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3 **Enhancement of organic matter degradation and methane gas**  
4 **production of anaerobic granular sludge by degasification of**  
5 **dissolved hydrogen gas**

6

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29

30 **Abstract**

31 A hollow fiber degassing membrane (DM) was applied to enhance organic matter  
32 degradation and methane gas production of anaerobic granular sludge process by reducing the  
33 dissolved hydrogen gas (D-H<sub>2</sub>) concentration in the liquid phase. DM was installed in the  
34 bench-scale anaerobic granular sludge reactors and D-H<sub>2</sub> was removed through DM using a  
35 vacuum pump. Degasification improved the organic matter degradation efficiency to 79% while  
36 the efficiency was 62% without degasification at 12,000 mg L<sup>-1</sup> of the influent T-COD  
37 concentration. Measurement of D-H<sub>2</sub> concentrations in the liquid phase confirmed that D-H<sub>2</sub> was  
38 removed by degasification. Furthermore, the effect of acetate concentrations on the organic matter  
39 degradation efficiency was investigated. At acetate concentrations above 3 g L<sup>-1</sup>, organic matter  
40 degradation deteriorated. Degasification enhanced the propionate and acetate degradation. These  
41 results suggest that degasification reduced D-H<sub>2</sub> concentration and volatile fatty acids  
42 concentrations, prevented pH drop, and subsequent enhanced organic matter degradation.

43

44 **Keywords**

45 Degassing membrane; Dissolved hydrogen gas; Process enhancement; pH drop; Shock  
46 load

47

48 **1. Introduction**

49 Anaerobic digestion (AD) has been widely used for the treatment of organic wastes and

50 wastewater due to the low operational cost and the formation of biogas as an energy resource. An  
51 AD process involves four key stages, hydrolysis, acidogenesis, acetogenesis and methanogenesis,  
52 each of which is accomplished by a different group of microorganisms. Many types of  
53 microorganisms are sensitive to and inhibited by operational parameters such as pH, temperature,  
54 dissolved hydrogen gas ( $D-H_2$ ) and volatile fatty acids (VFAs) (Feng et al., 2014).

55           Among these,  $D-H_2$  is one of the most critical parameters affecting the AD process.  $D-H_2$   
56 is an intermediate metabolite in many reactions in acidogenesis and acetogenesis (Giovannini et  
57 al., 2016). The  $H_2$  production, consumption and accumulation in the AD process are subject to the  
58 thermodynamics of the reactions. As a result, during oxidation of propionate and n-butyrate,  
59 which are two main VFAs in acidogenesis, the standard Gibbs free energy changes in the  
60 reactions are positive (76.1 kJ/mol for propionate degradation and 48.1 kJ/mol for n-butyrate  
61 degradation) (Giovannini et al., 2016). Hence, the reactions do not proceed spontaneously in the  
62 direction of oxidation under standard conditions in which the concentration of reactants and  
63 products in aqueous solution are 1M, the temperature is 30°C, the pH is 7 and the pressure is 1  
64 atm. For example, propionate and n-butyrate can only be decomposed into acetate when the  
65 hydrogen partial pressure ( $p_{H_2}$ ) is very low (e.g., 0.002 and 0.0008 atm for propionate and  
66 butyrate oxidation, respectively) (Giovannini et al., 2016). However, in practice, the  $p_{H_2}$  usually  
67 exceeds these levels because of stress conditions such as high organic loading rates, short  
68 hydrogen retention times (HRTs) or inflow of inhibitors (Ketheesan and Stuckey, 2015). Increases  
69 in  $p_{H_2}$  cause the accumulation of intermediate products (mainly VFAs), and may lead to process

70 failure, such as low organic matter removal, reduced biogas production, and poor effluent quality.  
71 The process failure has limited the widespread application of the AD process when compared  
72 with the aerobic process.

73 Several studies have focused on accelerating the AD by reducing the D-H<sub>2</sub> concentration.  
74 For example, Qiao et al. (2016) supplied sulfate as an external electron acceptor to enhance the  
75 degradation of propionate in a thermophilic anaerobic membrane reactor. Addition of sulfate  
76 enhanced propionate degradation by sulfate reduction and H<sub>2</sub> consumption by hydrogenotrophic  
77 methanogenesis. Furthermore, Qiao et al. (2013) attained stable operation of a thermophilic  
78 submerged anaerobic membrane reactor for more than 263 days, even under the conditions of  
79 high organic loading rates, persistent propionate and detectable H<sub>2</sub> in biogas by co-digestion of  
80 coffee grounds and dewatered activated sludge and addition of an alkali chemical (NH<sub>4</sub>HCO<sub>3</sub>). Li  
81 et al. accelerated H<sub>2</sub> consumption and VFAs decomposition by the addition of azo dye, which  
82 serves as an electron acceptor of H<sub>2</sub> oxidation (Li et al., 2014). Zero valent iron as a reducing  
83 material has commonly been used to accelerate AD processes (e.g., the decomposition of protein,  
84 cellulose, butyrate and propionate). Although zero valent iron chemically produces H<sub>2</sub> ( $\text{Fe}^0 +$   
85  $2\text{H}^+ \rightarrow \text{Fe}^{2+} + \text{H}_2$ ), D-H<sub>2</sub> concentrations became rather low in response to the addition of zero  
86 valent iron when compared with processes without zero valent iron because of the abundance of  
87 homoacetogens and hydrogenotrophic methanogens, resulting in H<sub>2</sub> consumption and therefore  
88 increased Chemical Oxygen Demand (COD) removal and methane (CH<sub>4</sub>) production (Feng et al.,  
89 2014; Kong et al., 2016; Meng et al., 2013). Bioaugmentation was also effective to reduce the

90 D-H<sub>2</sub> concentration (Romero-Güiza et al., 2016). However, all of the studies described above  
91 required additives, leading to deterioration of effluent water quality and increased costs. Therefore,  
92 simpler technology allowing removal of D-H<sub>2</sub> is required.

93           Degasification is a technology to remove dissolved gases from liquids. A degassing  
94 membrane (DM), which is a solid, microporous, hydrophobic (or hydrophilic) polymeric matrix,  
95 is one option for gas-liquid separation (Mandowara and Bhattacharya, 2009). A DM allows the  
96 gas phase to be in direct contact with the liquid phase. Vacuum is applied in the gas phase and  
97 creates a chemical concentration gradient of gases between fluids inside and outside the  
98 membrane, which facilitates transfer of the molecules through the membrane wall. A DM allows  
99 the gases dissolved in the liquid, but not the liquid itself, to pass through, which has the advantage  
100 of preventing the gas from being re-dissolved. DMs have been widely used for removal of O<sub>2</sub> and  
101 CO<sub>2</sub> from water as alternatives to vacuum towers, forced draft deaerators, and oxygen scavengers  
102 in many industries, including the microelectronics, pharmaceutical, power, food and beverage,  
103 industrial, photographic, ink and analytical markets (Drioli et al., 2015). We applied a DM to an  
104 anaerobic wastewater treatment process with granular sludge for removal of dissolved CH<sub>4</sub> gas  
105 (Bandara et al., 2013, 2012, 2011). Among many AD processes, the anaerobic granular sludge  
106 process has attracted attention because it has several advantages over dispersed sludge processes,  
107 such as higher biomass retention (especially slowly growing microorganisms), higher volumetric  
108 reaction rate, richer microbial diversity, higher tolerance to shock and toxic loadings, excellent  
109 settling properties, and reduced reactor volume (Liu et al., 2015; van Lier et al., 2015). A

110 hollow-fiber DM module was connected to the liquid outlet of a bench-scale upflow anaerobic  
111 sludge blanket (UASB) reactor treating synthetic wastewater (Bandara et al., 2011), which  
112 resulted in the dissolved CH<sub>4</sub> gas concentration decreasing from 104 mg COD L<sup>-1</sup> to 14 mg COD  
113 L<sup>-1</sup> at 15°C by degasification. In addition, degasification accelerated particulate matter  
114 sedimentation, probably because of removal of gas bubbles attached to the particulate matter.  
115 Furthermore, the DM module was applied to remove dissolved CH<sub>4</sub> gas from a bench-scale  
116 UASB reactor treating domestic wastewater (Bandara et al., 2012). The DM module could be  
117 operated with an average biogas flux through the DM of 55 ± 10 mL-biogas m<sup>-2</sup> day<sup>-1</sup> for 18  
118 months without significant membrane fouling.

