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1 **Excellent performance of anaerobic membrane bioreactor in treatment of**  
2 **distillery wastewater at pilot scale**

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## 11 **Abstract**

12 A pilot-scale anaerobic membrane bioreactor (AnMBR) was designed and optimized for  
13 the treatment of real distillery wastewater. A low hydraulic retention time of 3.5 days  
14 was reached after only 3 weeks. The AnMBR could treat up to 3.97 g<sub>COD</sub>/L/day with  
15 high biogas production at 1.36 NL<sub>biogas</sub>/L<sub>bioreactor</sub>/day. The performances of an AnMBR  
16 and an anaerobic packed-bed bioreactor for treating the same wastewater were  
17 compared. The AnMBR had a shorter start-up period (21 days), a higher COD removal  
18 efficiency (96.9%), and higher stability and methane production (0.26 L<sub>CH<sub>4</sub></sub>/g<sub>CODinput</sub>),  
19 indicating the interest of investigating AnMBR industrialization. The membrane  
20 performance was also studied, demonstrating a long cleaning cycle interval of at least  
21 44 days. The transmembrane pressure and Food-to-Microorganism ratio were defined to  
22 minimize membrane fouling without affecting the anaerobic digestion performance.

## 23 **Keywords**

24 Membrane bioreactor; Wastewater treatment; Anaerobic digestion; Packed-bed  
25 bioreactor; Biogas production

## 26 **Abbreviations**

27 AnMBR: Anaerobic membrane bioreactor

28 APBR: Anaerobic packed-bed reactor

29 COD: Chemical oxygen demand (g/L)

30 CSTR: Continuously stirred tank reactor

31 EPS: Extracellular polymeric substances

32 F/M: Food to microorganism ratio (g<sub>COD</sub>/g<sub>MLVSS</sub>/day)

33 FOS/TAC: Volatile fatty acids content / Buffer capacity

34 HRT: Hydraulic retention time (day)

35 MLSS: Mixed liquor suspended solids (g/L)

36 MLVSS: Mixed liquor volatile suspended solids (g/L)

37 NPOC: Non-purgeable organic carbon

38 OLR: Organic loading rate (g<sub>COD</sub>/L/day)

- 39 SMP: Soluble microbial particles
- 40 SRT: Sludge retention time (day)
- 41 TMP: Transmembrane pressure (bar<sub>g</sub>)
- 42 VFA: Volatile fatty acids (mg/L)

43 1 Introduction

44 The interest in and need for renewable energy are increasing with global warming.  
45 Wastewater treatment processes can become net producers of renewable energy by  
46 converting the organic pollutants of wastewater to biogas via anaerobic digestion [1].  
47 However, researching and selecting appropriate technology to treat wastewater and  
48 recover energy remain major challenges. Different types of bioreactors have already been  
49 used on an industrial scale for anaerobic digestion for wastewater treatment. The reactor  
50 design must consider the slow growth rate of microorganisms involved in anaerobic  
51 digestion. This must exceed the dilution rate to prevent biomass washout [2].

52 The most commonly used bioreactor type was the continuous stirred tank reactor (CSTR)  
53 until the 1960s [3]. However, the main drawback of this type of bioprocess relates to  
54 combining the sludge retention time (SRT) and hydraulic retention time (HRT). Thus, the  
55 retention time is limited by the growth rate of microorganisms. The HRT is usually 20–  
56 40 days, leading to an important bioreactor volume [2]. Few technologies have been  
57 developed to decouple the SRT and HRT [3]. The main biomass retention technologies  
58 used on an industrial scale can be separated into two classes: biofilm-based bioreactors,  
59 such as the anaerobic packed-bed reactor (APBR), and self-immobilized microorganism  
60 bioreactors, such as the upflow anaerobic sludge blanket (UASB) [2,3].

61 In packed-bed technology, the reactor is filled with inert and stationary material whose  
62 characteristics (e.g., size, shape, porosity, specific surface area) must be considered  
63 during bioreactor design. In the APBR, microorganisms are fixed on the surface of  
64 packing material, allowing a high SRT compared to the HRT. Sludge particles are also  
65 trapped in the interstices of the packing material, increasing the biomass retention [3].

66 The main advantages of this type of reactor are its simplicity of construction and easy  
67 operation and start-up. This technology also has low construction and operating costs,  
68 and no agitation is required [2], while a recirculation loop can sometimes be used.  
69 However, due to possible clogging of the fixed bed, this technology cannot be used to  
70 treat wastewater with a high concentration of suspended solids [3]. This technology can  
71 be used with a relatively high loading rate of up to 10 g<sub>COD</sub>/L/day [3].

72 New technologies based on membrane filtration for biomass retention are currently being  
73 studied for anaerobic digestion. The membrane bioreactor is an attractive technology that  
74 couples anaerobic wastewater treatment with membrane filtration. The anaerobic  
75 membrane bioreactor (AnMBR) exists in three different configurations: external cross-  
76 flow, internal submerged, and external submerged membrane. In the external cross-flow  
77 configuration, the membrane is separated from the reactor. Bioreactor sludge is pumped  
78 through the membrane, creating a positive pressure leading to permeate production, and  
79 the retentate returns to the bioreactor [2].

80 The main advantages of the AnMBR are its high treatment capacity, up to 30 g<sub>COD</sub>/L/day,  
81 and high effluent quality without suspended solids [2]. This high treatment capacity can  
82 be reached due to the high biomass concentrations and low HRT obtained with this  
83 technology. Membrane filtration allows complete decoupling of the HRT and SRT.  
84 Moreover, the permeate can be reused for some purposes without additional treatment  
85 [4]. The AnMBR has been shown to have high biological stability and carbon removal  
86 efficiency [5,6].

