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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

Abstract

A hollow fiber degassing membrane (DM) was applied to enhance organic matter degradation and methane gas production of anaerobic granular sludge process by reducing the dissolved hydrogen gas (D-H$_2$) concentration in the liquid phase. DM was installed in the bench-scale anaerobic granular sludge reactors and D-H$_2$ was removed through DM using a vacuum pump. Degasification improved the organic matter degradation efficiency to 79% while the efficiency was 62% without degasification at 12,000 mg L^{-1} of the influent T-COD concentration. Measurement of D-H$_2$ concentrations in the liquid phase confirmed that D-H$_2$ was removed by degasification. Furthermore, the effect of acetate concentrations on the organic matter degradation efficiency was investigated. At acetate concentrations above 3 g L^{-1}, organic matter degradation deteriorated. Degasification enhanced the propionate and acetate degradation. These results suggest that degasification reduced D-H$_2$ concentration and volatile fatty acids concentrations, prevented pH drop, and subsequent enhanced organic matter degradation.

1. Introduction

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 (pH_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 pH_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 pH_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_2 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_2 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_2 in biogas by co-digestion of

80　　coffee grounds and dewatered activated sludge and addition of an alkali chemical (NH_4HCO_3). Li

81　　et al. accelerated H_2 consumption and VFAs decomposition by the addition of azo dye, which

82　　serves as an electron acceptor of H_2 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_2 ($Fe^0 +$

85　　$2H^+ \rightarrow Fe^{2+} + H_2$), D-$H_2$ 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_2 consumption and therefore

88　　increased Chemical Oxygen Demand (COD) removal and methane (CH_4) production (Feng et al.,

89　　2014; Kong et al., 2016; Meng et al., 2013). Bioaugmentation was also effective to reduce the

90   D-H_2 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_2 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_2 and

101   CO_2 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_4 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_4 gas concentration decreasing from 104 mg COD L^{-1} to 14 mg COD

113   L^{-1} 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_4 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^{-2} day^{-1} for 18

118   months without significant membrane fouling.

119   In this study, $D-H_2$ 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_4 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_4 production, after which

123   degasification was applied. In addition, the effect of acetate concentrations on COD removal

124   efficiency and CH_4 production were investigated. Finally, the mechanisms of recovery from the

125   process failure were discussed based on the experimental data ($D-H_2$ concentration, pH level and

126   removed H_2 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 (0.8×30 cm) after filtering through a 0.2-mm membrane. The pH was determined using a

162   pH electrode. The concentrations of CH_4 and 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°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^{-1} 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^{-1} 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^{-1}-reactor day^{-1} 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^{-1} 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^{-1}-reactor day^{-1} from Phases 2.1 to 2.3. Conversely, degasification no longer significantly

10

190   improved T-COD removal when the T-COD loading rate was above 70 g L^{-1}-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 H$_2$ (D-H$_2$)

194   concentration, VFA concentrations and pH in DR and CR were determined. The D-H$_2$

195   concentrations were less than 10 μg L^{-1} under normal T-COD loading (approximately 24 g

196   L^{-1}-reactor day^{-1}) in Phases 1.4, 1.6, 1.8 and 1.10 (Figure 2A). Additionally, the D-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 H$_2$ gas removal rates by degasification confirmed removal of

200   D-H$_2$ through the DM. CH$_4$, CO$_2$ and 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

11

210   accumulation. The butyrate concentrations were <250 mg-COD L^{-1} and the other VFAs (lactate,

211   formate and isobutyrate) concentrations were <40 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_2 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_2 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 g L^{-1}-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_2

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_2 concentration should be reduced

224   below 20 μg 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 ±

226   220 mL-biogas m^{-2} 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

12

230   discontinuation of degasification resulted in increased D-H_2 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_4HCO_3 (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^{-1} and 4 g L^{-1} 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_2 gas removal and subsequent prevention of the

249   pH drop (Figure 5B and 5C).

250

251 3.3 Effect of degasification on CH_4 production

252   Figure 6 shows CH_4 the production rates ($mL\ L^{-1}$-reactor h^{-1}) in DR and CR. As the

253 T-COD loading rate increased by day 86, the CH_4 production rates increased. Further increases in

254 the T-COD loading rate in Phase 1.5 resulted in lower CH_4 production (about 150 $mL\ L^{-1}$-reactor

255 h^{-1}) in the CR because of deterioration of T-COD removal (Figure 1). In contrast, CH_4 production

256 rates were higher in the DR in Phases 1.5 (about 300 $mL\ L^{-1}$-reactor h^{-1}), 1.7, 2.1, 2.3, 2.5, 3.4

257 and 3.5 than those (about 250 $mL\ L^{-1}$-reactor h^{-1}) under normal T-COD loading (about 24 g

258 L^{-1}-reactor day^{-1}) because of the high T-COD removal efficiencies (Figure 1). These results

259 indicated that, contrary to expectations, H_2 loss by degasification did not reduce methanogenesis.