119 In this study, D-H<sub>2</sub> in the liquid phase of an anaerobic granular sludge was removed by a  
120 hollow fiber DM to enhance the organic matter degradation and CH<sub>4</sub> production. The COD  
121 loading rate was gradually increased by increasing organic matter concentration or reducing HRT  
122 to deliberately deteriorate COD removal efficiency and CH<sub>4</sub> production, after which  
123 degasification was applied. In addition, the effect of acetate concentrations on COD removal  
124 efficiency and CH<sub>4</sub> production were investigated. Finally, the mechanisms of recovery from the  
125 process failure were discussed based on the experimental data (D-H<sub>2</sub> concentration, pH level and  
126 removed H<sub>2</sub> amount).

127

## 128 **2. Materials and methods**

### 129 2.1 Experimental setup and operational conditions

130 Two identical bench-scale reactors (height, 40 cm; diameter, 7 cm; working volume, 1.3  
131 L) were used for granular sludge incubation (Bandara et al., 2011). A bundle of hollow fiber  
132 membranes for degasification was installed in both of the reactors. The physical characteristics of  
133 the membrane have been described in detail elsewhere (Bandara et al., 2011). Degasification was  
134 applied to one reactor (denoted degasification reactor (DR)), while another reactor was operated  
135 without degasification (denoted control reactor (CR)). The reactors were inoculated with 0.3 L of  
136 anaerobic granular sludge obtained from a full-scale UASB reactor treating wastewater from an  
137 isomerized sugar-processing plant (Bandara et al., 2011). The reactors were fed with synthetic  
138 wastewater containing powdered skim milk as a carbon and energy source, inorganic salts, and  
139 trace metals (Bandara et al., 2011) and operated at 37°C. The COD concentration and loading  
140 were controlled by changing the concentration of powdered milk or hydraulic retention time  
141 (HRT). Dissolved gas in the liquid was collected by the DM. The reactors were completely filled  
142 with the wastewater. The liquid in the reactors was not mixed. The characteristics of the DM  
143 module were described in detail elsewhere (Bandara et al., 2011). The dissolved biogas diffuses  
144 into the lumen of the hollow fibers of the DM under vacuum generated using a vacuum pump  
145 (Model APN-110KV-1; Iwaki Co., Ltd., Tokyo, Japan). The operational conditions are  
146 summarized in Table 1. Granular sludge was acclimated until day 33. From day 34 to 132 (Phase  
147 1), the total COD (T-COD) loading was increased by increasing influent T-COD concentration.  
148 Degasification was applied only to DR at higher T-COD loading rates (Phases 1.5, 1.7, and 1.9).  
149 After these shock load conditions, T-COD loading rate was switched back to the previous T-COD

150 loading to investigate reversibility of reactor performance. In Phase 2 (from day 133 to day 210),  
151 the T-COD loading was increased by reducing HRT while keeping the COD concentration  
152 constant. In Phase 3 (from day 211 to day 246), acetate salt was added to the influent synthetic  
153 wastewater to investigate the direct effect of acetate on T-COD removal.

154

## 155 2.2 Analytical methods

156 Total COD concentrations (T-COD) were measured using the Hach Method 8000.

157 T-COD removal efficiency was calculated as the ratio of T-COD removed to T-COD in the

158 influent. The concentrations of VFAs (the sum of formate, acetate, propionate, lactate, i-butyrate,

159 and n-butyrate) were determined using a high-performance liquid chromatography system

160 (LC-10AD; Shimadzu Co., Kyoto, Japan) equipped with a Shimadzu Shim-pack SCR-102H

161 column ( $0.8 \times 30$  cm) after filtering through a 0.2-mm membrane. The pH was determined using a

162 pH electrode. The concentrations of  $\text{CH}_4$  and  $\text{H}_2$  in the headspace of the reactors and inside the

163 lumen of the hollow fibers were measured using a gas chromatography system (GC-14B;

164 Shimadzu Co.) equipped with a thermal-conductivity detector and a Shincarbon-ST column

165 (Shimadzu Co.). The biogas volumes were measured at  $25^\circ\text{C}$  and the dissolved gas compositions

166 were determined using the headspace method (Bandara et al., 2011).

167

## 168 **3. Results and discussion**

### 169 3.1 Effects of T-COD loading on T-COD removal efficiencies

170 Granular sludge was incubated in two reactors, DR and CR, for 246 days. Figure 1 shows  
171 the T-COD removal efficiencies of DR and CR at day 210. The T-COD removal efficiencies of  
172 DR and CR were 78% at day 0, while they increased to 93% at day 6 and then reached a steady  
173 state with respect to T-COD removal efficiency (Figure 1A). The influent T-COD concentration  
174 gradually increased to about 8200 mg L<sup>-1</sup> until day 86 (Table 1), which resulted in a slight  
175 decrease in T-COD removal efficiencies (>84%). When the influent T-COD concentration was  
176 further increased to approximately 12,000 mg L<sup>-1</sup> at day 87, the T-COD removal efficiencies  
177 decreased significantly; therefore, degasification was applied only to DR. The T-COD removal  
178 efficiency decreased slightly (79%) in the DR, whereas it decreased to 62% in the CR. After  
179 switching back to the previous T-COD loading (about 24 g L<sup>-1</sup>-reactor day<sup>-1</sup> in Phase 1.4) in  
180 Phase 1.6, the CR immediately recovered from the shock effect with the same T-COD removal  
181 efficiency as the previous one in Phase 1.4. When shock loads were applied again in Phases 1.7  
182 and 1.9, degasification increased the T-COD removal efficiencies. The results obtained after  
183 switching back to the previous T-COD loading in Phases 1.6, 1.8 and 1.10 demonstrated that  
184 recovery from the disruption of T-COD removal was reversible.

185 The T-COD loading was also increased by reducing the HRT while keeping the COD  
186 concentration constant at about 8200 mg L<sup>-1</sup> from day 133 to day 195 (Figure 1B). The T-COD  
187 removal efficiencies were higher in the DR than in the CR in all Phases. The decrease in T-COD  
188 removal efficiencies of DR was not significant when the T-COD loading rates were lower than 50  
189 g L<sup>-1</sup>-reactor day<sup>-1</sup> from Phases 2.1 to 2.3. Conversely, degasification no longer significantly

190 improved T-COD removal when the T-COD loading rate was above  $70 \text{ g L}^{-1}\text{-reactor day}^{-1}$  from  
191 Phase 2.5 to Phase 2.9. Recovery from the disruption of T-COD removal by reducing HRT was  
192 also reversible in Phase 2.10.

