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Removal of residual dissolved methane gas in an upflow anaerobic sludge blanket reactor treating low-strength wastewater at low temperature with degassing membrane

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ABSTRACT

In this study, we investigated the efficiency of dissolved methane (D-CH_4) collection by degasification from the effluent of a bench-scale upflow anaerobic sludge blanket (UASB) reactor treating synthetic wastewater. A hollow-fiber degassing membrane module was used for degasification. This module was connected to the liquid outlet of the UASB reactor. After chemical oxygen demand (COD) removal efficiency of the UASB reactor became stable, D-CH_4 discharged from the UASB reactor was collected. Under 35°C and a hydraulic retention time (HRT) of 10 h, average D-CH_4 concentration could be reduced from 63 mg COD L^{-1} to 15 mg COD L^{-1}; this, in turn, resulted in an increase in total methane (CH_4) recovery efficiency from 89% to 97%. Furthermore, we investigated the effects of temperature and HRT of the UASB reactor on degasification efficiency. Average D-CH_4 concentration was as high as 104 mg COD L^{-1} at 15°C because of the higher solubility of CH_4 gas in liquid; the average D-CH_4 concentration was reduced to 14 mg COD L^{-1} by degasification. Accordingly, total CH_4 recovery efficiency increased from 71% to 97% at 15°C as a result of degasification. Moreover, degasification tended to cause an increase in particulate COD removal efficiency. The UASB reactor was operated at the same COD loading rate, but different wastewater feed rates and HRTs. Although average D-CH_4 concentration in the UASB reactor was almost unchanged (ca. 70 mg COD L^{-1}) regardless of the HRT value, the CH_4 discharge rate from the UASB reactor increased because of an increase in the wastewater feed rate. Because the D-CH_4 concentration could be reduced down to 12 ± 1 mg COD L^{-1} by degasification at an HRT of 6.7 h, the

CH_4 recovery rate was 1.5 times higher under degasification than under normal operation.

Keywords: Dissolved methane gas; Degassing membrane; Anaerobic wastewater treatment; Low temperature; Low-strength wastewater

Introduction

Anaerobic wastewater treatment is a well-established and proven technology for the treatment of various categories of industrial wastewaters (Cakir and Stenstrom, 2005). This technology has numerous advantages, such as low energy requirement and energy recovery as methane (CH_4) gas, over aerobic wastewater treatment systems. Most anaerobic wastewater treatments have been conducted within mesophilic (30–40°C) or thermophilic (45–60°C) temperature ranges (Kashyap et al., 2003; Dhaked et al., 2010; Fang et al., 2006; Lettinga et al., 2001). This is attributed to the fact that most of the biological reactions responsible for anaerobic biodegradation of organic matter proceeds much slower under psychrophilic (<20°C) conditions than under mesophilic conditions. However, domestic wastewater and a variety of industrial wastewaters, such as those from bottling, malting, and soft drinks manufacturing plants and breweries, are generally discharged at low ambient temperatures under temperate climatic conditions. Furthermore, domestic sewage wastewaters belong to the category of "low-strength wastewaters" that have a chemical oxygen demand (COD) concentration of ca. 1.0 g L^{-1} or lower. Therefore, a significant input of energy is required to heat the reactor to the treatment temperature (Lettinga et al., 2001; Angenent et al., 2001). If anaerobic wastewater treatment without heating the reactors can be applied to low-strength wastewater, the cost of anaerobic wastewater treatment can be reduced, thereby making this technology an attractive option for the treatment of a variety of wastewater categories.

Improvements in reactor design and operational conditions have helped overcome many of the disadvantages of anaerobic wastewater treatment that limited its application to high-strength wastewater treatment at mesophilic or thermophilic temperatures and have allowed for successful operation of anaerobic wastewater treatment reactors at low temperatures (Dhaked et al., 2010; Lettinga et al., 1999; Lettinga et al., 2001; Trzcinski and Stuckey 2010; Luostarinen and Rintala 2005; Madden et al., 2010; Xing et al., 2009; McKeown et al., 2009). In addition, several studies have focused on anaerobic wastewater treatment of low-strength wastewaters at lower temperatures (Elmitwalli et al., 2003; Angenent et al., 2001; Gomec et al., 2008; Matsushige et al., 1990). A drop in temperature causes a change in the physical and chemical properties of wastewater, and this can significantly affect the reactor performance. For instance, dissolved methane gas (D-CH_4) might play an important role in energy recovery efficiency of the reactor; however, to date, the role of D-CH_4 has been overlooked. Discharge of residual D-CH_4 in the reactor effluent represents a loss of energy that may be recovered; in addition, D-CH_4 is a source of CH_4, a greenhouse gas, that may be released into the environment (Hartley and Lant, 2006; Hatamoto et al., 2010; Matsuura et al., 2010). This knowledge is particularly important when low-strength wastewaters are anaerobically treated at low temperature, because the solubility of CH_4 in the liquid phase increases with a decrease in temperature. Few studies have investigated the removal of D-CH_4 in anaerobic wastewater treatment processes by biological oxidation, physical gasification based on gas–liquid equilibrium, and mixing with gas or a paddle (Hartley and Lant, 2006;

Hatamoto et al., 2010; Matsuura et al., 2010; Pauss et al., 1990). However, the recovery efficiency of D-CH_4 was low and/or the recovered CH_4 gas composition was low in these processes. Hence, another technology is required for the removal of D-CH_4 (Voolapalli and Stuckey, 1998).

In this study, we employed a hollow-fiber membrane to recover residual D-CH_4 in the effluent of an anaerobic wastewater treatment reactor by degasification. A bench-scale upflow anaerobic sludge blanket (UASB) reactor was operated, and the liquid outlet of the UASB reactor was connected to another reactor for degasification. After achieving stable COD removal efficiency, the D-CH_4 discharged from the UASB reactor was recovered by the degassing membrane (DM) reactor. In addition, we investigated the effects of temperature and hydraulic retention time (HRT) of the UASB reactor on the efficiency of degasification.

1. Materials and methods

2.1. Experimental setup and operational conditions

A bench-scale UASB reactor (height, 40 cm; diameter, 7 cm; working volume, 1.3 L) was operated in this study. The reactor was inoculated with 0.3 L of anaerobic granular sludge, which had total and volatile solids concentrations of 28 g L^{-1} and 22 g L^{-1}, respectively, obtained from a full-scale UASB reactor treating wastewater from an isomerized sugar-processing plant. The UASB reactor was covered with a water jacket to maintain the reactor at a constant temperature (15, 25, or 35°C). The reactor was fed with synthetic wastewater containing powdered skim milk as carbon and energy sources, inorganic salts, and trace metals (Satoh et al., 2007). COD concentration was controlled by changing the concentration of powdered milk. HRT was adjusted by altering the wastewater feed rate. These operational conditions are summarized in Table 1. pH of the bulk liquid in the UASB reactor was maintained by adding NaOH at 7.6 after 51 days.