260 It should be noted that CH_4 was also recovered by degasification, which accounted for 5%–18%

261 of the CH_4 recovered from the reactor headspace. CH_4 and H_2 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 cascadic reaction; specifically, reduction of dissolved hydrogen

269 concentration, enhancement of volatile fatty acids degradation, and prevention of pH drop. The

14

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.

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348

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_2 (D-H_2) 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_2

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_2 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_2 and pH (B), and acetate concentrations (C)

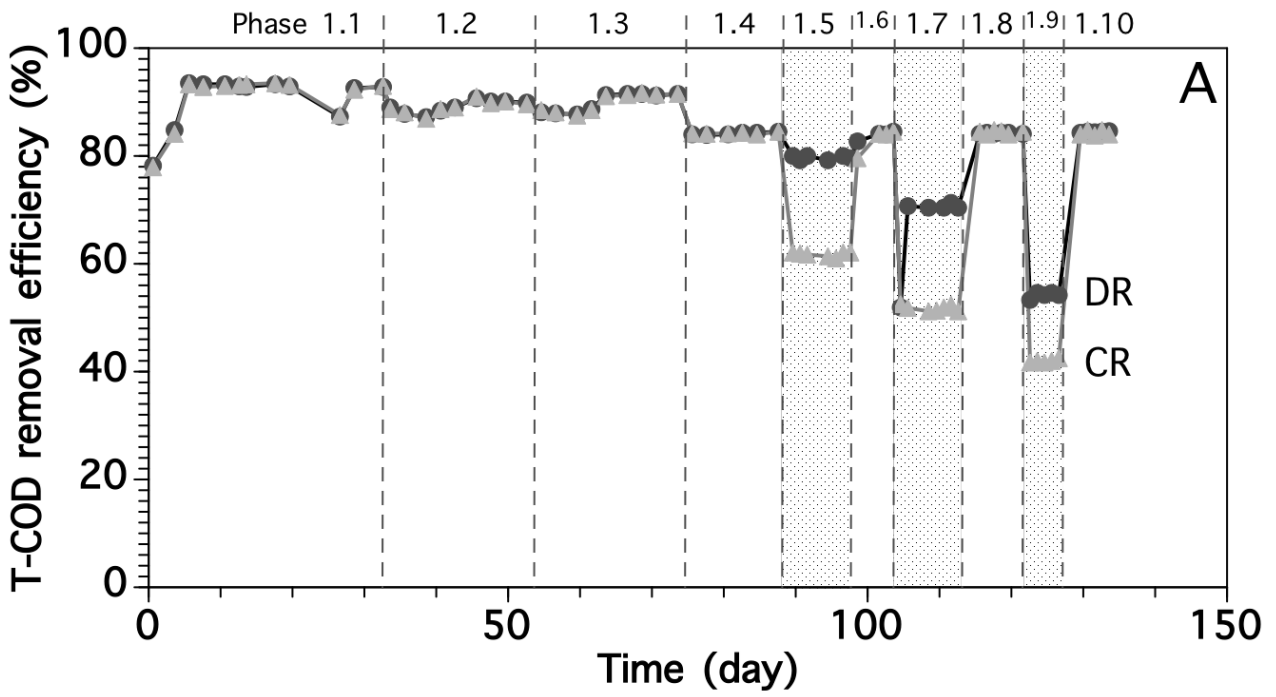365   in DR and CR in Phase 3. The gray areas represent degasification periods.