193 To elucidate the reason for the process stability of DR, the dissolved  $\text{H}_2$  (D- $\text{H}_2$ )  
194 concentration, VFA concentrations and pH in DR and CR were determined. The D- $\text{H}_2$   
195 concentrations were less than  $10 \mu\text{g L}^{-1}$  under normal T-COD loading (approximately  $24 \text{ g}$   
196  $\text{L}^{-1}\text{-reactor day}^{-1}$ ) in Phases 1.4, 1.6, 1.8 and 1.10 (Figure 2A). Additionally, the D- $\text{H}_2$   
197 concentrations in the DR were lower than those in the CR under shock load conditions.  
198 Interestingly, they were even lower in Phases 1.5 and 1.7 than under normal T-COD loading  
199 (Phase 1.4). The measurement of  $\text{H}_2$  gas removal rates by degasification confirmed removal of  
200 D- $\text{H}_2$  through the DM.  $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{H}_2$  concentrations in the DM were between 46% and 16%,  
201 83% and 54%, and 0.62% and 0.1%, respectively. Acetate and propionate concentrations were  
202 lower in the DR during degasification periods than in the CR at high T-COD loading rates in  
203 Phases 1.5, 1.7 and 1.9 (Figure 2B). Degradation of propionate is a central issue for improving the  
204 performance of an AD process because propionate is an intermediate product that generally  
205 accumulates in anaerobic digesters. Propionate can only be oxidized if a syntrophic association is  
206 conducted by propionate-oxidizing bacteria and hydrogen-consuming bacteria. As propionate  
207 degraders, syntrophic propionate-oxidizing bacteria play an important role in the anaerobic food  
208 chain (Qiao et al., 2016). However, propionate-degradation is often inhibited by propionate  
209 (substrate inhibition). Degasification can be an option to prevent process failure by propionate

210 accumulation. The butyrate concentrations were  $<250 \text{ mg-COD L}^{-1}$  and the other VFAs (lactate,  
211 formate and isobutyrate) concentrations were  $<40 \text{ mg-COD L}^{-1}$ , although Voolapalli and Stuckey  
212 (2001) reported formate accumulation under shock load conditions. Higher VFAs concentrations  
213 could cause a decrease in pH in the CR, but not in the DR. These results could be attributed to the  
214 D-H<sub>2</sub> gas being removed by degasification followed by stimulation of syntrophic oxidation of  
215 VFAs to prevent the pH drop, resulting in improved T-COD removal.

216 Relationships between T-COD loading rates and T-COD removal efficiencies (Figure  
217 3A), D-H<sub>2</sub> concentrations and pH (Figure 3B), acetate (Figure 3C) or propionate concentrations  
218 (Figure 3D) for 246 days were investigated. Enhancement of T-COD removal efficiency by  
219 degasification was most significant at about  $25 \text{ g L}^{-1}\text{-reactor day}^{-1}$  of T-COD loading rate (Figure  
220 3A). Effect of degasification on enhancement of T-COD removal efficiency became lower with  
221 increase in T-COD loading rate. Figure 3B demonstrated that degasification reduced D-H<sub>2</sub>  
222 concentrations to about one half of those in CR and prevented acidification of the liquid in DR by  
223 increase pH by 0.2. Based on the result, it is suggested that D-H<sub>2</sub> concentration should be reduced  
224 below  $20 \mu\text{g L}^{-1}$ . Degasification also enhanced acetate and propionate degradation (Figure 3C and  
225 3D). The DM module could be operated with an average biogas flux through the DM of  $1580 \pm$   
226  $220 \text{ mL-biogas m}^{-2} \text{ day}^{-1}$  for 246 days without significant membrane fouling.

227 For further verification, degasification in the DR was temporarily stopped at day 94 in  
228 Phase 1.5 and day 108 in Phase 1.7. The T-COD removal efficiencies immediately decreased  
229 when degasification was stopped (Figure 4A). The results clearly demonstrated that

230 discontinuation of degasification resulted in increased D-H<sub>2</sub> concentrations and VFAs  
231 accumulation, and a subsequent pH drop (Figure 4A and 4B), which emphasizes the contribution  
232 of degasification to process stability. Slower degradation of organic matter by anaerobic than by  
233 aerobic processes hampers widespread application of AD. Based on these findings, we conclude  
234 that degasification is a powerful tool for temporal and long-term stabilization and enhancement of  
235 AD. To date, there have been many attempts to enhance organic matter removal and biogas  
236 production in AD processes, such as the addition of zero-valent iron (Feng et al., 2014; Kong et  
237 al., 2016; Meng et al., 2013), trace elements (W. Zhang et al., 2015), bacteria (J. Zhang et al.,  
238 2015) and NH<sub>4</sub>HCO<sub>3</sub> (Qiao et al., 2013), and the implementation of co-digestion (the  
239 simultaneous digestion of two or more substrates) (Qiao et al., 2013). In contrast, degasification  
240 might be a simpler method to enhance process performance of an AD process because it does not  
241 require the addition of any chemicals or the introduction of complicated equipment.

242

### 243 3.2 Effect of addition of acetate on T-COD removal

244 To investigate the direct effect of acetate on T-COD removal, acetate salt was added to  
245 the influent synthetic wastewater from day 211 to day 246 (Table 1). When the acetate  
246 concentration reached 3 g L<sup>-1</sup> and 4 g L<sup>-1</sup> in Phases 3.4 and 3.5, T-COD removal efficiency  
247 decreased. Therefore, degasification was only applied to DR, and T-COD removal was slightly  
248 improved (Figure 5A). This was attributed to D-H<sub>2</sub> gas removal and subsequent prevention of the  
249 pH drop (Figure 5B and 5C).

250

### 251 3.3 Effect of degasification on CH<sub>4</sub> production

252 Figure 6 shows CH<sub>4</sub> the production rates (mL L<sup>-1</sup>-reactor h<sup>-1</sup>) in DR and CR. As the  
253 T-COD loading rate increased by day 86, the CH<sub>4</sub> production rates increased. Further increases in  
254 the T-COD loading rate in Phase 1.5 resulted in lower CH<sub>4</sub> production (about 150 mL L<sup>-1</sup>-reactor  
255 h<sup>-1</sup>) in the CR because of deterioration of T-COD removal (Figure 1). In contrast, CH<sub>4</sub> production  
256 rates were higher in the DR in Phases 1.5 (about 300 mL L<sup>-1</sup>-reactor h<sup>-1</sup>), 1.7, 2.1, 2.3, 2.5, 3.4  
257 and 3.5 than those (about 250 mL L<sup>-1</sup>-reactor h<sup>-1</sup>) under normal T-COD loading (about 24 g  
258 L<sup>-1</sup>-reactor day<sup>-1</sup>) because of the high T-COD removal efficiencies (Figure 1). These results  
259 indicated that, contrary to expectations, H<sub>2</sub> loss by degasification did not reduce methanogenesis.  
260 It should be noted that CH<sub>4</sub> was also recovered by degasification, which accounted for 5%–18%  
261 of the CH<sub>4</sub> recovered from the reactor headspace. CH<sub>4</sub> and H<sub>2</sub> concentrations in the reactor  
262 headspace were between 73% and 43% and < 5270 ppm, respectively.