After gas production reached a steady state, dissolved gas in the liquid was collected by the DM. A reactor for degasification (height, 30 cm; diameter, 7 cm; working volume, 1.1 L) was connected to the liquid outlet of the UASB reactor to collect the residual D-CH_4 in the UASB effluent by degasification. A hollow-fiber membrane module (a multi-layered composite hollow-fiber membrane; MHF0504MBFT) provided by the Mitsubishi Rayon Co., Ltd. (Tokyo, Japan) was installed in the DM reactor. The DM reactor was completely filled with the wastewater treated in the UASB reactor. The liquid in the DM reactor was not mixed. The characteristics of the DM module are summarized in Table 2. The dissolved biogas diffuses into the lumen of the hollow fibers of the DM under vacuum generated using a vacuum pump (Model APN-110KV-1; Iwaki Co., Ltd., Tokyo, Japan). Although gas molecules can pass through the non-porous layer of the membrane, liquids cannot. Thus, the DM effectively separates dissolved gas from the liquid. The DM reactor was operated at same temperatures as the UASB reactor. Transmembrane pressures were set at 50 kPa and 80 kPa (absolute pressure) by using a vacuum gauge, and HRTs of the

DM reactor were altered in accordance with variations in the HRTs of the UASB reactor (Table 1). The operation of the DM reactor without degasification is referred to as normal operation.

2.2. Analytical methods

The concentrations of total COD (T-COD) and dissolved fraction of COD (D-COD) after filtration with a 0.45-μm-pore-size membrane (Advantec Co., Ltd., Tokyo, Japan) were measured using a Hach method (Method 8000). Particulate COD (P-COD) concentration was calculated by subtracting the D-COD concentration from the T-COD concentration. The concentrations of volatile fatty acids (VFAs; formate, acetate, propionate, lactate, i-butyrate, and n-butyrate) were determined by using a high-performance liquid chromatography system (LC-10AD system; Shimadzu Co., Kyoto, Japan) equipped with a Shimadzu Shim-pack SCR-102H column (0.8 × 30 cm) after filtering it through a 0.2-μm-pore-size membrane. The oxidation–reduction potential (ORP) and pH were directly determined by using an ORP and a pH electrode, respectively.

Concentrations of CH_4, CO_2, N_2, O_2, and H_2 in the headspace of the UASB reactor and inside the lumen of the hollow fibers of the DM were measured by using a gas chromatography system (GC-14B; Shimadzu Co., Kyoto, Japan) equipped with a thermal-conductivity detector and a Shincarbon-ST column (Shimadzu Co., Kyoto, Japan). The biogas volumes were measured at 25°C. The dissolved gas compositions were measured by using the headspace method (Bandara et al., submitted). Liquid samples (50 mL) were obtained from the UASB and DM reactors and were injected with a syringe into a sealed vial (125 mL) that was prefilled with 100% argon gas. We then added 1 mL of 20 mM mercury (II) chloride to the vial to inhibit biological reactions. The vial was vigorously shaken for 5 min to allow for diffusion of the dissolved gas in the liquid sample into the headspace. The liquid sample was allowed to stand for 30 min at room temperature to equilibrate the gas and liquid phases. Then, the composition of the headspace gas was determined using gas chromatography. The standard liquid sample with known concentration of each standard gas was prepared as follows. A vial (125 mL) was filled with 50 mL of distilled water. Argon gas was blown into the distilled water in the vial for 5 min to remove air in the liquid and gas phases. The vial was then sealed with a butyl rubber septum. Next, we injected 1 mL of 100% of each gas, corresponding to 820 μmol at 25°C, to the vial. The vial was shaken and allowed to stand for 30 min, and then, the composition of the headspace gas was determined by gas chromatography. The concentrations of dissolved gases in the sample liquid were calculated on the basis of the ratio of the amount of gas in the sample to that in the standard sample. All measurements were conducted at a constant temperature of 25°C.

On the basis of these results, D-CH_4 collection efficiency was calculated as a ratio of D-CH_4 concentration collected by the DM to D-CH_4 discharged from the UASB reactor. The D-CH_4 concentration in the DM was calculated as the difference between the D-CH_4 concentrations discharged from the UASB and DM reactors. Total

CH_4 recovery efficiency was calculated as a ratio of the CH_4 recovery rate (mg COD L^{-1} day^{-1}) to the total CH_4 production rate (mg COD L^{-1} day^{-1}). The CH_4 recovery rate is defined as the sum of the CH_4 evolution rate in the UASB headspace and CH_4 collection rate in the DM. The total CH_4 production rate is defined as the sum of the CH_4 evolution rate, CH_4 collection rate, and CH_4 discharge rate from the DM reactor.

2. Results and discussion

3.1. Performance of the UASB reactor

The bench-scale UASB reactor was operated at different temperatures and HRTs for 170 days, and concentrations of influent T-COD and effluent T-COD and D-COD in the UASB reactor were measured (Figure 1). The average T-COD concentration (± standard deviation) of the influent was 1480 ± 240 mg COD L^{-1} from startup to day 128 (by phase 4), after which the influent T-COD concentrations decreased. Even at the beginning of reactor operation, D-COD removal efficiency was high (Figure 1), probably because of the high amount of biomass, although the influent and effluent T-COD concentrations fluctuated. The average effluent D-COD concentration was 120 ± 40 mg COD L^{-1} by day 128, resulting in a D-COD removal efficiency as high as 92 ± 2%. Total VFA concentration in the UASB reactor was less than 68 mg COD L^{-1} for 170 days (data not shown). The dominant VFA was acetate, with a maximal concentration of 53 mg COD L^{-1} and average concentration of 28 ± 14 mg COD L^{-1} for

170 days. The second dominant VFA was propionate (9 ± 6 mg COD L^{-1}); the concentrations of other VFAs (lactate, formate, i-butyrate, and n-butyrate) were less than 1 mg COD L^{-1}.

3.2. Performance of the DM reactor

Since the reactor performance (e.g., the D-COD removal efficiency of the UASB reactor) became stable after adjusting the pH to 7.6 after day 51, degasification was conducted at a transmembrane pressure (absolute pressure) of 50 kPa from day 64 (phase 1.2) to recover residual dissolved CH_4 (D-CH_4) in the UASB effluent. The average D-CH_4 concentration was 61 ± 6 mg COD L^{-1} during normal operation (phase 1.1); in contrast, it was 20 ± 4 mg COD L^{-1} in the effluent of the DM reactor during a degasification period (Phase 1.2) (Figure 2). The difference between the D-CH_4 concentrations in the effluents of the UASB and DM reactors indicated that D-CH_4 was successfully collected by the DM during a degasification period. The transmembrane pressure was further reduced by 20 kPa (absolute pressure) at day 78 (phase 1.3) to reduce the residual D-CH_4 in the UASB effluent. The D-CH_4 concentration was 15 ± 5 mg COD L^{-1} in the DM reactor effluent in phase 1.3 (Figure 2). On the basis of these results, D-CH_4 collection efficiencies were calculated to be 68 ± 7% and 77 ± 7% in phases 1.2 and 1.3, respectively. When the DM reactor was applied to pure water at 0.3 kPa, dissolved oxygen concentration was decreased to less than 0.1 mg L^{-1} after 5 min, indicating that the theoretical dissolved gas collection efficiency of the DM was almost

100%. To check for reproducibility of the efficiency of degasification, vacuum was discontinued at day 85 (phase 1.4). The average D-CH_4 concentration was 63 ± 1 mg COD L^{-1} in the DM reactor effluent in phase 1.4, and this concentration was comparable to that before degasification (phase 1.1) (Figure 2). These results clearly indicated that D-CH_4 was successfully collected by degasification with the DM.

