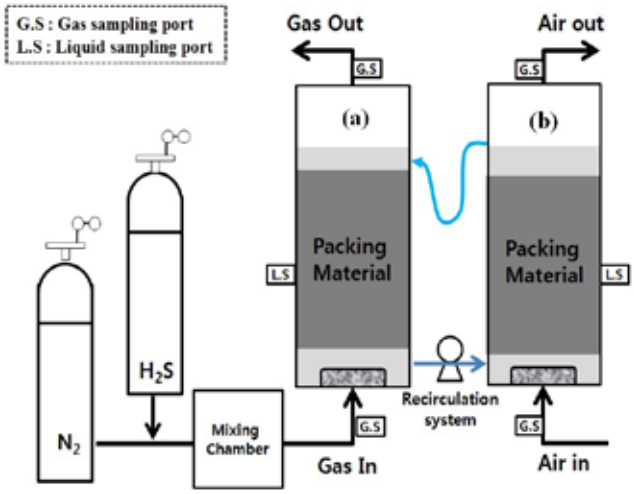

Fig. 1. Schematic diagram of the two-phase bioreactor, (a) the anaerobic absorption column and (b) the aerobic biofilter

2.2. Microorganism

In order to effectively remove hydrogen sulfide, the SOB strain, *Acidithiobacillus thiooxidans* was inoculated to the bioreactor. Prior to the inoculation, the SOB culture was pre-cultivated in a 2-L sterile medium in a 5-L flask in the presence of oxygen. The liquid medium used to cultivate the SOB strain contained sodium thiosulfate ($Na_2S_2O_3$) 5.1 g/L, K_2HPO_4 2.0 g/L, KH_2PO_4 2.0 g/L, NH_4Cl 0.4 g/L, $MgCl_2 \cdot 7H_2O$ 0.2 g/L, and $FeSO_4 \cdot 7H_2O$ 0.01 g/L. For the bioreactor operation, sodium thiosulfate was not added to the liquid medium.

2.3. Operation condition of bioreactor

At the beginning of the bioreactor operation, an inlet concentration of hydrogen sulfide was maintained at 180 ppm for 30 days, and the inlet concentration was step-wise increased to 400 ppm for three additional days. At the given hydrogen sulfide concentrations, the overall loading rates were 3.75 and 8.0 $g/m^3/hr$, respectively. The liquid medium was circulated through the two units at 24 L/hr, which was equivalent to a recirculation ratio of six reactor volume per hour. Also, once every two days, 200 mL of the liquid medium was withdrawn and replaced with a fresh medium to supply necessary trace metals.

2.4. Method of analysis

In this study, the hydrogen sulfide concentration in the gas phase was directly measured using an electrochemical detector (GFM series, GASDATA, UK). The liquid samples were measured for pH

(Satorious, USA), dissolved oxygen concentration (YSI, USA), and optical density at 600 nm using a spectrophotometer (Simadzu, Japan).

3. Result and Discussion

3.1. Microbial activity and pH

In order to determine the activity of SOB inoculated to the bioreactor, changes in pH and optical density (OD @ 600 nm) was measured. Following the bioreactor start-up, the pH of the microbial medium was maintained at the initial value, because the microorganisms did not effectively remove hydrogen sulfide from the gas phase. After five days from the start-up of the bioreactor experiment, the pH sharply decreased from 6.3 to lower than 1.5 as shown in Figure 2. During the same operational period, the OD as a measure of microbial cell quantity increased from 0.05 to 0.4, indicating that the SOB strain was in an exponential growth stage and its activity was not affected at the extremely low pH condition. Therefore, *A. thiooxidans*, the SOB strain used in this study was suitable for the removal of hydrogen sulfide at a high concentration, and no chemical additives were required to maintain pH in a neutral condition.