87 However, membrane bioreactors are not usually used on an industrial scale for anaerobic  
88 treatment. Control of membrane fouling by liquid/gas recirculation and chemicals makes

89 this process more complex to design than other bioreactor types and leads to higher  
90 operating costs [2]. Membrane fouling in the bioreactor is usually caused by organic  
91 matter attached to the membrane surface or trapped in the membrane pores. This organic  
92 matter is composed of microorganisms, soluble microbial particles (SMP), and  
93 extracellular polymeric substances (EPS). Generally, SMP and EPS are proteins or  
94 hydrocarbon molecules secreted by microorganisms during growth and methane  
95 production. Meng et al. [7] showed that SMP and EPS are the main foulants in AnMBR.  
96 The concentration, composition, and size of SMP and EPS were impacted by controlling  
97 parameters such as the organic loading rate (OLR, expressed as  $\text{g}_{\text{COD}}/\text{L}/\text{day}$ ), SRT, and  
98 food-to-microorganism ratio (F/M, expressed as  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$ ) [8,9,10]. Controlling  
99 the SRT for a given OLR allows control of the biomass concentration in the bioreactor.  
100 Thus, controlling the SRT enables control of the F/M ratio. The amount of SMP and EPS  
101 produced by microorganisms depends on the F/M ratio.

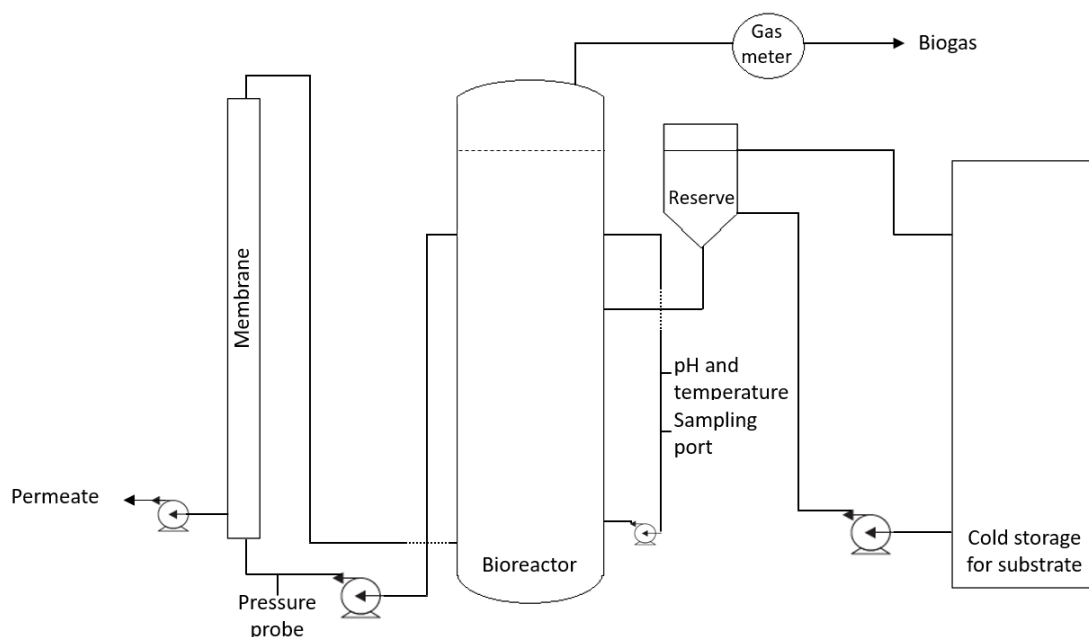
102 This study investigates the start-up phase and overall performance of an AnMBR treating  
103 distillery wastewater at pilot-scale. The anaerobic digestion performance of AnMBR  
104 technology was studied together with an APBR process treating the same wastewater.  
105 APBR technology was used as a reference for this wastewater, as this technology is  
106 currently used for industrial-scale treatment in the distillery. Membrane fouling was also  
107 investigated.

## 108 2 Materials and methods

### 109 2.1 AnMBR

#### 110 2.1.1 Plant description

111 Fig. 1 shows the outline of the pilot-scale AnMBR. The total reactor volume was 170 L  
112 with a working volume of 150 L and an internal diameter of 0.4 m.  
113 The reactor was connected to an external membrane module with a liquid recirculation  
114 pump (IWAKI MDT15). Two membranes were tested in this study. The first was a spiral-  
115 wound membrane made of hydrophilic polysulfone, with a surface area of 0.5 m<sup>2</sup> and  
116 pore size of 10 kD (Alfa Laval). The second membrane was a multichannel tube made of  
117 ceramic with 19 channels. The surface area was 0.25 m<sup>2</sup>, pore size 0.1 μm (Orelis  
118 Environnement – KLEANSEP™ BW), and channel diameter 3.5 mm. The reactor was  
119 equipped with a pH probe and temperature sensor (JUMO 201020) in a second external  
120 recirculation loop with a sampling port.



121  
122 **Figure 1: Schematic of the pilot-scale anaerobic membrane bioreactor used in this**  
123 **study.**

124 The bioreactor was fed continuously with substrate in the reserve, which was kept cold  
125 by recirculation through the cold storage (4 °C) with a peristaltic pump (Watson Marlow  
126 Series 300). Overflow substrate was returned to the cold storage by gravity. The substrate



127 was kept cold to prevent degradation. The permeate flow rate was controlled with a  
128 peristaltic pump (MasterFlex – 7518-10) on the permeate side of the membrane, which  
129 controlled the feed flow rate by natural level adjustment. No sludge was discarded during  
130 the experiment except for sampling (SRT > 100 days).

131 Bioreactor mixing was ensured by continuous gas and liquid recirculation. The liquid  
132 recirculation flow rate in the membrane was 700 L/h. The liquid recirculation flow rate  
133 for the pH probe (JUMO 201020) and sludge sampling was 600 L/h. Gas recirculation  
134 occurred by cycles of 10 minutes' recirculation at 480 NL/h and 10 minutes without  
135 recirculation. The temperature was set at 37 °C with a water bath and coil in the  
136 bioreactor. No pH adjustment was performed in the bioreactor.