Discharge of residual D-CH_4 in the effluent of wastewater treatment reactors contributes to an increase in atmospheric CH_4, which is 21 times more potent than CO_2 as a greenhouse gas (Hartley and Lant, 2006). In addition, the D-CH_4 discharge represents a loss of energy that is generated in anaerobic wastewater treatment processes. Therefore, recovery of D-CH_4 from the effluent of the anaerobic wastewater treatment reactors is a prerequisite for the discharge of the effluent into the environment.

Gas compositions in the headspace of the UASB reactor (Figure 3A) and inside the lumen of the hollow fibers of the DM (Figure 3B) are shown in Figure 3. Although gas compositions fluctuated until day 50, they became relatively stable thereafter because of pH adjustment to 7.6. Average CH_4 compositions in the UASB headspace during the degasification period (phases 1.2 and 1.3) were 51 ± 4% and 58 ± 5%, respectively, and they were 49 ± 12% and 56 ± 3% during normal operation (phases 1.1 and 1.4, respectively) (Figure 3A). Thus, degasification did not significantly affect the CH_4 composition in the UASB headspace. Average compositions of CH_4 and CO_2 collected through the DM during the degasification period were 22 ± 13% and 10 ± 7%, respectively, in phase 1.2 and 20 ± 2% and 27 ± 6%, respectively, in phase 1.3 (Figure

3B).

O_2 was detected in the biogas collected through the DM; this indicated the ingress of air into the dissolved gas collection line. The percentage of N_2 plus O_2 in the collected biogas varied from 39% to 87% in phase 1. The ratios of N_2 to O_2 were 3.9 or higher, and these ratios were greater than the ratio of N_2 to O_2 in air (79/21 or 3.76). This was because the dissolved biogas itself contained N_2 but not O_2. If air had been excluded from the collected biogas, the average CH_4 composition in the collected gas would have been 63 ± 15% in phase 1.2, which would have been comparable to that in the UASB headspace.

Several approaches for removal and/or recovery of D-CH_4 in wastewater treatment processes have been investigated. Hatamoto et al. (2010) used an encapsulated down-flow hanging sponge (DHS) reactor to biologically oxidize the D-CH_4; although this technique enabled oxidization of 95% of the D-CH_4, no D-CH_4 was recovered as CH_4 gas. In contrast, Matsuura et al. (2010) employed a two-stage DHS reactor for post-treatment of the effluent from a UASB reactor treating municipal sewage to recover and oxidize D-CH_4. The first-stage reactor could recover D-CH_4 with 77% recovery efficiency. However, CH_4 compositions in the recovered biogas were relatively low because of an inherent limitation of this method—the D-CH_4 is released from a liquid in the DHS reactor by physical gasification based on the gas–liquid equilibrium. Another approach for removing D-CH_4 involved mixing the liquid in an anaerobic wastewater treatment reactor with gas or a paddle. Hartley and Lant (2006) applied intermittent gas mixing by micro-aeration with biogas-containing air, and this resulted in the release of D-CH_4 into the gas phase of the reactor. However, the CH_4 composition in the recovered biogas was lower than that in the reactor headspace in the absence of aeration, because the gas used for aeration diluted the biogas. Recovered biogas with low CH_4 concentration is unsuitable for subsequent energy generation processes. Pauss et al. (1990) mixed the liquid phase in reactors with a paddle or by recirculating the supernatant liquid to enhance the evolution of D-CH_4 and gas bubbles attached to solid particles from the liquid phase to gas phase. However, in this method, the D-CH_4 concentration would theoretically never be lower than the saturation concentration of D-CH_4. Another possible process is to apply vacuum directly to the reactor. It does not seem to be realistic, because the reactor must be closed completely and a pressure tight case.

In order to check the mass balance of CH_4, the CH_4 evolution rate in the UASB headspace, CH_4 collection rate from the DM, and CH_4 discharge rate from the DM reactor were calculated using the CH_4 concentrations and flow rates of wastewater and biogas described above. Figure 4 shows the rates of CH_4 discharged from the UASB reactor and total CH_4 recovery efficiency in addition to the 3 above-mentioned CH_4 production rates and their averaged values were summarized in Table 1. The average CH_4 evolution rates were 1000 ± 94 mg COD L^{-1} day^{-1} and 1120 ± 110 mg COD L^{-1} day^{-1} during the degasification period (phases 1.2 and 1.3, respectively). These rates were comparable to those during normal operation (960 ± 220 mg COD L^{-1} day^{-1} in phase 1.1 and 1010 ± 230 mg COD L^{-1} day^{-1} in phase 1.4). Thus, degasification did not significantly affect the CH_4 evolution rate in the UASB reactor. In contrast, the average

CH_4 discharge rates were lower during the degasification period (49 ± 10 mg COD L^{-1} day^{-1} in phase 1.2 and 35 ± 11 mg COD L^{-1} day^{-1} in phase 1.3) than during normal operation (140 ± 14 mg COD L^{-1} day^{-1} in phase 1.1 and 150 ± 3 mg COD L^{-1} day^{-1} in phase 1.4) because of the collection of D-CH_4 with the DM. The D-CH_4 discharge rate accounted for 11 ± 3% of the total CH_4 production rate during the normal operation (Phase 1.1), whereas it was 2.6 ± 0.8% during the degasification period (phase 1.3). The average CH_4 collection rates were 180 ± 85 mg COD L^{-1} day^{-1} in phase 1.2 and 210 ± 26 mg COD L^{-1} day^{-1} in phase 1.3 (Figure 4). On the basis of these results, the total CH_4 production rates were calculated to be 1090 ± 230 mg COD L^{-1} day^{-1} in phase 1.1 and 1160 ± 230 mg COD L^{-1} day^{-1} in phase 1.4. In contrast, these rates were slightly higher (1220 ± 140 mg COD L^{-1} day^{-1} in phase 1.2 and 1370 ± 130 mg COD L^{-1} day^{-1} in phase 1.3) during the degasification period, probably because of CH_4 production in the DM reactor. Accordingly, D-CH_4 collection efficiencies were 68 ± 7% and 77 ± 7% in phases 1.2 and 1.3, respectively, and total CH_4 recovery efficiencies increased up to 96 ± 1% and 97 ± 1% during the degasification period (phases 1.2 and 1.3, respectively), as compared to those during normal operation (89 ± 3% in phase 1.1 and 90 ± 2% in phase 1.4).