366

367   Figure 6. The rates of CH_4 production collected from headspace in the DR and CR and degassing
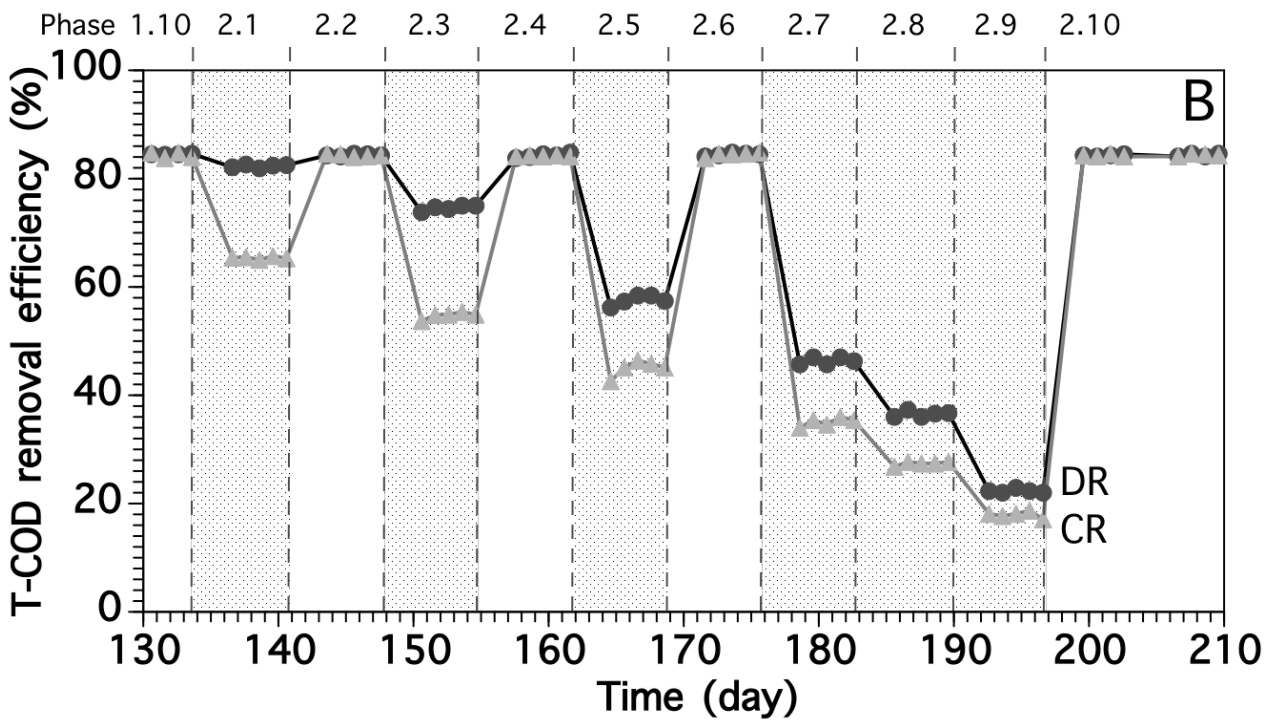