263

## 264 5. Conclusions

265 Degasification with degassing membrane enhanced organic matter degradation and  
266 methane gas production in an anaerobic granular sludge reactor under stress conditions, such as  
267 higher organic concentration, shorter hydraulic retention time and addition of acetate. Process  
268 enhancement was attributed to a cascading reaction; specifically, reduction of dissolved hydrogen  
269 concentration, enhancement of volatile fatty acids degradation, and prevention of pH drop. The

270 anaerobic processes often show instability, which has hampered the widespread application of  
271 anaerobic digestion. Overall, the results of this study indicate that degasification is a powerful tool  
272 for temporal and long-term stabilization and enhancement of anaerobic digestion.

273

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280

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349 Figure captions

350

351 Figure 1. T-COD removal efficiencies of the degasification reactor (DR) and the control reactor  
352 (CR) by day 210. The gray areas represent degasification periods.

353

354 Figure 2. Dissolved H<sub>2</sub> (D-H<sub>2</sub>) concentrations, pH (A) and VFA concentrations (B) in DR and CR  
355 in Phase 1. The gray areas represent degasification periods.

356

357 Figure 3. Relationships between T-COD loading rates and T-COD removal efficiencies (A), D-H<sub>2</sub>  
358 concentrations and pH (B), acetate (C) or propionate concentrations (D) for 246 days.

359

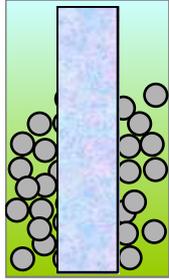
360 Figure 4. T-COD removal efficiencies, VFA concentrations (A) D-H<sub>2</sub> and pH (B) in DR. The gray  
361 area represents a degasification period. Degasification in DR was temporarily stopped at day  
362 94 in Phase 1.5 and day 108 in Phase 1.7.

363

364 Figure 5. The T-COD removal efficiencies (A), D-H<sub>2</sub> and pH (B), and acetate concentrations (C)  
365 in DR and CR in Phase 3. The gray areas represent degasification periods.

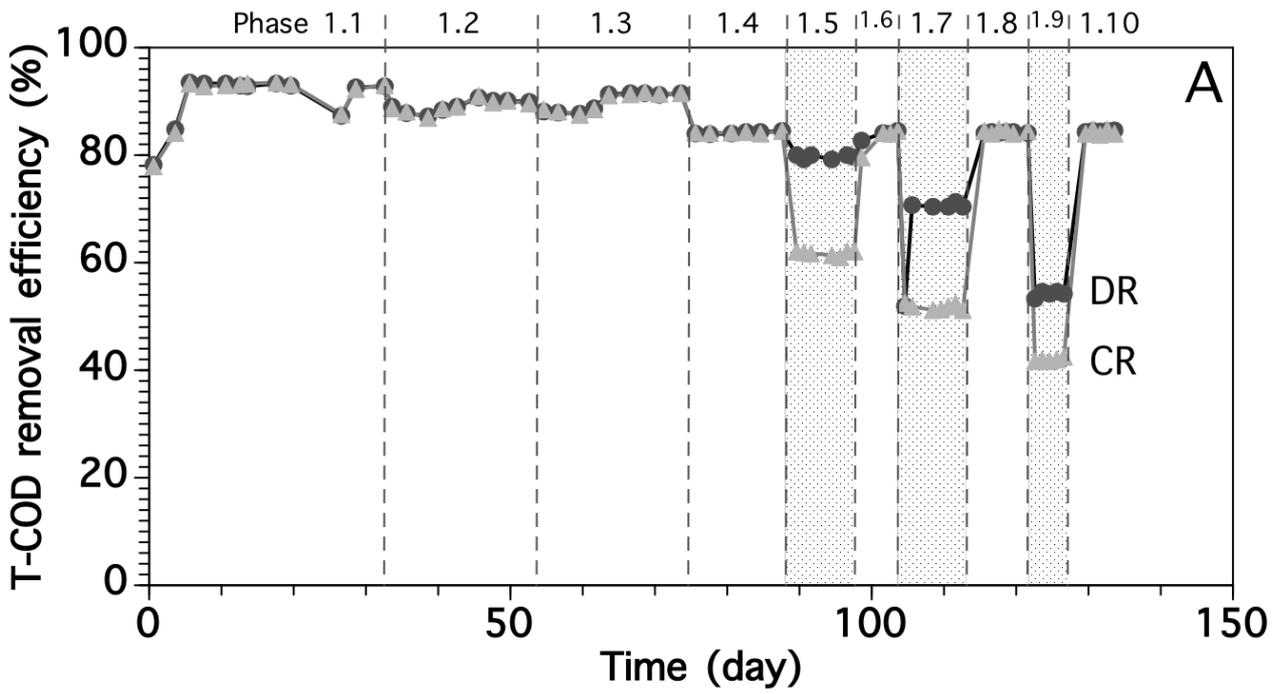
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367 Figure 6. The rates of CH<sub>4</sub> production collected from headspace in the DR and CR and degassing  
368 membrane (degas) in the DR.