3.3. Effect of temperature

The UASB and DM reactors were operated at low temperatures (25°C and 15°C in phases 2 and 3, respectively). The average D-CH_4 concentrations in the UASB reactor were 63 ± 4 mg COD L^{-1} at 35°C throughout phase 1, 82 ± 7 mg COD L^{-1} at 25°C throughout phase 2, and 104 ± 5 mg COD L^{-1} at 15°C throughout phase 3 (Figure 2). The increase in D-CH_4 concentrations at lower temperatures was attributed to an increase in the solubility of CH_4 in the liquid phase with decreasing temperature. Corresponding with the increase in D-CH_4 concentration, average D-CH_4 discharge rates increased from 150 ± 12 mg COD L^{-1} day^{-1} at 35°C to 200 ± 20 mg COD L^{-1} day^{-1} at 25°C and 250 ± 13 mg COD L^{-1} day^{-1} at 15°C, thereby indicating that the loss of D-CH_4 from the UASB reactor was more significant at lower temperatures.

Degasification enabled successful collection of D-CH_4 by the DM regardless of temperature. The D-CH_4 concentrations decreased to 13 ± 4 mg COD L^{-1} at 25°C (phase 2.2) and 14 ± 2 mg COD L^{-1} at 15°C (phase 3.2) in the DM reactor. The ratio of D-CH_4 concentration to CH_4 composition in the UASB headspace increased with decreasing temperature. Therefore, the CH_4 collection rates showed a relative increase from 210 ± 26 mg COD L^{-1} day^{-1} at 35°C (phase 1.3) to 300 ± 84 mg COD L^{-1} day^{-1} at 25°C (phase 2.2) and to 370 ± 100 mg COD L^{-1} day^{-1} at 15°C (phase 3.2). Accordingly, total CH_4 recovery efficiency increased from 71% in phase 3.1 to 97% at 15°C by degasification (phase 3.2), and D-CH_4 collection efficiencies increased with a decrease in temperature from 77 ± 7% at 35°C to 85 ± 4% at 25°C and 86 ± 2% at 15°C.

CH_4 evolution rates in the UASB headspace decreased with a decrease in temperature (Figure 4); 1000 ± 170 mg COD L^{-1} day^{-1} throughout phase 1, 860 ± 140 mg COD L^{-1} day^{-1} throughout phase 2, and 650 ± 100 mg COD L^{-1} day^{-1} throughout phase 3. D-COD removal efficiency was not considerably decreased at lower temperatures (94%, 89% and 91% in Phases 1.3, 2.2 and 3.2, respectively). This finding indicated that microbial CH_4 production activity was maintained even at low temperatures probably because of the presence of sufficient amount of biomass and high bioavailability of influent organic materials. Hence, the decrease in the CH_4 evolution rate might be due to an increase in the solubility of CH_4 in the liquid phase (Lettinga et al., 2001).

The CH_4 recovery rates at 25°C (1230 ± 54 mg COD L^{-1} day^{-1} in phase 2.2) and at 15°C (1060 ± 150 mg COD L^{-1} day^{-1} in phase 3.2) were higher than the CH_4 evolution rate at 35°C during normal operation (1000 ± 170 mg COD L^{-1} day^{-1} throughout phase 1). Thus, it can be concluded that the degasification technology enables us to recover CH_4 in a UASB process at low temperature without heating, and the amount of CH_4 produced in the UASB process is comparable to that produced in a conventional process under mesophilic conditions. In general, many types of wastewaters are discharged at low ambient temperatures under temperate climatic conditions. To treat such wastewaters under mesophilic condition, their temperature must be increased to the optimal mesophilic range. This requires a significant amount of energy, and the high energy cost is a heavy economic burden on such wastewater treatment systems (Lettinga et al., 2001). A UASB process that does not require heating is economically attractive (Dhaked et al., 2010; Angenent et al., 2001). However, the degasification technology also consumed much energy in this study (42 J s^{-1}). The maximal CH_4 collection rate of 660 mg COD L^{-1} day^{-1} (Fig. 4) was comparable only to 0.14 J s^{-1}, assuming that 1 L of CH_4 has the energy of 35800 J.

Future studies must aim to reduce the energy required for degasification so that the degasification technology may be applied in real wastewater treatment processes.

Although the D-COD removal efficiency was slightly decreased at lower temperatures (phases 2 and 3), it was still higher than 87%, and the effluent D-COD concentration was less than 200 mg COD L^{-1} (Figure 1). In contrast, the P-COD concentration in the DM reactor, which was calculated as the difference between the effluent T-COD and D-COD concentrations, increased at lower temperatures, indicating a decrease in P-COD removal efficiency. At lower temperatures, particles might settle more slowly because of lower liquid–solid separation efficiency, which in turn may be attributed to higher liquid viscosity and/or attachment of gas bubbles onto the particles. Interestingly, the P-COD removal efficiency in the DM reactor was improved by degasification in phase 3. This finding may be explained by the fact that a decrease in the dissolved gas concentration in the liquid in the DM reactor caused by degasification, followed by redissolution of the gas bubbles attached to the particles, caused the particles to efficiently settle down. The degasification technology might improve the efficiency of particle separation in a clarifier for a UASB reactor.

To verify the reproducibility of temperature effects, the temperature in the UASB reactor was set at 35°C again at day 123. The average D-COD removal efficiency (92 ± 1%) and the D-CH_4 concentration (66 ± 5 mg COD L^{-1}) in phase 4 were comparable to those in phase 1 (94 ± 2% and 63 ± 4 mg COD L^{-1}, respectively). This finding indicated that changes in chemical parameters (e.g., the D-CH_4 concentration and the CH_4 evolution rate in the UASB reactor) in phases 2 and 3 were attributed to lower temperatures.

3.4. Effect of HRT

The UASB reactor was again operated at 35°C but at lower COD concentrations (the concentrations throughout phase 5 and phase 6 were two-third and one-third, respectively, of that in phase 4) (Figure 1 and Table 1). In order to keep the COD loading rate of the UASB reactor constant, HRTs in phase 5 (6.7 h) and phase 6 (3.3 h) were also reduced to two-third and one-third, respectively, of the HRT in phase 4 (10 h) by increasing the wastewater feed rate. The average D-CH_4 concentration in the UASB reactor was ca. 70 mg COD L^{-1} in phases 4 to 7 because of the identical solubility of CH_4 at a constant temperature (35°C). The average D-CH_4 concentration was ca. 90% of the theoretical saturation concentration of D-CH_4 at 35°C (Hartley and Lant, 2006). However, an increase in the wastewater feed rate resulted in an increase in the CH_4 discharge rate from the UASB reactor (Figure 4). The D-CH_4 discharge rates were 160 ± 12 mg COD L^{-1} day^{-1} in phase 4, 240 ± 15 mg COD L^{-1} day^{-1} throughout phase 5, and 500 ± 35 mg COD L^{-1} day^{-1} throughout phase 6. Degasification could reduce the D-CH_4 concentration to 12 ± 1 mg COD L^{-1} in the DM reactor at an HRT of 6.7 h (phase 5.2). The average CH_4 collection rate was 480 ± 130 mg COD L^{-1} day^{-1} in phase 5.2, and this was much higher than that in phase 1.3 (210 ± 26 mg COD L^{-1} day^{-1}) because of the higher D-CH_4 concentration. The corresponding total CH_4 recovery efficiency was 98% and the CH_4 recovery rate was 1830 ± 160 mg COD L^{-1}