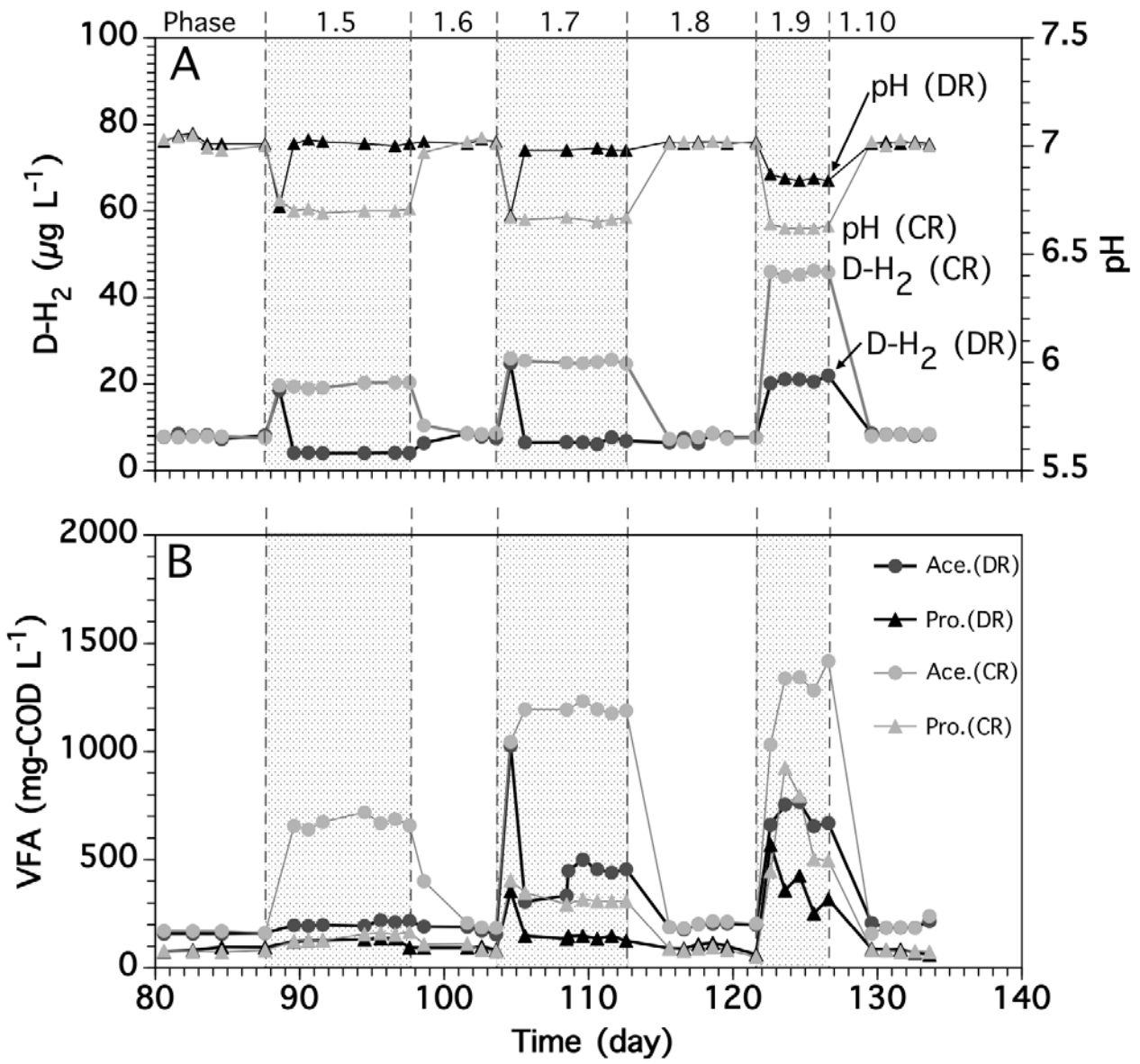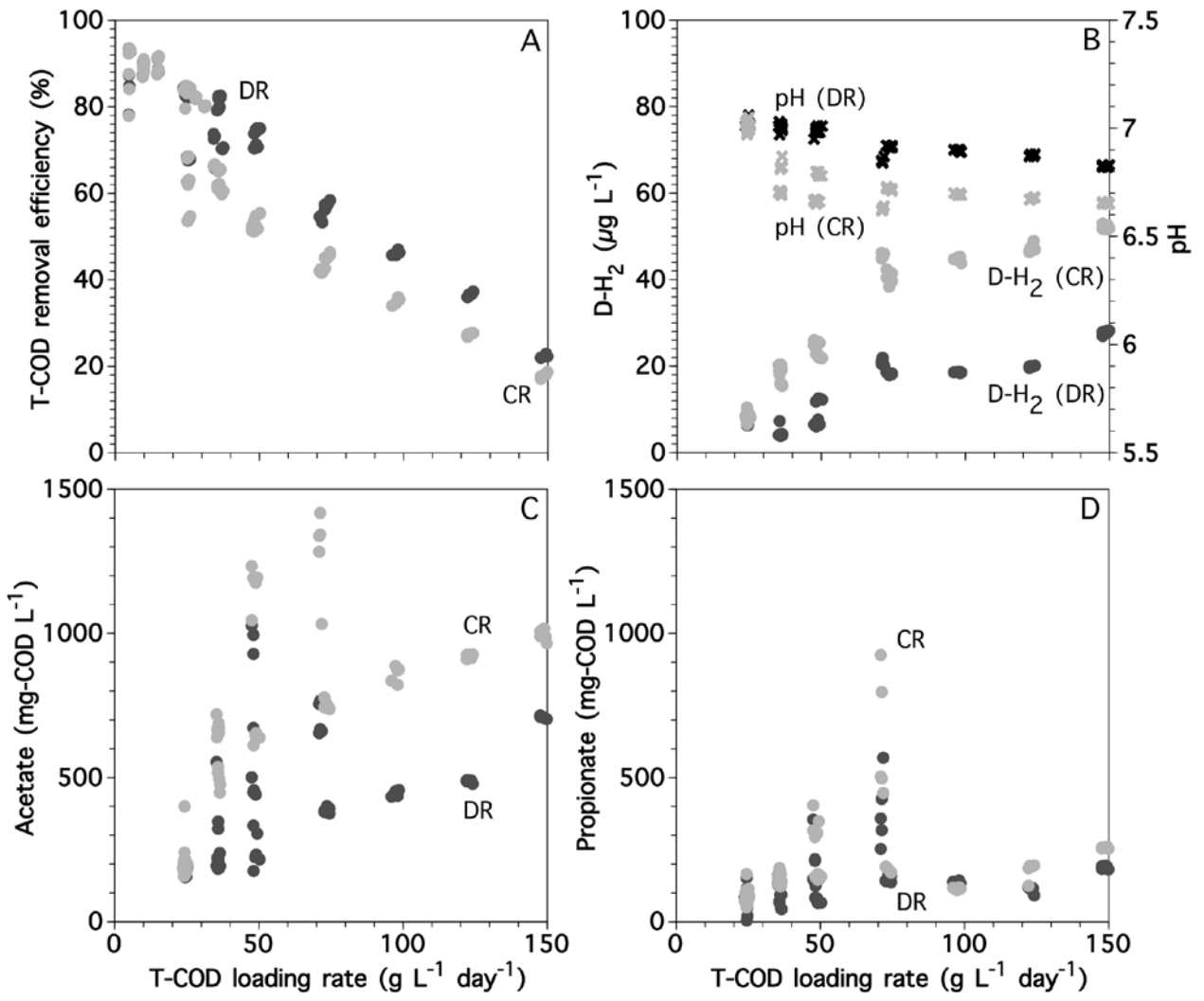368   membrane (degas) in the DR.