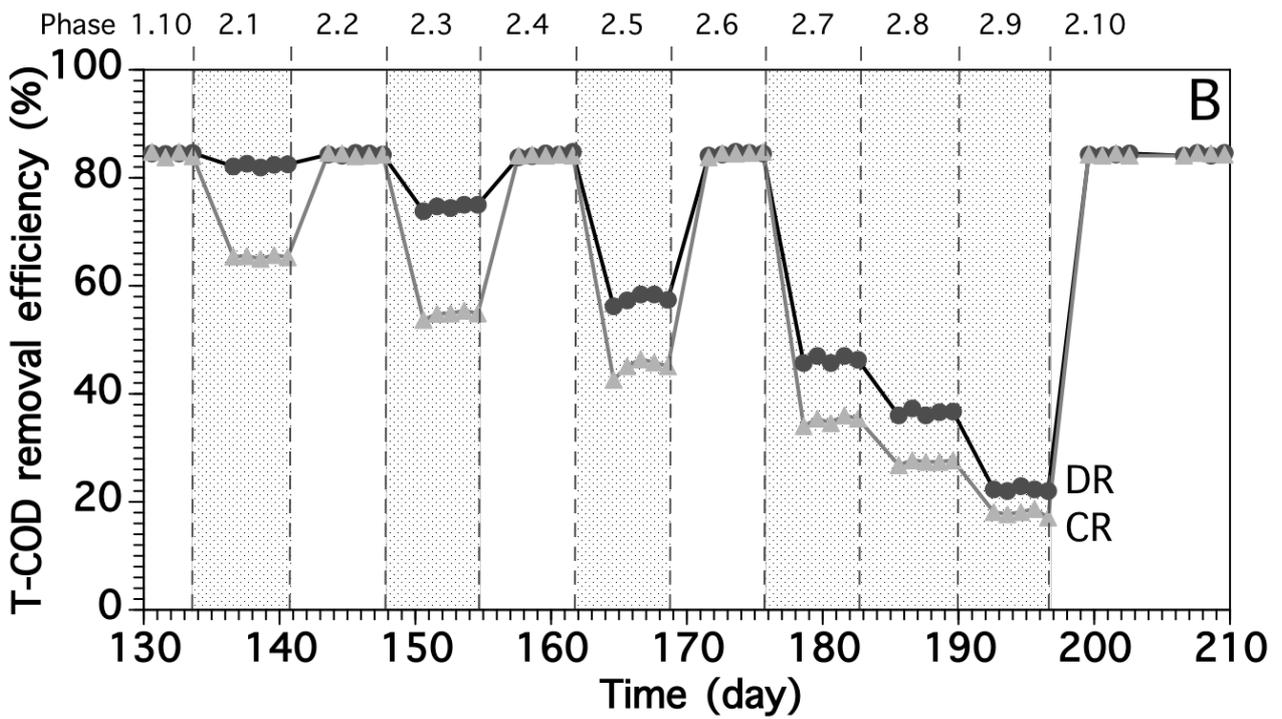


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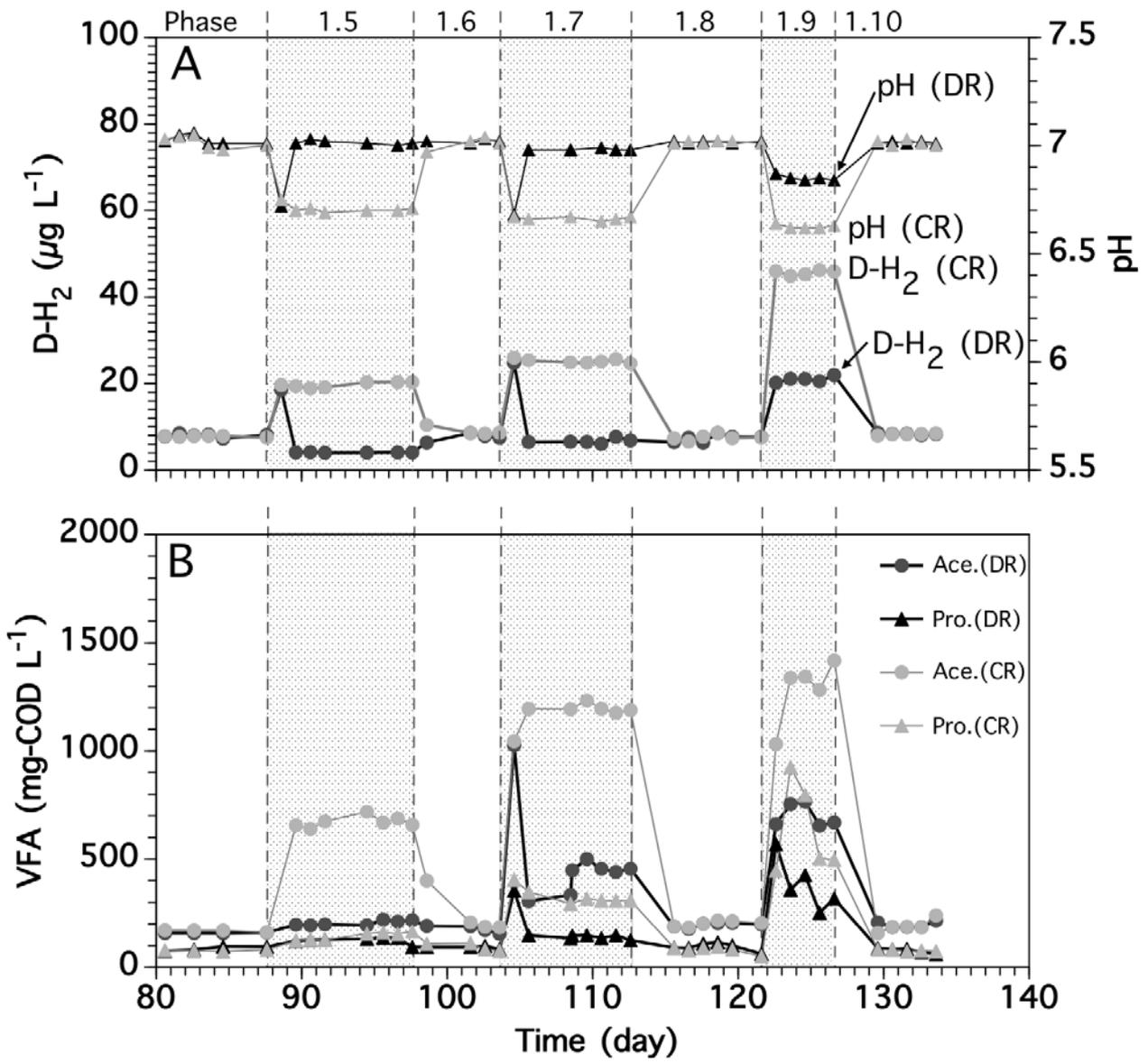
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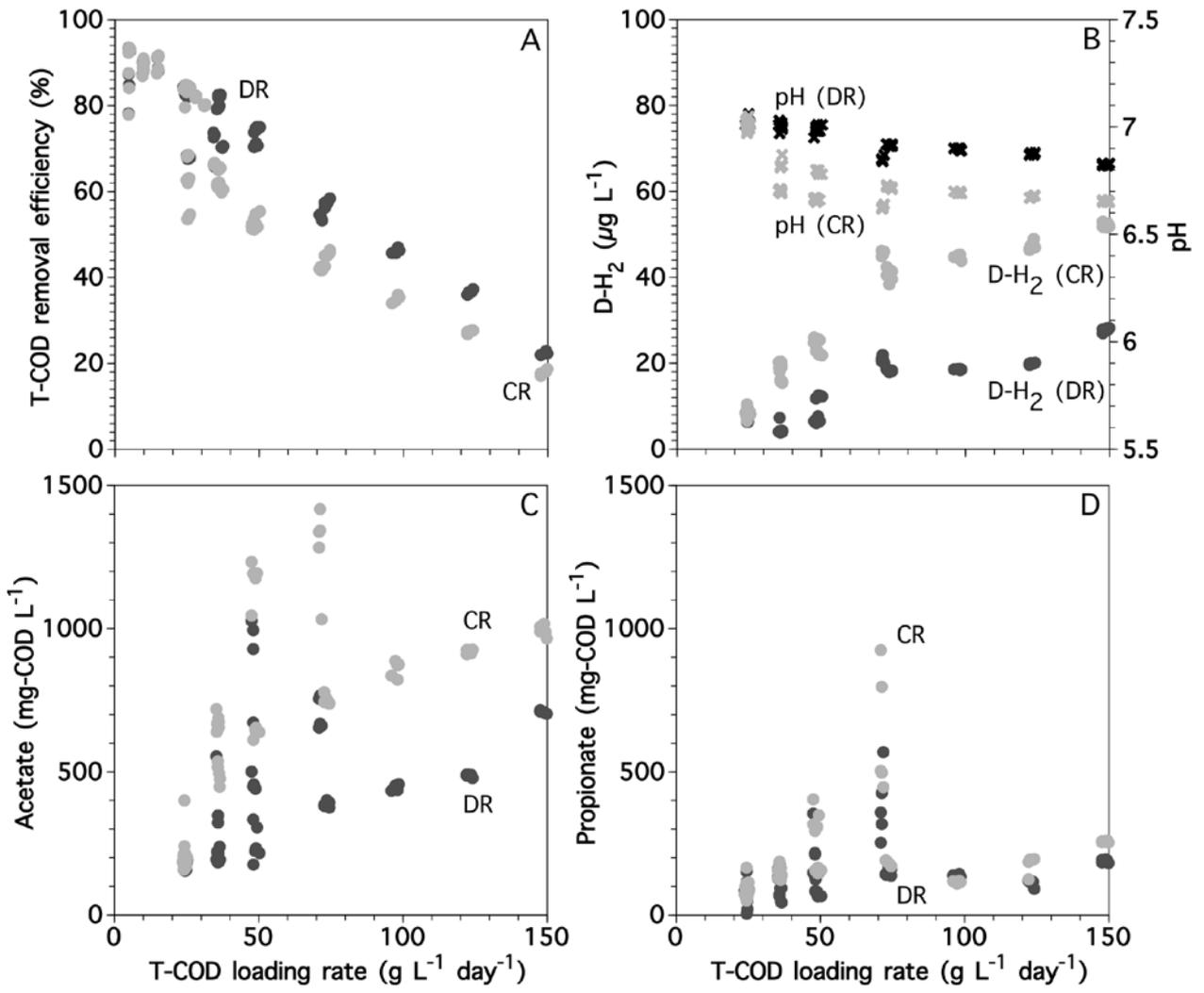
373 Figure 1.