#### 137 2.1.2 Analytical procedure

138 The following parameters were recorded regularly during all operations: FOS/TAC was  
139 recorded daily by pH titration using 0.1 N H<sub>2</sub>SO<sub>4</sub>, performed according to the Nordmann  
140 method [11]. The buffer capacity (TAC) of the system was determined through titration  
141 of 20 mL of sample up to pH 5. The volatile organic acids (FOS) were then obtained after  
142 a second titration step between pH 5.0 and pH 4.4. The biogas composition was recorded  
143 daily using a Micro-GC 490 instrument from Agilent Technologies. The sample was  
144 introduced through a heated line at 100 °C. Two columns were used: a molecular sieve 5  
145 Å (MS-5A, 10 m) to separate O<sub>2</sub>, N<sub>2</sub>, and CH<sub>4</sub> and a PoraPLOT U (PPU, 10 m) to quantify  
146 CO<sub>2</sub>. Separation was achieved at 100 °C and 60 °C with backflushes of 4.7 and 16 s,  
147 respectively. Columns pressure was 29 psi. The carrier gas was helium, and the detector  
148 was a thermal conductivity detector (TCD). The total run took 80 s. Calibration was  
149 performed with standard gas from Air Liquide, diluted using flowmeters from

150 Bronkhorst. Quantification was achieved using the peak areas in external calibration; the  
151 concentrations ranged from 0.05% to 100%. The permeate weight was recorded daily  
152 with a scale (OHAUS-type DEFENDER 5000). The total volume of biogas was recorded  
153 daily using a RITTER drum-type gas meter (TG 0.5). Volatile fatty acids (VFA) were  
154 recorded 3 times a week by HPLC (Ultimate 3000 Thermo Fisher) equipped with an  
155 Aminex HPX-87H ( $7.8 \times 300$  mm,  $9 \mu\text{m}$ ) column at  $50 \text{ }^\circ\text{C}$  with UV detection. The mobile  
156 phase was  $8 \text{ mM H}_2\text{SO}_4$  with a flow rate of  $0.8 \text{ mL/min}$ , and  $20 \mu\text{L}$  of sample was injected  
157 for analysis. Acetic, butyric, and propionic acids were quantified. The chemical oxygen  
158 demand (COD) was recorded 3 times a week using Spectroquant® cell tests (1.14541)  
159 with a Spectroquant® Multy photometer. Mixed liquor suspended solids (MLSS) and  
160 mixed liquor volatile suspended solids (MLVSS) were recorded weekly. Two  
161 centrifugations of  $50 \text{ mL}$  of sample ( $4,500 \text{ g}$ ,  $15 \text{ min}$ ) were performed, followed by  
162 sequential drying at  $105 \text{ }^\circ\text{C}$  and  $550 \text{ }^\circ\text{C}$ .

### 163 2.1.3 Membrane fouling and cleaning

164 The membrane flux was measured regularly for the ceramic membrane by measuring the  
165 flow rate of permeate produced from wastewater after disconnecting the peristaltic pump.  
166 Thus, the pressure at the permeate side was assumed to be atmospheric pressure. The  
167 pressure at the retentate side was measured using a probe (Brukert 8316).

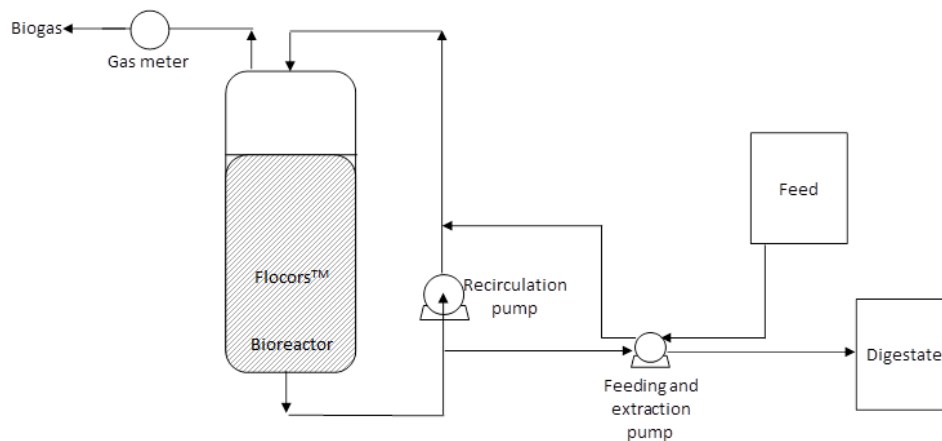
168 The membrane was cleaned whenever the flux decreased below  $12 \text{ L/h/m}^2$ . First, it was  
169 ensured that no clogging occurred in the membrane channels. Chemical cleaning was then  
170 conducted using  $\text{NaOH}$  solution ( $10 \text{ g/L}$ ) for 30 minutes at  $70 \text{ }^\circ\text{C}$  and rinsing with distilled  
171 water. Afterward,  $\text{HNO}_3$  solution ( $5 \text{ g/L}$ ) was used, and the membrane was submerged

172 for 20 minutes at 50 °C. The membrane was rinsed with water before being reused in the  
173 bioreactor.

## 174 2.2 APBR

### 175 2.2.1 Plant description

176 Fig. 2 shows the outline of the APBR. The reactor used was a CSTR-10S model provided  
177 by Bioprocess Control and adapted for APBR use. The total reactor volume was 12 L  
178 with a working volume of 10.6 L and an internal diameter of 0.2 m.



179

180 **Figure 2: Schematic of the anaerobic packed-bed reactor used in this study.**

181 The stirrer was removed, and the working volume of the reactor was filled with disordered  
182 plastic hollow cylinders with ribs (Flocors™). These had an average height of 30 mm, an  
183 internal diameter of 30 mm, and an external diameter of 35 mm. They had a porosity of  
184 95% and a specific area of 230 m<sup>2</sup>/m<sup>3</sup>. The reactor was filled with 0.55 L of Flocors™  
185 corresponding to a surface area of 0.127 m<sup>2</sup> available for the microorganisms. Flocors™  
186 was kept in the bioreactor by using a pipeline of lower diameter than the Flocors™ size.  
187 The reactor was equipped with a pH probe, a redox probe, and a temperature sensor.

188 Substrate feeding, stored at 4 °C, and digestate discharge were performed with a  
189 peristaltic pump (Gilson Minipuls Evolution®). Bioreactor mixing was ensured with an  
190 external loop with another peristaltic pump (Gilson Minipuls Evolution®) at a flow rate  
191 of 0.7 L/h. The temperature was set at 37 °C with a water bath and the bioreactor wall  
192 jacket.