day^{-1}, which was 1.5 times higher than that in normal operation at the same HRT in phase 5.1 (1220 ± 100 mg COD L^{-1} day^{-1}). These results indicate that the UASB process with degasification is a promising technology for effective CH_4 recovery in low-strength wastewater treatment. Because of the economical benefits offered by the UASB process, its application to low-strength wastewater treatment has recently attracted much attention (Elmitwalli et al., 2003; Angenent et al., 2001; Gomec et al., 2008; Matsushige et al., 1990). At an HRT of 3.3 h, D-CH_4 remained at a high concentration (51 ± 3 mg COD L^{-1}), and hence, the total CH_4 recovery efficiency was as low as 76 ± 2% during the degasification period (phase 6.2). This was probably because of insufficient HRT to collect D-CH_4; however, the average CH_4 collection rate was still high (370 ± 10 mg COD L^{-1} day^{-1}). Future studies should aim to improve the gas flux rate of the DM, for example, by changing membrane materials (Liang et al., 2002). The average biogas collection rates calculated by excluding the amount of air from the collected biogas (i.e., biogas flux through the DM) were 150 ± 14 mL m^{-2} day^{-1} in phase 1.3 and 150 ± 24 mL m^{-2} day^{-1} in phase 6.2 (Figure 3B), indicating that the decrease in biogas flux due to membrane fouling was negligible in this study.

3. Conclusions

In this study, the bench scale UASB reactor equipped with the DM reactor was operated at different temperatures and HRTs for 170 days. D-CH_4 was successfully collected by degasification with the DM. Under lower temperatures or shorter HRTs, the D-CH_4 concentrations increased; therefore, the D-CH_4 collection efficiencies increased. Moreover, the P-COD concentration was decreased by degasification. These results indicated that degasification is a promising technology for improving CH_4 recovery and P-COD removal efficiencies of the UASB process for treating low-strength wastewater at low temperature.

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References

Angenent, L.T., Banik, G.C., Sung, S.W. (2001) Anaerobic migrating blanket reactor treatment of low-strength wastewater at low temperatures. Water Environment Research 73(5), 567-574.

Bandara, W.M.K.R.T.W., Ikeda, M., Satoh, H., Sasakawa, M., Nakahara, Y., Takahashi, M., Okabe, S. (Submitted) Recovery of dissolved methane gas discharged from a UASB reactor with a degassing membrane. Water Science and Technology.

Cakir, F.Y., Stenstrom, M.K. (2005) Greenhouse gas production: A comparison between aerobic and anaerobic wastewater treatment technology. Water Research 39(17), 4197-4203.

Dhaked, R.K., Singh, P., Singh, L. (2010) Biomethanation under psychrophilic conditions. Waste Management 30(12), 2490-2496.

Elmitwalli, T.A., van Lier, J., Zeeman, G., Lettinga, G. (2003) Treatment of domestic sewage at low temperature in a two-anaerobic step system followed by a trickling filter. Water Science and Technology 48(11-12), 199-206.

Fang, H.H.P., Liang, D.W., Zhang, T., Liu, Y. (2006) Anaerobic treatment of phenol in wastewater under thermophilic condition. Water Research 40(3), 427-434.

Gomec, C.Y., Letsiou, I., Ozturk, I., Eroglu, Y., Wilderer, P.A. (2008) Identification of archaeal population in the granular sludge of an UASB reactor treating sewage at low temperatures. Journal of Environmental Science and Health, Part A 43(13), 1504-1510.

Hartley, K., Lant, P. (2006) Eliminating non-renewable CO_2 emissions from sewage treatment: An anaerobic migrating bed reactor pilot plant study. Biotechnology and Bioengineering 95(3), 384-398.

Hatamoto, M., Yamamoto, H., Kindaichi, T., Ozaki, N., Ohashi, A. (2010) Biological oxidation of dissolved methane in effluents from anaerobic reactors using a down-flow hanging sponge reactor. Water Research 44(5), 1409-1418.

Kashyap, D.R., Dadhich, K.S., Sharma, S.K. (2003) Biomethanation under psychrophilic conditions: a review. Bioresource Technology 87(2), 147-153.

Lettinga, G., Rebac, S., Zeeman, G. (2001) Challenge of psychrophilic anaerobic wastewater treatment. Trends in Biotechnology 19(9), 363-370.

Lettinga, G., Rebac, S., Parshina, S., Nozhevnikova, A.N., van Lier, J.B., Stams, A.J. (1999) High-rate anaerobic treatment of wastewater at low temperatures. Applied and Environmental Microbiology 65(4), 1696-1702.

Liang, T.M., Cheng, S.S., Wu, K.L. (2002) Behavioral study on hydrogen fermentation reactor installed with silicone rubber membrane. International Journal of Hydrogen Energy 27(11-12), 1157-1165.

Luostarinen, S.A., Rintala, J.A. (2005) Anaerobic on-site treatment of black water and dairy parlour wastewater in UASB-septic tanks at low temperatures. Water Research 39(2-3), 436-448.

Madden, P., Chinalia, F.A., Enright, A.M., Collins, G., O'Flaherty, V. (2010)

Perturbation-independent community development in low-temperature anaerobic biological wastewater treatment bioreactors. Biotechnology and Bioengineering 105(1), 79-87.

Matsushige, K., Inamori, Y., Mizuochi, M., Hosomi, M., Sudo, R. (1990) The effects of temperature on anaerobic filter treatment for low-strength organic wastewater. Environmental Technology 11(10), 899-910.

Matsuura, N., Hatamoto, M., Sumino, H., Syutsubo, K., Yamaguchi, T., Ohashi, A. (2010) Closed DHS system to prevent dissolved methane emissions as greenhouse gas in anaerobic wastewater treatment by its recovery and biological oxidation. Water Science and Technology 61(9), 2407-2415.

McKeown, R.M., Scully, C., Mahony, T., Collins, G., O'Flaherty, V. (2009) Long-term (1243 days), low-temperature (4-15°C), anaerobic biotreatment of acidified wastewaters: Bioprocess performance and physiological characteristics. Water Research 43(6), 1611-1620.

Pauss, A., Andre, G., Perrier, M., Guiot, S.R. (1990) Liquid-to-gas mass transfer in anaerobic processes: Inevitable transfer limitations of methane and hydrogen in the biomethanation process. Applied and Environmental Microbiology 56(6), 1636-1644.

Satoh, H., Miura, Y., Tsushima, I., Okabe, S. (2007) Layered structure of bacterial and archaeal communities and their in situ activities in anaerobic granules. Applied and Environmental Microbiology 73(22), 7300-7307.