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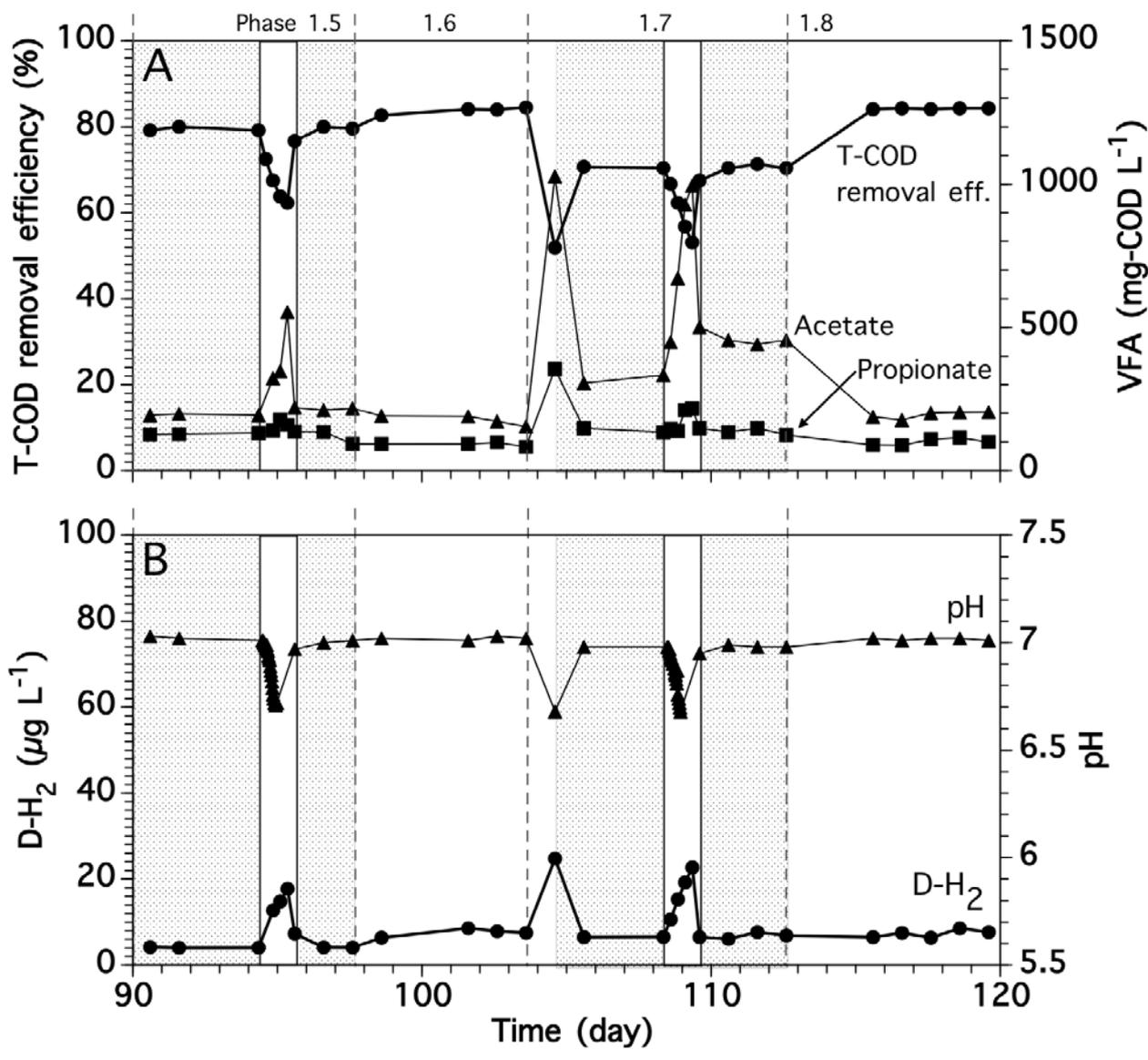
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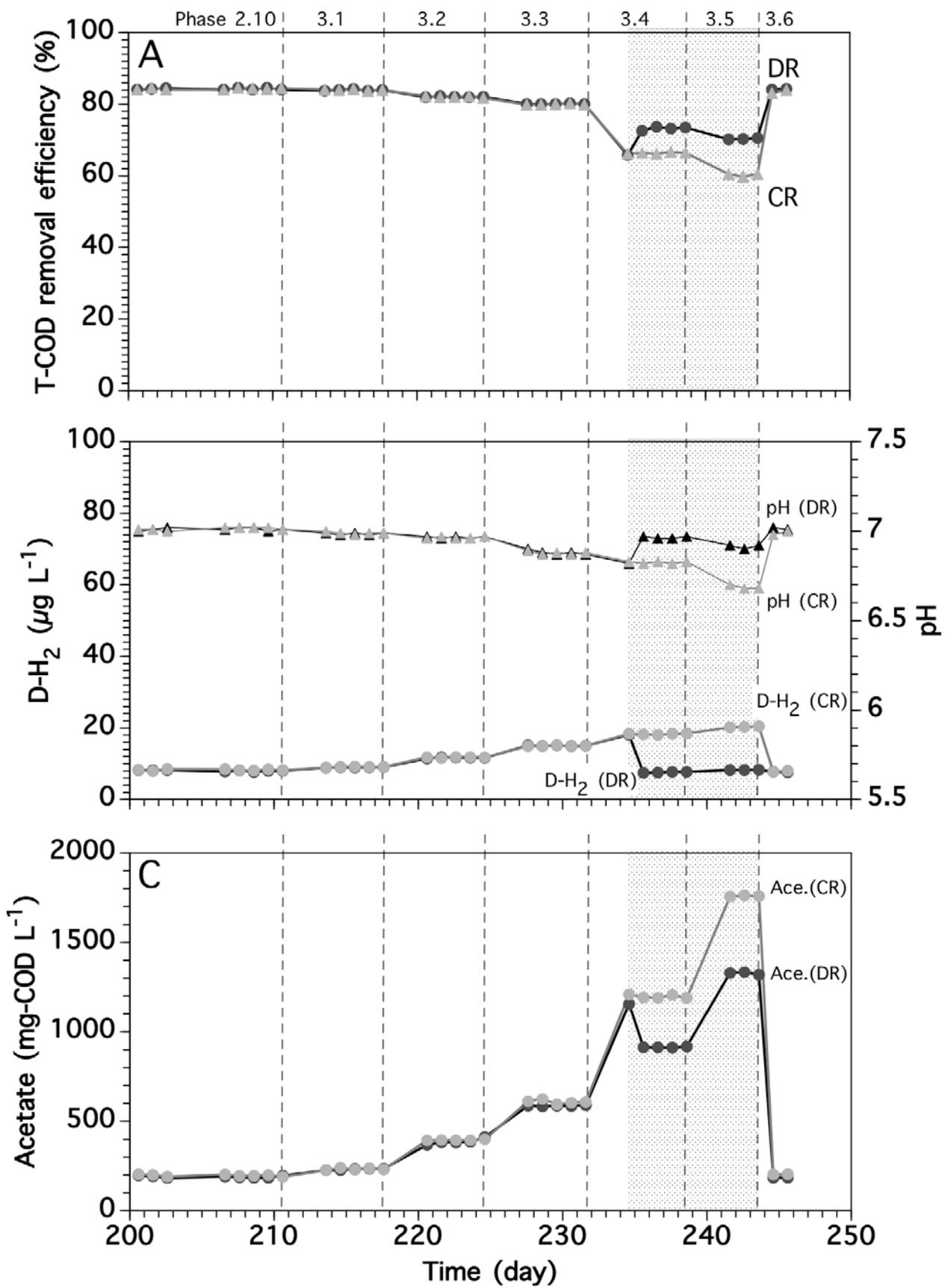
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378 Figure 3.