### 193 2.2.2 Analytical procedure

194 During all operations, the consumed substrate weights and extracted digestate were  
195 recorded daily. The total biogas volume was measured daily using a volumetric gas  
196 flowmeter ( $\mu$ Flow, Bioprocess Control). The FOS/TAC was recorded 3 times a week and  
197 determined using a TitraLab AT1000 Series titrator from Hach. The non-purgeable  
198 organic carbon (NPOC) was recorded 3 times a week using a total organic carbon  
199 analyzer, TOC-L CSH/CSN, from Shimadzu after filtration of the sample with a  
200 0.2  $\mu$ m filter. For each batch of wastewater, the COD and NPOC were determined to  
201 correlate both values to compare the AnMBR and APBR. The COD and NPOC are  
202 proportional for a given wastewater type [12]. However, the carbon content in the APBR  
203 effluent could be underestimated while samples were filtered before NPOC analysis. The  
204 low suspended solids with the APBR suggest that underestimation was limited. The  
205 biogas composition was recorded 3 times a week using a GEMBIO biogas analyzer  
206 (Gruter & Marchand). The concentrations of CH<sub>4</sub> and CO<sub>2</sub> in the biogas were measured  
207 with an IR projector (2 Hz pulsations).

### 208 2.3 Sludge inoculum

209 The AnMBR was initially inoculated with sludge obtained from the 10-L APBR. It was  
210 composed of 0.38 g/L total suspended solids and 0.26 g/L volatile suspended solids.

211 The reactor was filled with 300 L inoculum by continuously filtering 150 L of inoculum  
 212 through the membrane to increase the biomass in the bioreactor, as digestate from the  
 213 APBR had low volatile suspended solids. Thus, the initial suspended solids in the pilot  
 214 bioreactor were doubled compared to the sludge.

215 No inoculum was used for the APBR. Consortia were developed from microorganisms  
 216 found in the wastewater used as substrate.

217 2.4 Experimental setup

218 During all experiments, the feed flow rate was regulated by following the  
 219 recommendations in Table 1, according to FOS/TAC values for the AnMBR and APBR.  
 220 The aim was to achieve optimal biogas production by balancing organic acid production  
 221 (FOS) with the buffer capacity of the system (TAC).

FOS/TAC ratio	Interpretation of FOS/TAC ratio	Feed flow rate control
>0.6	Highly excessive feed flow rate	Stop feeding
0.5–0.6	Excessive feed flow rate	Reduce feed flow rate by 10%
0.4–0.5	Plant is heavily loaded	Monitor the plant more closely
0.3–0.4	Biogas production at a maximum	Keep feed flow rate constant
0.2–0.3	Feed flow rate is too low	Increase feed flow rate by 10%
<0.2	Feed flow rate is far too low	Increase feed flow rate by 20%

222 **Table 1: Followed recommendations for feed flow rate during start-up period**

223 For the AnMBR, the experiment was conducted in three phases. The first was the study  
 224 of the start-up period. During the second phase, the substrate composition was changed  
 225 to evaluate the resilience performance of the AnMBR process instability. The third phase  
 226 was the process performance evaluation of the AnMBR running at a stable stage with a  
 227 high OLR.

228 The 10-L APBR ran for a few months at a stable stage with a performance similar to the  
229 industrial APBR used by the distillery (Cristanol, France) and was used as a model for  
230 evaluating the AnMBR performance.

## 231 2.5 Wastewater

232 The substrate used as nutrient feedstock was wastewater from the industrial distillery  
233 (Cristanol, France). It was a blend of evaporation condensate of the distillery from sugar  
234 beet and wheat ethanol production and cleaning water from the different distillery tanks.  
235 Suspended solids represented  $3 \pm 1$  g/L. A new batch of fresh wastewater was collected  
236 every 2 to 3 weeks. Variability was observed between each batch. Especially, higher COD  
237 was observed during sugar beet campaign periods. The pH was adjusted to  $7 \pm 0.2$  with  
238 potassium hydroxide before feeding. During the first phase, the bioreactor was fed with  
239 wastewater only (average COD:  $6.9 \pm 1.4$  g/L). During Phase 2, saccharose was added to  
240 the wastewater to evaluate the resilience performance of the AnMBR to instability.  
241 Saccharose represented half the COD during this phase (average COD:  $20.1 \pm 4.1$  g/L).  
242 Urea was added to the effluent with saccharose to maintain a constant C/N ratio, which  
243 was different for every wastewater batch. The third phase was performed during sugar  
244 beet campaign periods when wastewater had a high COD content (average COD:  $12.2 \pm$   
245  $0.8$  g/L); no saccharose was added in Phase 3.

## 246 2.6 Calculations

### 247 2.6.1 FOS/TAC

248 The FOS/TAC ratio was used to evaluate the biological stability of the process. The  
249 formula used for the FOS/TAC calculation was based on the Nordmann method [11],  
250 which empirically estimates FOS and TAC. The FOS value was expressed in mg/L of

251 acetic acid, and the TAC value was expressed in mg/L of calcium carbonate. Both values  
252 were estimated by titration of 20 mL sample with 0.1 N H<sub>2</sub>SO<sub>4</sub>.

$$253 \quad FOS = ((B \times 1.66) - 0.15) \times 500 \quad TAC = A \times 250$$

254 With A: Titration volume at pH 5 (mL) and B: Titration volume from pH 5 to pH 4.4  
255 (mL).

### 256 2.6.2 Food-to-microorganism ratio

257 The MLVSS was measured weekly. This was used as the microorganism concentration  
258 for calculating the F/M ratio. The MLVSS was shown to increase exponentially during  
259 Phase 1; thus, the MLVSS concentration was estimated for each day using the variation  
260 rate estimated for each week and the last MLVSS measurement.

$$261 \quad \textit{Variation rate} = \frac{\textit{Ln}(\textit{MLVSS at day } n) - \textit{Ln}(\textit{MLVSS at day } n + 7)}{7}$$

$$262 \quad \textit{MLVSS at day } n + i = \textit{Exp}(\textit{Ln}((\textit{MLVSS at day } n) + i \times \textit{Variation rate}))$$

263 The F/M ratio was calculated by dividing the OLR, expressed as g<sub>COD</sub>/L/day, and the  
264 microorganism concentration, expressed as g<sub>MLVSS</sub>/L<sub>reactor</sub>. The OLR was calculated from  
265 the feed flow rate and feed COD concentration. The average OLR of the last 2 days was  
266 used for the F/M ratio calculation.