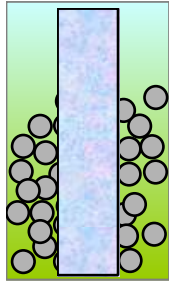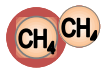
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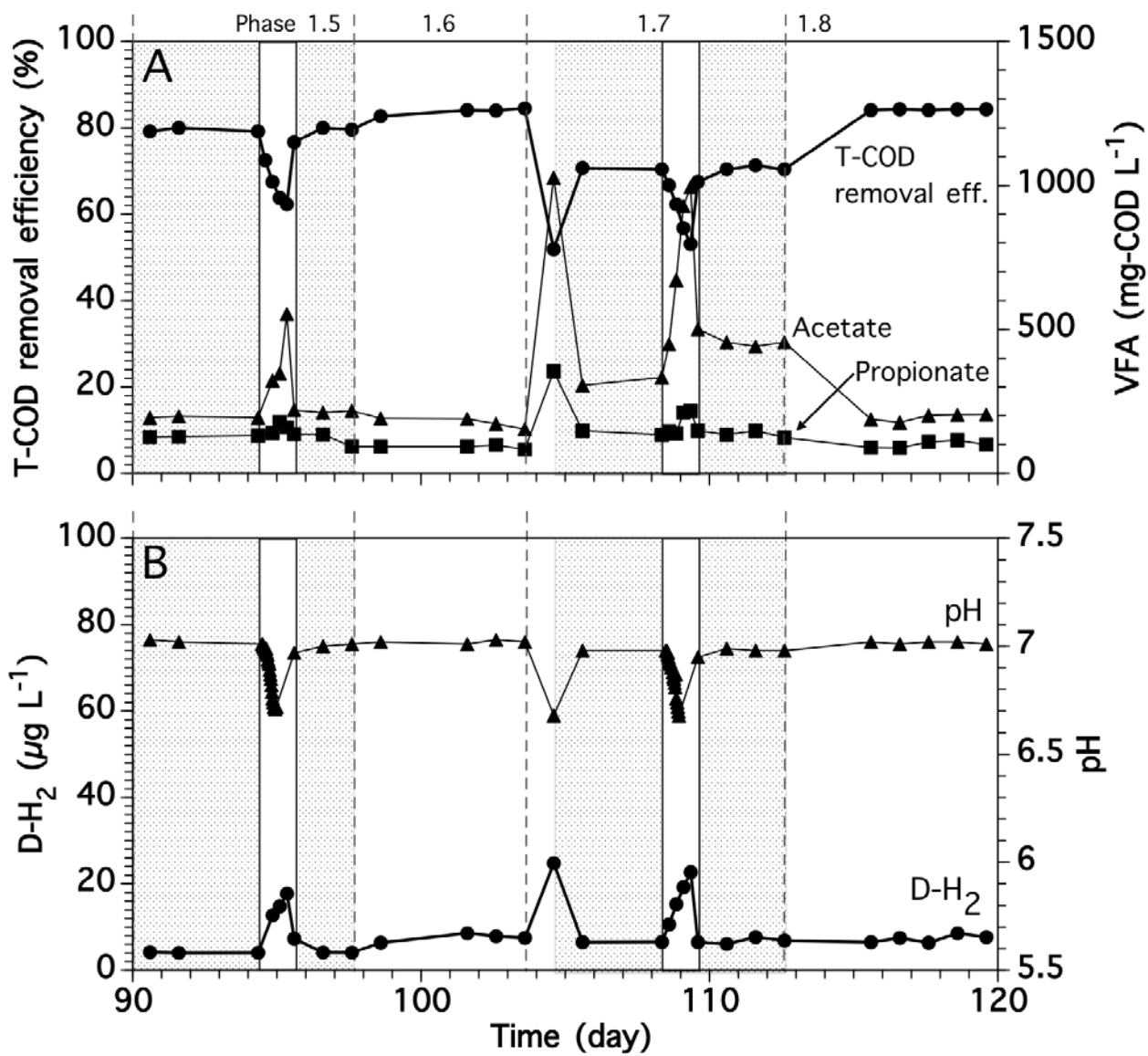
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373   Figure 1.
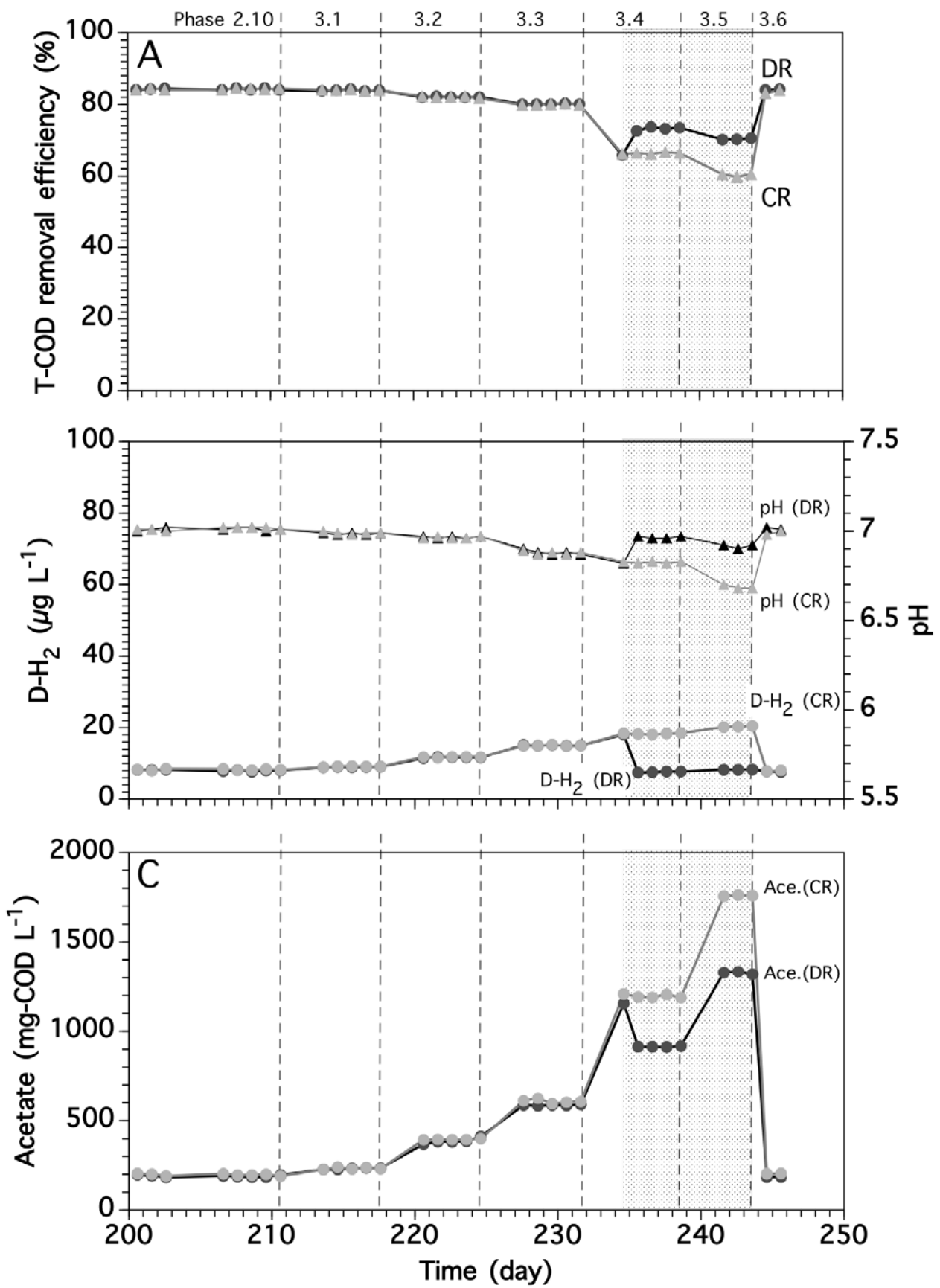