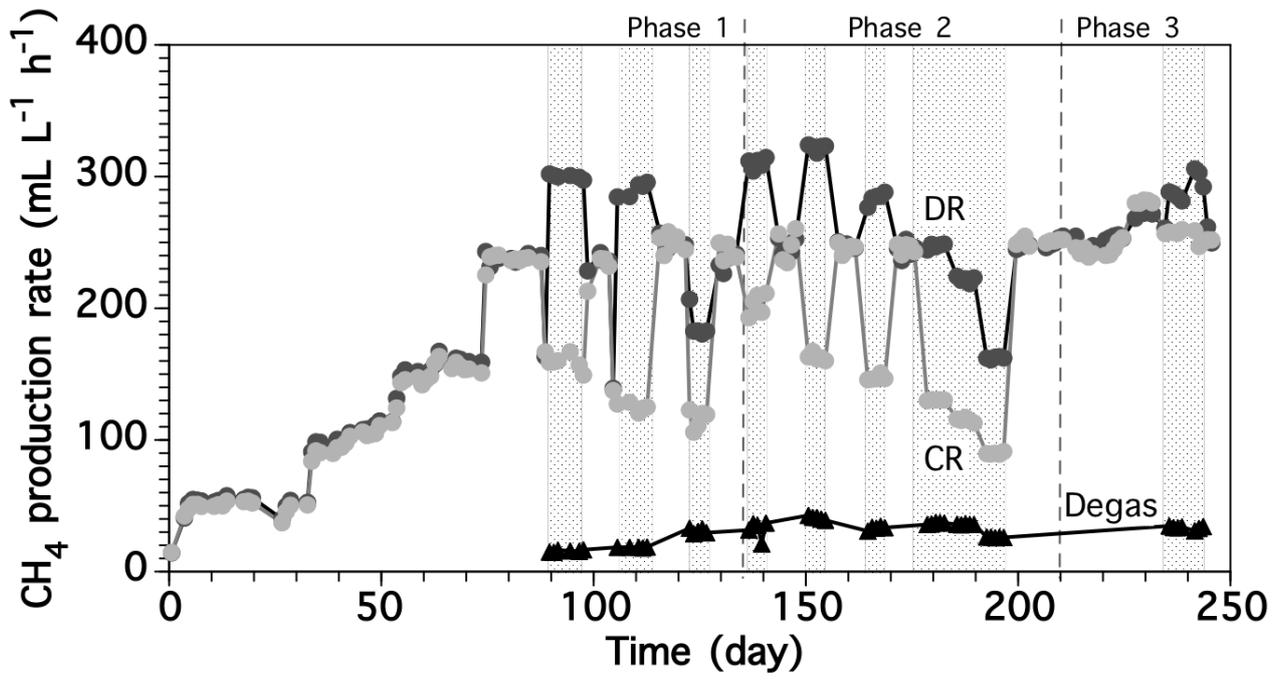
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380 Figure 4.



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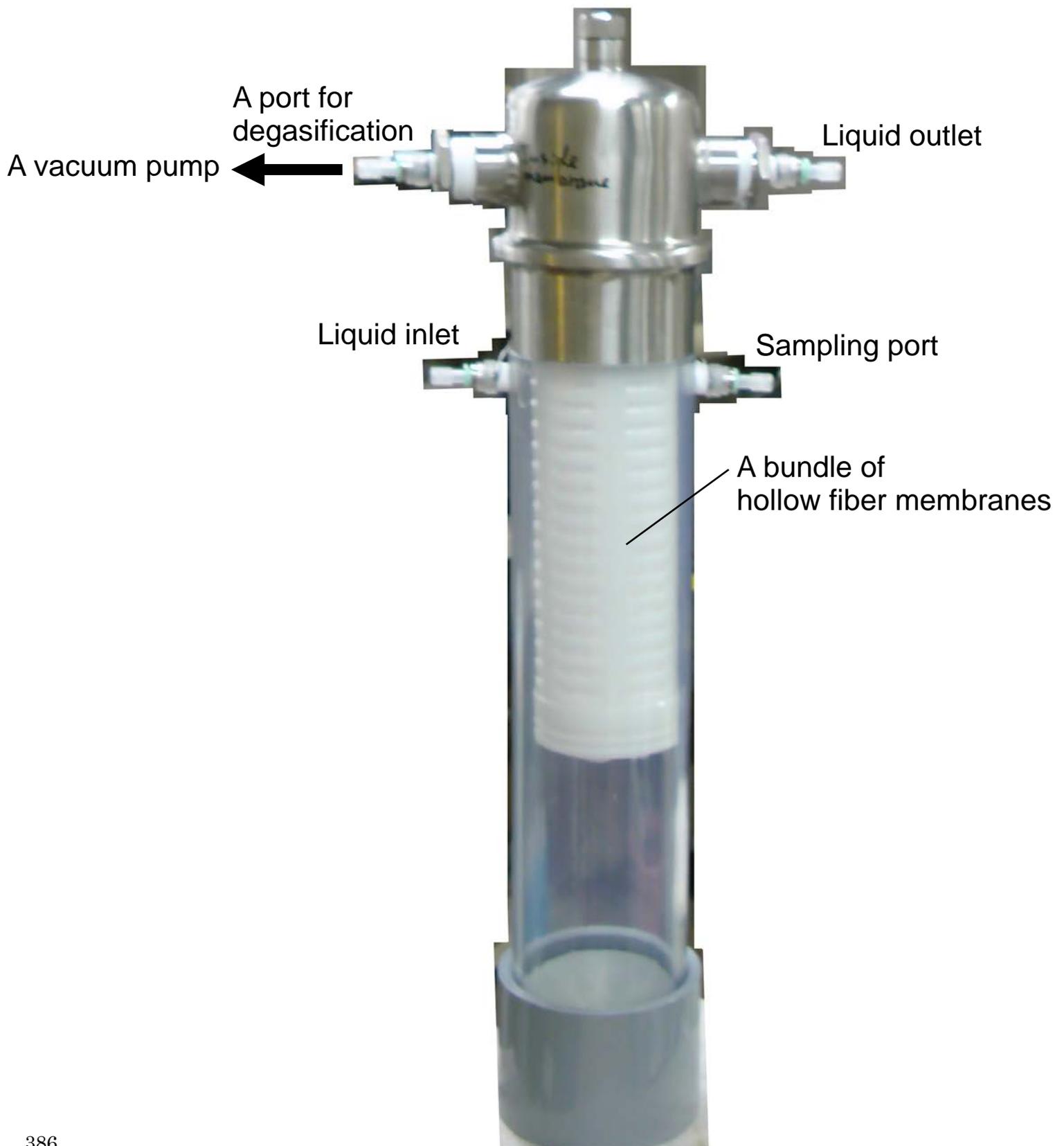
382 Figure 5.