267 The F/M ratio is a crucial parameter that significantly affects the process performance  
268 and membrane fouling in the AnMBR system. When this ratio is high, a high biomass  
269 yield is obtained, causing a low SRT, contributing to high sludge production [9]. It is also  
270 a source of biomass deflocculation and higher SMP and EPS production, which both  
271 increase membrane fouling [9]. Liu et al. [9] showed that a high F/M ratio of 3.6

272  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$  led to higher membrane fouling than  $0.1 \text{ g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$ , even if the  
273 MLVSS was higher when a low F/M ratio was applied. Membrane fouling is mainly  
274 explained by SMP and EPS concentrations in the AnMBR [7,9]. A high F/M ratio can  
275 also affect the balance between acidogenesis and methanogenesis, affecting the digestion  
276 performance. One of the main causes of anaerobic digestion failure is acidification due to  
277 overloading. The bacterial community of the acidogenesis step has a higher growth rate  
278 than the archaeal community of the methanogenesis step. Thus, a high F/M ratio risks  
279 VFA accumulation.

280 Thus, a low F/M ratio is preferred to stabilize the process and achieve a higher removal  
281 efficiency. However, if the F/M ratio is low, a higher MLVSS is required for the same  
282 OLR, increasing membrane fouling and the start-up time to reach the desired MLVSS.

### 283 2.6.3 Membrane flux

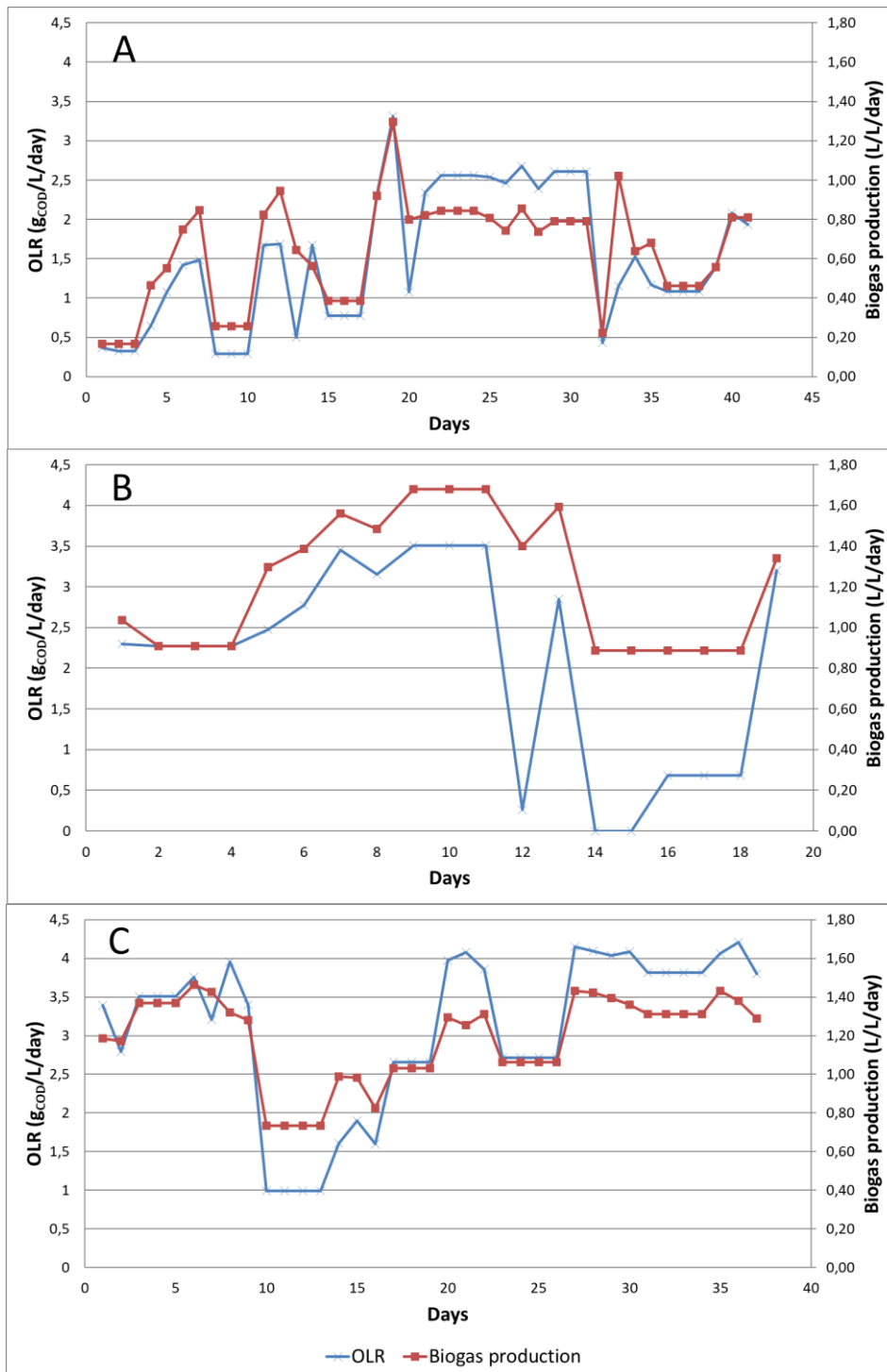
284 The membrane flux was measured in liters per hour per square meter ( $\text{L}/\text{h}/\text{m}^2$ ). The  
285 permeate pressure was assumed to be atmospheric pressure. The membrane flux was  
286 calculated as follows:

$$287 \quad \text{Membrane flux} = \text{Measured flowrate (L/h)} / \text{Membrane surface area (m}^2\text{)}$$

## 288 3 Results and discussion

### 289 3.1 AnMBR process performance





290

291 **Figure 3: Evolution of the OLR and biogas production of the AnMBR. A: Phase 1:**  
 292 **Start-up phase; B: Phase 2: Resilience trial by adding saccharose to the wastewater;**  
 293 **C: Phase 3: Wastewater during sugar beet campaign.**

### 294 3.1.1 Phase 1: Start-up phase

295 Fig. 3A shows the OLR ( $\text{g}_{\text{COD}}/\text{L}/\text{day}$ ) applied to the AnMBR and the biogas production  
296 ( $\text{L}_{\text{biogas}}/\text{L}_{\text{bioreactor}}/\text{day}$ ) during the first phase when the spiral-wound membrane was used.  
297 The experiment started with an OLR fixed at  $0.40 \text{ g}_{\text{COD}}/\text{L}/\text{day}$ . The OLR increased rapidly  
298 according to the FOS/TAC value. After 21 days, the process had been stabilized at a short  
299 HRT reaching 3.5 days, corresponding to an OLR of  $2.48 \text{ g}_{\text{COD}}/\text{L}/\text{day}$ , with low volatile  
300 fatty acids (VFA) in the permeate ( $<50 \text{ mg/L}$ ).