Trzcinski, A.P., Stuckey, D.C. (2010) Treatment of municipal solid waste leachate using a submerged anaerobic membrane bioreactor at mesophilic and psychrophilic temperatures: Analysis of recalcitrants in the permeate using GC-MS. Water Research 44(3), 671-680.

Voolapalli, R.K., Stuckey, D.C. (1998) Stability enhancement of anaerobic digestion through membrane gas extraction under organic shock loads. Journal of Chemical Technology and Biotechnology 73(2), 153-161.

Xing, W., Zuo, J.E., Dai, N., Cheng, J., Li, J. (2009) Reactor performance and microbial community of an EGSB reactor operated at 20 and 15°C. Journal of Applied Microbiology 107(3), 848-857.

Figure Captions

Figure 1. Concentrations of influent total chemical oxygen demand (T-COD) and effluent T-COD and dissolved fraction of COD (D-COD) in the upflow anaerobic sludge blanket (UASB) reactor, and D-COD removal efficiency of the UASB reactor. The gray area represents a degasification period. Operational conditions in each phase are summarized in Table 1.

Figure 2. Dissolved methane (D-CH_4) concentrations in the upflow anaerobic sludge blanket (UASB) and degassing membrane (DM) reactors, and D-CH_4 collection efficiency of the DM reactor. The gray area represents a degasification period. Operational conditions in each phase are summarized in Table 1.

Figure 3. Gas compositions in the headspace of the upflow anaerobic sludge blanket (UASB) reactor (A) and inside the lumen of the hollow fibers of the degassing membrane (DM), and gas flux into the hollow fibers of the DM (B). The gray area represents a degasification period. Operational conditions in each phase are summarized in Table 1.

Figure 4. Rates (mg COD L^{-1} day^{-1}) of CH_4 evolution in the upflow anaerobic sludge blanket (UASB) headspace, CH_4 collection from the degassing membrane (DM), and CH_4 discharge from the UASB and DM reactors, as well as total CH_4 recovery efficiency. The gray area represents a degasification period. Operational conditions in

each phase are summarized in Table 1.

TABLE 1. Summary of operational conditions and reactor performances in the UASB and DM reactors

Phase	Influent T-COD concentration (mg/L)	T-COD loading rate (mg/L/h)	HRT of UASB (hour)	HRT of DM (hour)	Temperature (°C)	Transmembrane pressure (kPa)
1.1	1650 ± 132	150 ± 10	10.9 ± 0.3	9.2 ± 0.2	35	0
1.2	1580 ± 95	150 ± 4	10.3 ± 0.4	8.7 ± 0.3	35	50
1.3	1620 ± 48	160 ± 10	10.0 ± 0.3	8.5 ± 0.3	35	80
1.4	1640	160	10.4	8.8	35	0
2.1	1530 ± 21	150 ± 7	10.2 ± 0.4	8.6 ± 0.4	25	0
2.2	1410 ± 13	140 ± 5	10.1 ± 0.3	8.5 ± 0.3	25	80
2.3	1390	140	10.2	8.6	25	0
3.1	1390 ± 0	130 ± 7	10.3 ± 0.6	8.7 ± 0.5	15	0
3.2	1340 ± 18	130 ± 4	10.1 ± 0.2	8.5 ± 0.2	15	80
3.3	1400	140	10.0	8.5	15	0
4	1460 ± 37	150 ± 4	10.0 ± 0.1	8.4 ± 0	35	0
5.1	810 ± 30	110 ± 23	5.5 ± 1.0	4.7 ± 0.8	35	0
5.2	880 ± 55	89 ± 5	6.6 ± 0	5.6 ± 0	35	80
5.3	950 ± 4	96 ± 1	6.6 ± 0	5.6 ± 0	35	0
6.1	450 ± 48	45 ± 5	3.3 ± 0	2.8 ± 0	35	0
6.2	480 ± 5	48 ± 1	3.3 ± 0	2.8 ± 0	35	80
6.3	500	50	3.3	2.8	35	0
7	1510 ± 46	150 ± 5	9.9 ± 0.1	8.4 ± 0	35	0

TABLE 1–Continued

Phase	CH_4 evolution rate (mg-COD/L/day)	CH_4 collection rate (mg-COD/L/day)	CH_4 discharge rate from UASB (mg-COD/L/day)	CH_4 discharge rate from DM (mg-COD/L/day)	Total CH_4 production rate (mg-COD/L/day)	CH_4 recovery rate (mg-COD/L/day)	D-CH_4 collection efficiency (-)	Total CH_4 recovery efficiency (-)
1.1	960 ± 220		140 ± 14		1090 ± 230	960 ± 220		89 ± 3
1.2	1000 ± 94	180 ± 85	150 ± 7	49 ± 10	1220 ± 140	1180 ± 150	68 ± 7	96 ± 1
1.3	1120 ± 110	210 ± 26	150 ± 4	35 ± 11	1370 ± 130	1340 ± 130	77 ± 7	97 ± 1
1.4	1010 ± 230		150 ± 3		1160 230	1010 ± 230		90 ± 2
2.1	880 ± 91		190 ± 16		1080 ± 85	880 ± 91		85 ± 2
2.2	930 ± 69	300 ± 84	210 ± 27	32 ± 11	1260 ± 59	1230 ± 54	85 ± 4	97 ± 1
2.3	580 ± 93		190 ± 3		770 ± 97	580 ± 93		78 ± 1
3.1	530 ± 25		240 ± 19		780 ± 37	530 ± 25		71 ± 2
3.2	700 ± 73	370 ± 100	240 ± 7	33 ± 4	1100 ± 150	1060 ± 150	86 ± 2	97 ± 1
3.3	720		250 ± 20		610 ± 490	720		76
4	1390 ± 160		160 ± 12		1550 ± 170	1390 ± 160		90 ± 1
5.1	1220 ± 100		240 ± 14		1460 ± 110	1220 ± 100		84 ± 1
5.2	1350 ± 47	480 ± 130	250 ± 19	45 ± 4	1870 ± 160	1830 ± 160	82 ± 1	98 ± 0
5.3	1150 ± 190		240 ± 14		1390 ± 190	1150 ± 190		84 ± 2
6.1	830 ± 63		480 ± 35		1320 ± 95	830 ± 63		64 ± 1
6.2	770 ± 33	370 ± 12	500 ± 41	370 ± 25	1500 ± 16	1140 ± 31	26 ± 7	76 ± 2
6.3	960 ± 280		510 ± 19		1480 ± 300	960 ± 280		69 ± 5
7	810 ± 540		150 ± 125		1220 ± 600	1300 ± 76		90 ± 1

TABLE 2. Characteristics of the degassing membrane module

Item	Characteristics
Non-porous layer for degassing	Polyethylene
Porous layer	Polyurethane
Outer diameter of a hollow fiber	280 μm
Inner diameter of a hollow fiber	200 μm
Thickness	40 μm
Length	360 mm
Number of hollow fiber	5500
Total membrane area	1.7 m^2
Membrane volume	0.4 L