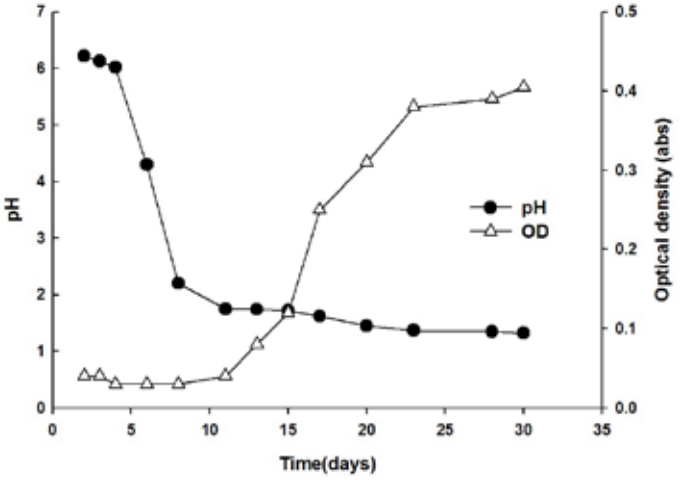

Fig. 2. Optical density and pH changes in the two-phase bioreactor

3.2. Removal efficiency of hydrogen sulfide

During the first 30-day period, the inlet concentration of hydrogen sulfide introduced to the two-phase bioreactor was maintained at 180 ppm. Initially, as shown in Figure 3, the removal efficiency of hydrogen sulfide microbial was unstable and varied from 30 to 60% due to low cell density and activity of the microbial strain in the bioreactor. With increasing microbial density and activity, the overall efficiency increased to greater than 97% after 8 days. An interesting finding of this study was that the microbial strain showed a high growth rate and elimination capacity for hydrogen sulfide after the pH value dropped below 2.0.

On day 30, the inlet concentration of hydrogen sulfide was increased to 400 ppm to determine whether the two-phase bioreactor could effectively treat a higher loading rate. At the higher loading rate, the

overall removal efficiency of hydrogen sulfide did not drop, implying that the activity of the microbial strain was high enough to handle the high loading rate of hydrogen sulfide. At the high loading rate, the pH value was also maintained at approximately 1.5. These results indicated that the two-phase bioreactor can be an effective control method for the removal of high strength hydrogen sulfide from biogas.

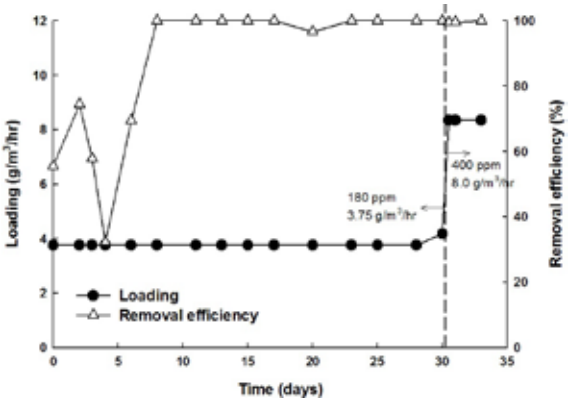

Fig. 3. Hydrogen sulfide loading rates and overall removal efficiencies in the two-phase bioreactor

3.3. DO concentration

As one of the important operating parameters of the two-phase bioreactor, the concentration of dissolved oxygen (DO) in each bioreactor unit was monitored. The SOB requires liquid-phase oxygen for microbial respiration and hydrogen sulfide oxidation, but the biogas stream generally does not contain oxygen. In addition, this pre-treatment process should not alter the methane content to ensure the biogas quality. As a result, the two-phase process needed to maintain different DO concentrations in each bioreactor unit, and appropriate DO concentrations were suggested to be less than 2 mg/L in the anaerobic absorption column and greater than 3 mg/L in the aerobic biofilter to increase the biological oxidation rate. The actual DO concentrations were found to be 2 and 7.8 mg/L in the anaerobic absorption column and the aerobic biofilter, respectively. Consequently, the results showed that each unit of the two-phase bioreactor was operated under proper conditions.

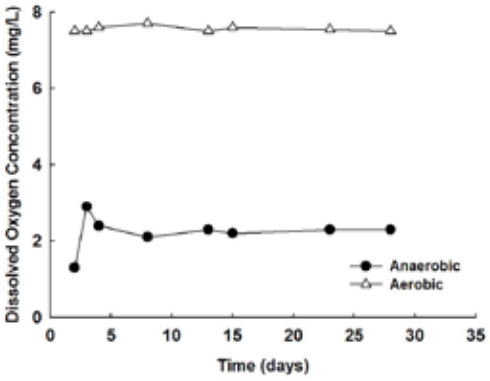

Fig. 4. Dissolved oxygen concentration during operation of a two-phase bioreactor

4. Conclusions

In this study, the two-phase bioreactor consisting of the anaerobic absorption column and the aerobic biofilter, was applied to treat high strength hydrogen sulfide from biogas. A microbial strain of SOB, *Acidithiobacillus thiooxidans* was inoculated in the bioreactor. The key observations and conclusions that can be drawn from this study are given below.

- The SOB strain, *A. thiooxidans* used in this study was suitable for the removal of hydrogen sulfide at a high concentration. The microbial strain was in the exponential growth stage and its activity was not affected at the extremely low pH condition; therefore, no chemical additives were required to maintain pH in a neutral condition.
- At the pseudo-steady-state, the activity of the SOB strain was high enough to treat the high loading rate of hydrogen sulfide. The two-phase bioreactor could be applicable for the removal of high strength hydrogen sulfide from biogas.
- Consequently, the two-phase bioreactor using the separated absorption and biodegradation units in series could be an effective alternative over conventional biofilter systems and other physicochemical technologies treating biogas impurities.

Acknowledgements

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