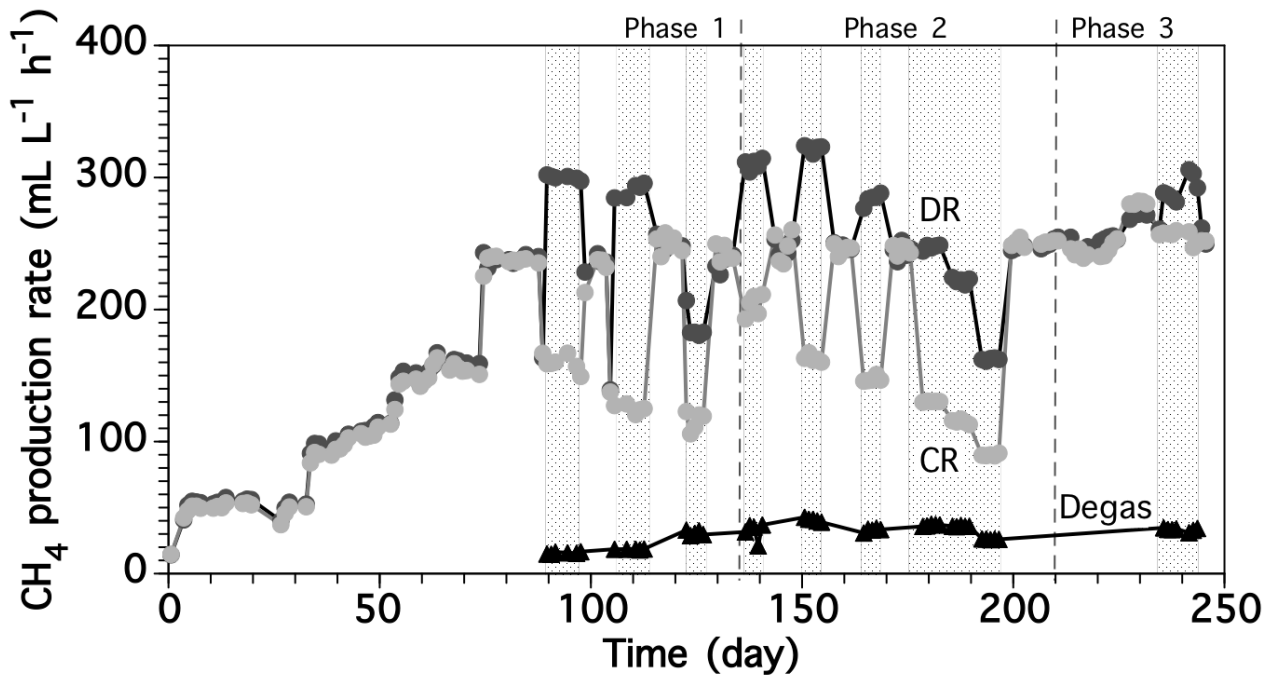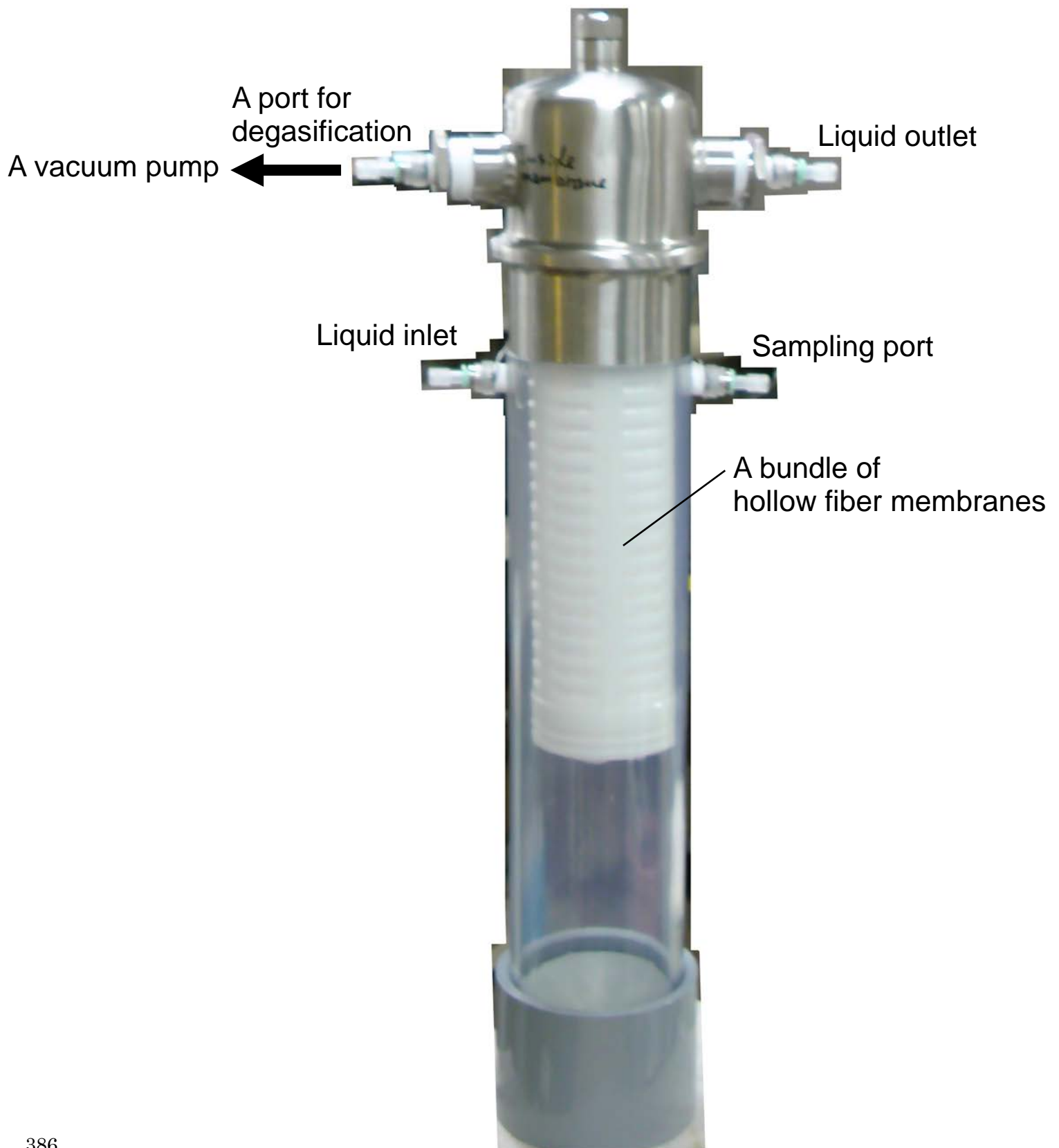
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375