301 This result indicates that the AnMBR approach induces a short start-up period limited to  
302 21 days to reach a low HRT, while the start-up phase can take several months in a  
303 conventional anaerobic digester. Moreover, other studies showed that a short HRT can  
304 be reached in less than 2 weeks in an AnMBR [6]. The ability of the AnMBR to have  
305 complete control of the SRT is the main reason for both anaerobic digestion performance  
306 improvements.

### 307 3.1.2 Phase 2: Resilience trial

308 During the second phase, the resilience of the AnMBR was investigated by adding  
309 saccharose to the wastewater. Phase 2 was started with a similar OLR ( $2.30 \text{ g}_{\text{COD}}/\text{L}/\text{day}$ )  
310 to Phase 1 ( $2.48 \pm 0.09 \text{ g}_{\text{COD}}/\text{L}/\text{day}$ ), which was increased according to the FOS/TAC  
311 value. The first batch of wastewater in this phase had a COD concentration of  $8.02 \text{ g/L}$ ,  
312 and saccharose was added to double the COD concentration. Fig. 3B shows the OLR  
313 applied to the AnMBR and the biogas production during this phase. Until day 9, the  
314 FOS/TAC ratio remained low ( $0.19 \pm 0.02$ ) despite the change in organic carbon in the  
315 wastewater ( $\text{FOS/TAC} = 0.30 \pm 0.02$  in Phase 1). It was feasible to treat up to  $3.37 \pm 0.16$   
316  $\text{g}_{\text{COD}}/\text{L}/\text{day}$  with high removal efficiency ( $98.5\% \pm 0.3\%$ ).

317 On day 9, a new batch was used with a higher COD concentration (12.11 g/L), and  
318 saccharose was added to reach a COD concentration of 24.22 g/L. Easily fermentable  
319 sugar led to a rapid pH decrease in the cold storage, to pH 5 before feeding, inducing  
320 bioreactor acidification to pH 6.6. The pH decrease arose from the VFA accumulation,  
321 which reached 920 mg/L. This led to a FOS/TAC increase to 1.06 due to decreased  
322 methanogenic activity compared to the acetogenesis activity. The bacterial community of  
323 the acetogenesis step had a faster growth rate and activity than the archaeal community  
324 of the methanogenesis step. Thus, saccharose was quickly converted into acetic acid,  
325 which accumulated. Moreover, the archaeal community growth and activity were  
326 inhibited when the pH decreased to 6.6, further increasing the VFA accumulation. Thus,  
327 the feed pH was corrected, and the feed flow rate was decreased while the VFA  
328 concentration remained high (1,311 mg/L) 2 days later. The pH correction of the substrate  
329 was insufficient for VFA degradation. Feeding was then stopped for 2 days and slowly  
330 restarted. Four days after stopping the feed, no VFA was detected in the permeate (<50  
331 mg/L), and an OLR similar to that before acidification was applied (3.20 g<sub>COD</sub>/L/day).  
332 This result confirms that the AnMBR is highly resilient toward organic carbon variations  
333 since the FOS/TAC ratio remained low after adding saccharose for a similar OLR before  
334 day 9. Moreover, after the pH shock and VFA accumulation, recovery of the process  
335 stability and consumption of VFA was achieved within 4 days by reducing feeding. Thus,  
336 the AnMBR appears to be a promising technology to prevent biomass losses during  
337 uncontrolled acidification events due to its ability to rapidly resume control.  
338 Other literature results showed the high resilience of the AnMBR technology [5,13].  
339 Basset et al. [5] investigated the AnMBR performance under a fluctuating COD input.  
340 They showed that increasing the specific OLR to 0.47 g<sub>COD</sub>/g<sub>MLVSS</sub>/day led to a decrease

341 in COD removal from 95.7% to 63.6%. However, stable operation was recovered  
342 immediately by decreasing the specific OLR to 0.30 g<sub>COD</sub>/g<sub>MLVSS</sub>/day. In CSTR  
343 technology, if acidification occurs, biomass growth is stopped and lost, and a longer  
344 period is required to recover a proper balance.

### 345 3.1.3 Phase 3: Stable stage during sugar beet campaign

346 During the last period, the bioreactor was supplied with effluent without adding  
347 saccharose. Moreover, the bioreactor was equipped with a ceramic membrane. Fig. 3C  
348 shows the results for the OLR and biogas production in Phase 3.

349 After 10 days of operation, fouling caused a decrease in membrane flux. Consequently,  
350 an OLR decrease was measured. After membrane cleaning, the feed flow rate could be  
351 increased again according to the FOS/TAC value to increase the OLR. After a month, the  
352 process reached a stable stage at an HRT of 2.9 days and an OLR of 3.97 g<sub>COD</sub>/L/day with  
353 high COD removal (95.4%) and high digestion stability (FOS/TAC = 0.25).