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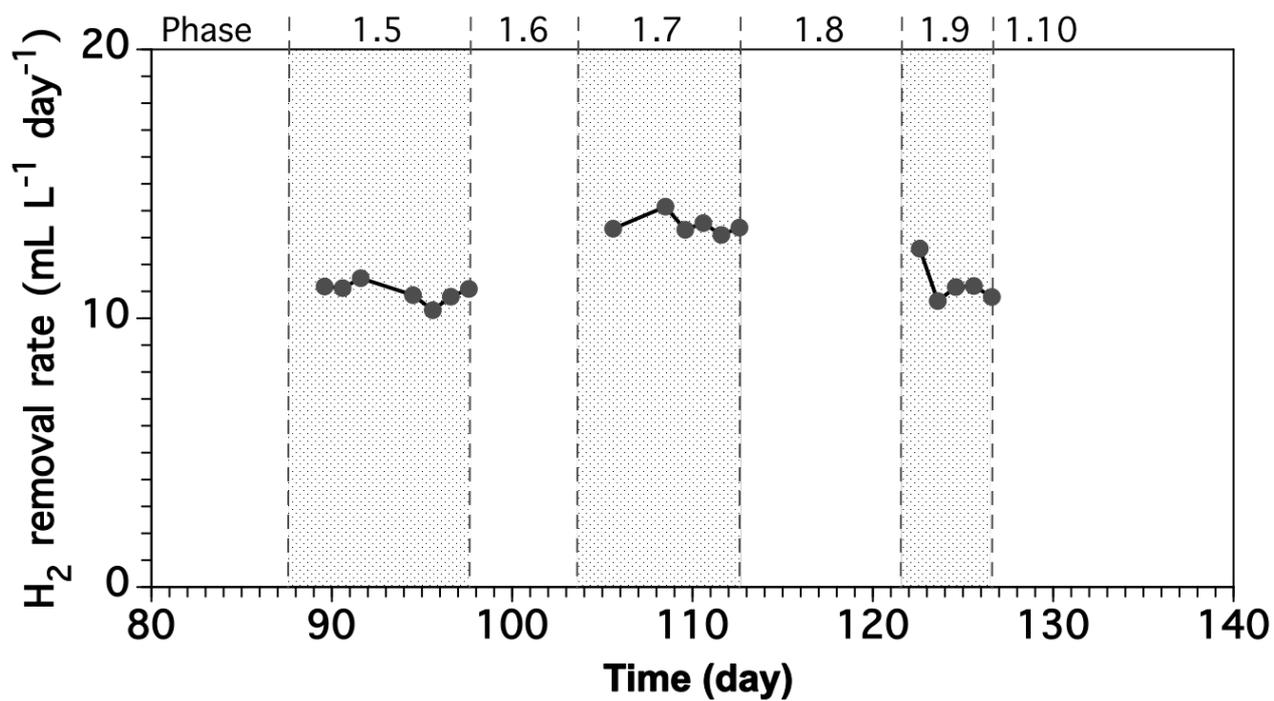
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387 Figure S1. Diagram of a bench-scale reactor in which a bundle of hollow fiber membranes for  
388 degasification was installed.

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394 Figure S2 Dissolved H<sub>2</sub> recovery rates through the degassing membrane.

395

396 Table 1. Summary of the operational conditions of two bench-scale anaerobic granular sludge reactors.

Phase	Degasification	Time (day)	HRT of DR <sup>a</sup> (h)	HRT of CR <sup>b</sup> (h)	Influent T-COD Conc. (mg L <sup>-1</sup> )	T-COD loading rate (g L <sup>-1</sup> -reactor day <sup>-1</sup> )	Conc. of sodium acetate and sodium propionate added
1.1		1-	8.05±0.07	8.08±0.05	1620±61	4.8±0.2	
1.2		34-	8.04±0.06	8.02±0.06	3276±37	9.8±0.1	
1.3		54-	8.05±0.05	8.01±0.05	4969±63	14.8±0.2	
1.4		75-	8.04±0.04	8.04±0.05	8167±82	24.3±0.3	
1.5	Applied	87-	8.00±0.05	8.00±0.05	11927±101	35.8±0.3	
1.6		97-	8.00±0.03	8.04±0.03	8150±58	24.4±0.1	
1.7	Applied	103-	8.06±0.05	8.05±0.05	16200±126	48.2±0.5	
1.8		112-	8.02±0.05	8.03±0.04	8150±55	24.4±0.2	
1.9	Applied	121-	8.05±0.03	8.07±0.03	23920±110	71.2±0.4	
1.10		126-	8.06±0.03	8.04±0.05	8120±84	24.2±0.3	
2.1	Applied	133-	5.36±0.02	5.36±0.02	8080±84	36.2±0.3	
2.2		141-	8.05±0.07	8.06±0.05	8140±55	24.3±0.2	
2.3	Applied	148-	4.04±0.04	4.04±0.04	8260±89	49.0±0.8	
2.4		154-	8.03±0.09	8.03±0.09	8160±55	24.4±0.4	
2.5	Applied	161-	2.67±0.00	2.67±0.00	8200±100	73.6±0.9	
2.6		168-	7.99±0.06	8.02±0.04	8160±55	24.5±0.2	
2.7	Applied	175-	2.00±0.01	2.01±0.01	8160±55	97.6±1.0	
2.8	Applied	183-	1.61±0.00	1.61±0.01	8240±55	122.9±0.9	
2.9	Applied	190-	1.33±0.00	1.33±0.00	8260±55	148.7±1.0	
2.10		196-	7.96±0.04	7.97±0.06	8222±67	25.9±0.2	
3.1		211-	7.94±0.03	7.94±0.05	8560±55	27.9±0.1	0.25 g-Ace L <sup>-1</sup>
3.2		221-	7.97±0.04	7.95±0.03	9260±55	31.0±0.2	1.0 g-Ace L <sup>-1</sup>
3.3		228-	7.93±0.05	7.93±0.03	10260±55	34.3±0.2	2.0 g-Ace L <sup>-1</sup>
3.4	Applied	235-	7.98±0.03	7.93±0.05	11360±55	37.2±0.3	3.0 g-Ace L <sup>-1</sup>
3.5	Applied	242-245	7.93±0.03	8.02±0.06	12367±58	24.9±0.2	4.0 g-Ace L <sup>-1</sup>

3.6		245-246	8.00±0.04	7.94±0.04	8250±71	24.9±0.2	
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398 <sup>a</sup> The reactor conducted by degasification.

399 <sup>b</sup> The reactor in which a degassing membrane was installed but degasification was not applied.