376   Figure 2.

377

378   Figure 3.

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380   Figure 4.

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382   Figure 5.

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384   Figure 6.

385

A port for
degasification

Liquid outlet

A vacuum pump

Liquid inlet

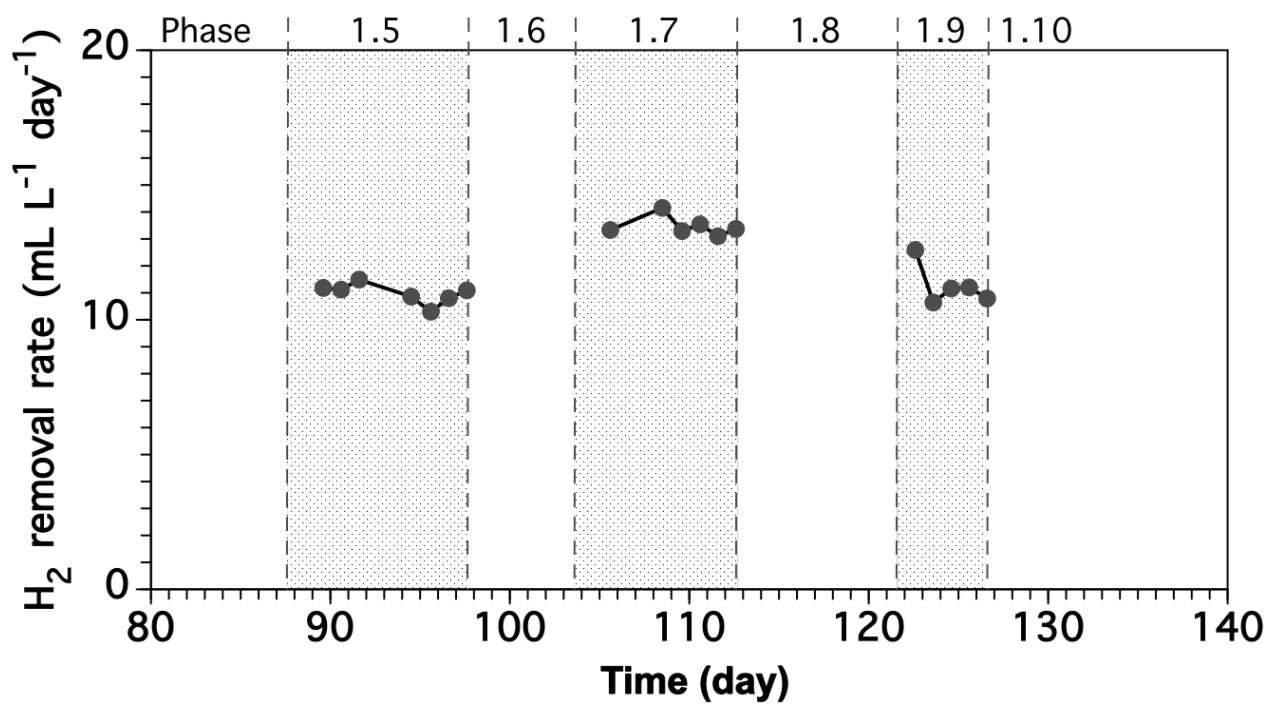Sampling port

A bundle of
hollow fiber membranes

386
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_2 recovery rates through the degassing membrane.

395

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

Phase	Degasification	Time (day)	HRT of DR[a] (h)	HRT of CR[b] (h)	Influent T-COD Conc. (mg L^{-1})	T-COD loading rate (g L^{-1}-reactor day^{-1})	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^{-1}
3.2		221-	7.97±0.04	7.95±0.03	9260±55	31.0±0.2	1.0 g-Ace L^{-1}
3.3		228-	7.93±0.05	7.93±0.03	10260±55	34.3±0.2	2.0 g-Ace L^{-1}
3.4	Applied	235-	7.98±0.03	7.93±0.05	11360±55	37.2±0.3	3.0 g-Ace L^{-1}
3.5	Applied	242-245	7.93±0.03	8.02±0.06	12367±58	24.9±0.2	4.0 g-Ace L^{-1}

| 3.6 | | 245-246 | 8.00±0.04 | 7.94±0.04 | 8250±71 | 24.9±0.2 | |

397

398   [a] The reactor conducted by degasification.

399   [b] The reactor in which a degassing membrane was installed but degasification was not applied.