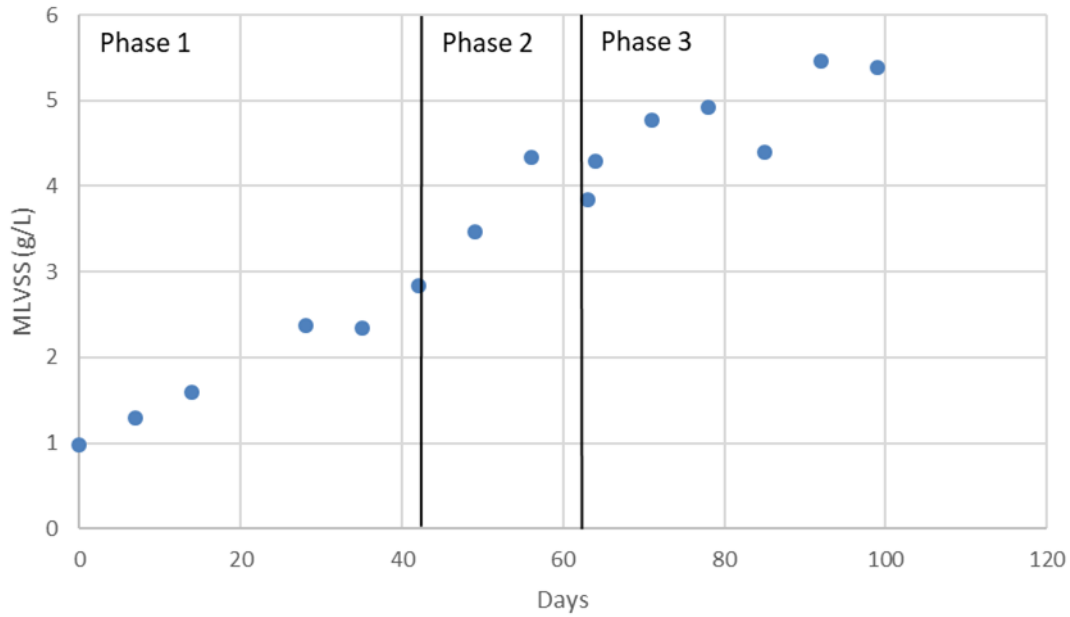
354 The AnMBR represents a physical barrier, inducing COD particle accumulation inside  
355 the bioreactor and releasing a permeate with a low COD content. The COD value in the  
356 AnMBR effluent was 0.53 g/L in Phase 3, while the total COD value was 30-fold higher  
357 inside the bioreactor. The high removal efficiency of the AnMBR was also observed in  
358 other studies [17,18,19]. In a study by Ozgun et al. [18], the total COD values reached  
359 approximately 1 to 2 g/L in the bioreactor, while the permeate COD remained low (0.10  
360 g/L).

361 However, the OLR reached in our process remained two to three fold lower compared to  
362 some studies using an AnMBR. Mnif et al. [14] reached an OLR of 8.0 g<sub>COD</sub>/L/day  
363 without affecting the reactor performance. Hu et al. [16] reached an OLR of 12.6

364  $\text{g}_{\text{COD}}/\text{L}/\text{day}$  with high COD removal efficiency (91.8%). This difference in the OLR can  
365 be explained by a higher COD concentration in the wastewater used in their study (up to  
366  $22 \text{ g}_{\text{COD}}/\text{L}$ ) and a higher surface membrane area:  $4.6 \text{ m}^2/\text{m}^3$ , 2.7 times larger than in our  
367 study. A higher membrane surface area leads to a higher flux and thus a higher feed flow  
368 rate.

#### 369 3.1.4 MLVSS evolution in AnMBR

370 Fig. 4 shows the evolution of MLVSS. The MLVSS is usually used to estimate the  
371 microorganism concentration [15]. The exponential variation in the MLVSS in Phase 1  
372 suggests exponential growth of microorganisms. In this phase, the average variation rate  
373 of MLVSS was  $0.031 \pm 0.005 \text{ day}^{-1}$ . The variation rate of MLVSS at the beginning of  
374 Phase 3 was lower:  $0.009 \pm 0.004 \text{ day}^{-1}$ . At the end of the experiment, the MLVSS was  
375 stabilized at  $5.6 \text{ g/L}$ . These results suggest that no apparent microorganism growth  
376 occurred (stationary phase of growth). The difference in the MLVSS variation rate might  
377 explain the slightly lower methane yield in Phase 1 ( $0.24 \text{ L}_{\text{CH}_4}/\text{g}_{\text{CODinput}}$ ) compared to  
378 Phase 3 ( $0.26 \text{ L}_{\text{CH}_4}/\text{g}_{\text{CODinput}}$ ), as a higher proportion of the COD input was used for  
379 microorganism growth in Phase 1, leading to higher accumulation of MLVSS.



380

381 **Figure 4: Evolution of MLVSS in AnMBR over time to evaluate the biomass growth.**

382 **Phase 1: Start-up phase; Phase 2: Resilience trial by adding saccharose to the**

383 **wastewater; Phase 3: Wastewater during sugar beet campaign.**

### 384 3.2 Performance comparison

385 Table 2 shows the AnMBR performance reached in recent studies using this technology

386 to treat real industrial wastewater. It also shows the performance at the stable stage

387 reached in Phase 3 of our study compared with the performance of the APBR treating the

388 same batch of wastewater.

#### 389 3.2.1 Performance comparison with recent studies using AnMBR

390 Other recent studies (2020–2021) used AnMBR technology to treat real industrial

391 wastewater. Table 2 shows their performances [20,21,22,23]. The performance achieved

392 in our study based on the OLR is higher than in most studies shown. Only one study [22]

393 showed a higher OLR (7.9 g<sub>COD</sub>/L/day). However, the OLR mainly depends on the

394 wastewater COD concentration and biodegradability. The relatively high COD

395 concentration of the distillery wastewater may explain the higher OLR. The higher OLR  
396 reached in our study led to higher biogas production (1.36 NL/L/day) than in most other  
397 studies. Only the study with a higher OLR showed higher biogas production (2.42  
398 NL/L/day) [22].

399 The HRT reached in our experiment (2.9 days) is similar to that in other studies (1.5 to 5  
400 days) despite the lower membrane surface area used ( $1.7 \text{ m}^2/\text{m}^3$ ), showing the advantage  
401 of the external tubular membrane, which has a higher flux in our study. However, this  
402 requires more energy for fouling control than for submerged membranes.

403 Moreover, the other studies confirm a high COD removal efficiency, which can be  
404 reached with the AnMBR (92% and 99%) [20,22]. Only one study [21] reported a lower  
405 COD removal efficiency (82%), explained by the complex structure and lower  
406 biodegradability of organic compounds in real textile wastewater. Additionally, other  
407 studies [20,22] report a high methane production of at least  $0.21 \text{ NL CH}_4/\text{gCOD}_{\text{input}}$ .

### 408 3.2.2 Performance comparison of AnMBR and APBR

#### 409 3.2.2.1 Start-up phase

410 In the APBR, the start-up period lasted 121 days to reach an HRT of 3 days, while only  
411 21 days were required for the AnMBR. This result suggests a shorter start-up phase in  
412 the AnMBR compared to the APBR. However, no inoculum was used in the APBR  
413 experiments, while the AnMBR was inoculated with a culture medium obtained from  
414 the APBR. Moreover, the short start-up period of the AnMBR may be due to the  
415 inoculum used, which was already adapted to the wastewater used in this study.