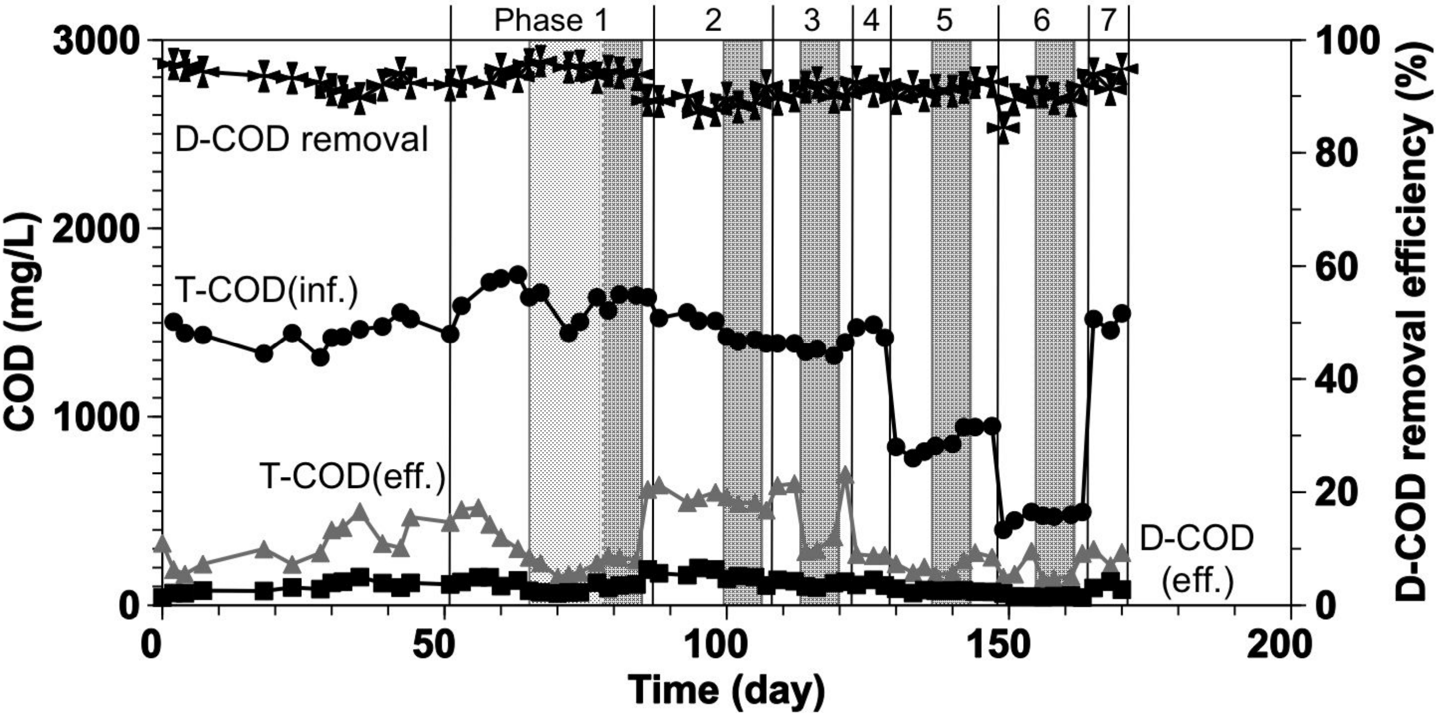

Figure 1
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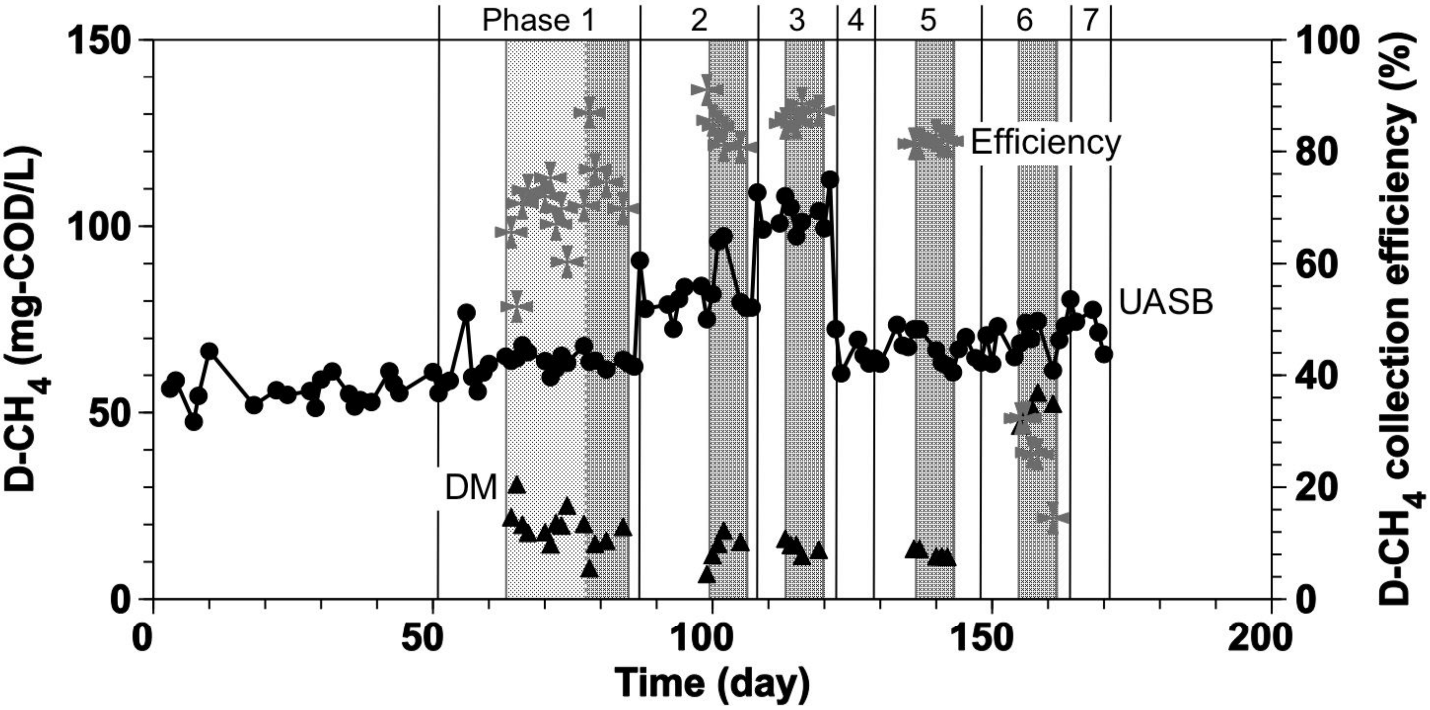

Figure 2

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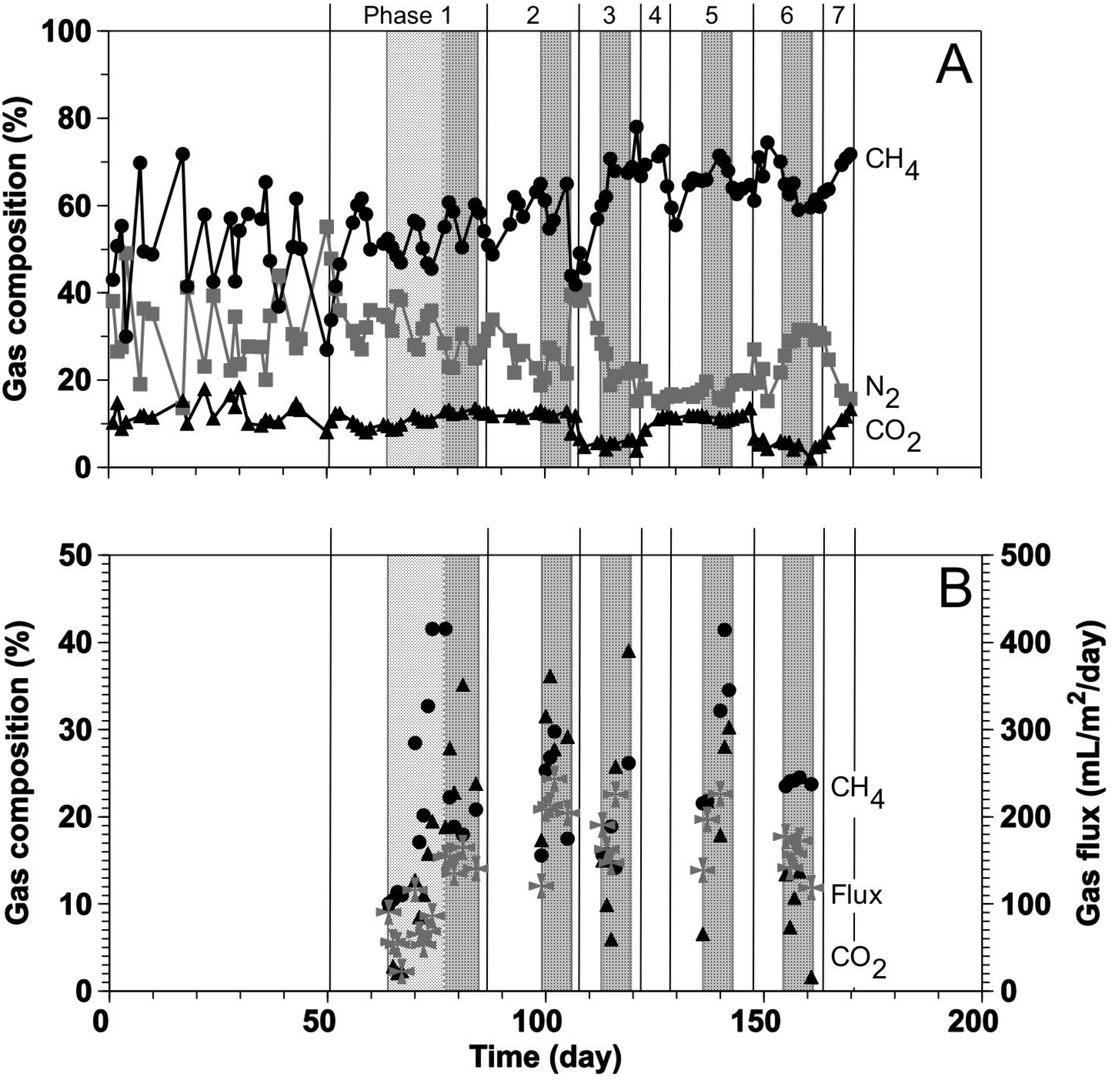

Figure 3
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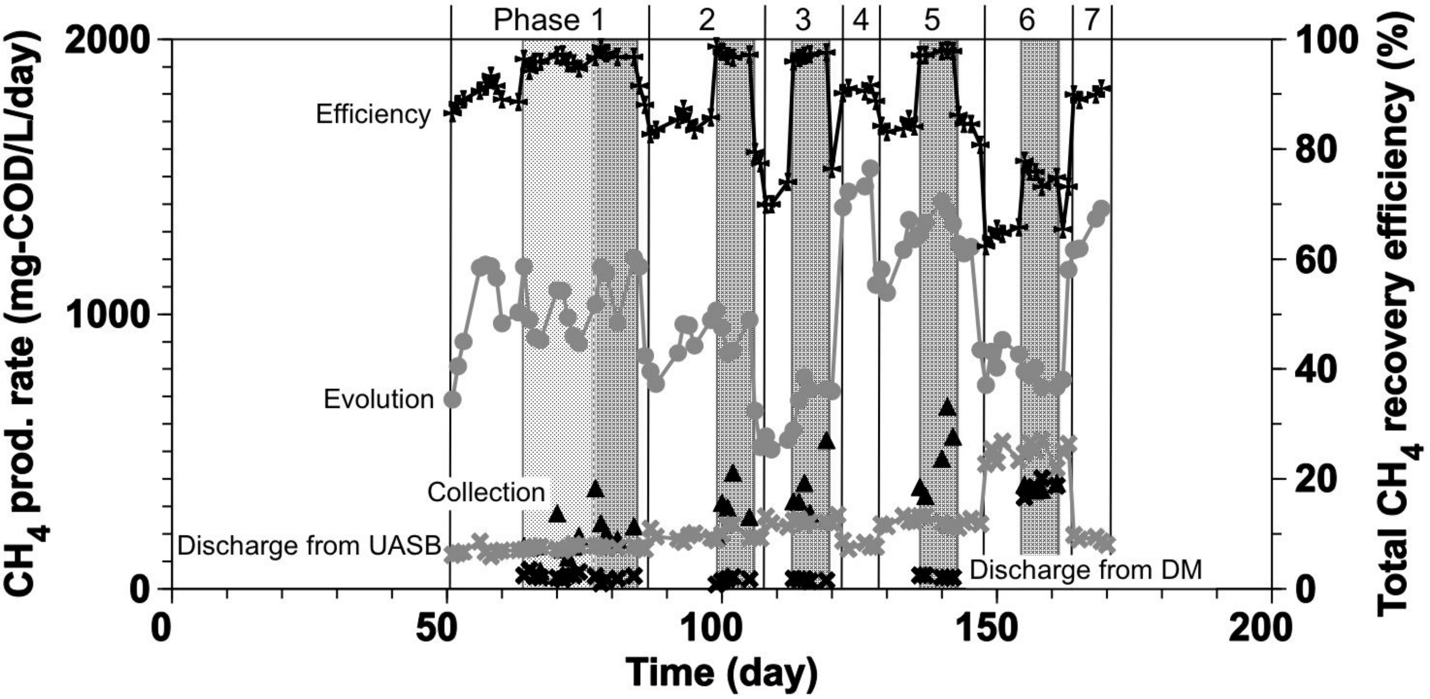

Figure 4
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