416 3.2.2.2 Stable stage

417 Table 2 shows the performance of the AnMBR at the stable stage reached in Phase 3  
418 compared with the performance of the APBR when treating the same batch of wastewater.

419 These results reveal the main differences between the AnMBR and APBR. A higher  
420 specific biogas production was obtained in the AnMBR,  $0.26 \text{ L}_{\text{CH}_4}/\text{g}_{\text{COD}_{\text{input}}}$ , while  $0.15$   
421  $\text{L}_{\text{CH}_4}/\text{g}_{\text{COD}_{\text{input}}}$  was obtained in the APBR for the same wastewater batch. Furthermore, the  
422 COD removal efficiency was higher in the AnMBR than in the APBR, and the VFA (eq  
423  $\text{CH}_3\text{COOH}$ ) content was lower in the AnMBR. Moreover, the lower FOS/TAC ratio  
424 provided higher stability in the AnMBR. Despite the scale difference of the two  
425 bioreactors, the performance differences can mainly be attributed to the technology used  
426 since the 10-L APBR is a model of the industrial-scale anaerobic bioreactor (Cristanol,  
427 France).

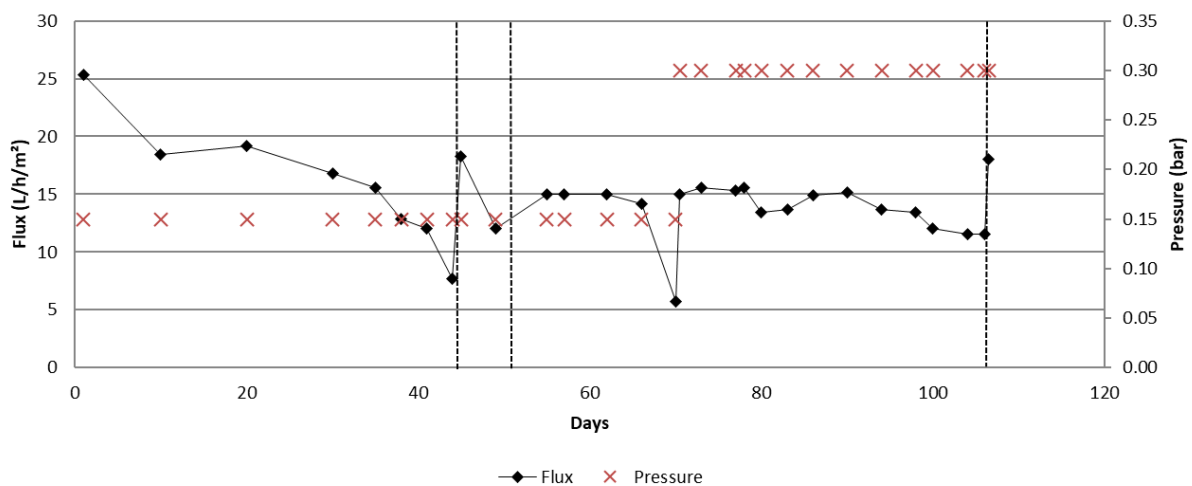
428 The high COD removal efficiency of the AnMBR resulted from a higher retention time  
429 of suspended COD particles, which are trapped in the bioreactor and can thus be  
430 consumed by microorganisms. Inside the AnMBR, suspended solid represented 77% of  
431 the total COD, which is consistent with the scientific literature [8,18,24]. These  
432 suspended particles were not retained in the APBR. This led to a greater methane yield  
433 for the AnMBR ( $0.26 \text{ L}_{\text{CH}_4}/\text{g}_{\text{COD}_{\text{input}}}$ ) than that obtained in the APBR ( $0.15 \text{ L}_{\text{CH}_4}/\text{g}_{\text{COD}_{\text{input}}}$ ).  
434 Thus, compared to the APBR process, the AnMBR process resulted in a 1.7-fold greater  
435 methane yield and biogas production. This result shows better use of the COD in the  
436 AnMBR compared to the APBR. This difference is not reflected in the COD removal  
437 efficiency since the APBR effluent COD measurement did not consider suspended solids.



438 The higher soluble COD removal efficiency observed in the AnMBR (95.4%) compared  
 439 to the APBR (93.0%) can be explained by the better hydraulic conditions in the  
 440 AnMBR compared to the APBR. A high biomass concentration with free cells in the  
 441 AnMBR led to better contact between microorganisms and the COD. Conversely, the  
 442 APBR struggles to maintain intimate and prolonged contact between the  
 443 microorganisms and COD. Moreover, clogging can occur [3]. Effluents of the anaerobic  
 444 wastewater treatment plant still require further aerobic treatment to reach the required  
 445 quality for disposal in a river or spreading in the environment. With a higher COD  
 446 removal in the AnMBR, less energy will be required for the aerobic treatment.

447 **3.3 Membrane fouling**

448 Although the AnMBR showed much promise for wastewater treatment and biogas  
 449 production, membrane fouling remains a major limiting factor regarding industrial  
 450 applications. In this study, the ceramic membrane lifespan was investigated to estimate  
 451 the cleaning frequency and flux recovery.



452 **Figure 5: Evolution of the ceramic membrane flux in AnMBR to evaluate the**  
 453 **filtration performance of the membrane through the experiment. The dotted lines**  
 454 **represent membrane cleaning days.**

455 Fig. 5 shows the membrane flux throughout the AnMBR experimental operation using a  
456 ceramic membrane. The initial membrane flux was 25 L/h/m<sup>2</sup>. This value quickly  
457 decreased to 19 L/h/m<sup>2</sup> and remained between 15 and 20 L/h/m<sup>2</sup> for more than 30 days.  
458 Beyond this, a faster membrane flux decrease was observed from 15 to 7.7 L/h/m<sup>2</sup> in less  
459 than 10 days, probably caused by cake formation at the membrane surface and clogged  
460 pores.

461 First, membrane chemical cleaning was performed using cold acid and base solutions  
462 after 44 days of operation at low transmembrane pressure (TMP), under 0.15 bar<sub>g</sub>. This  
463 led to flux recovery up to 18 L/h/m<sup>2</sup> (72% recovery compared to the new membrane).  
464 However, after only 4 days, the flux decreased to 12 L/h/m<sup>2</sup>, showing that a cold solution  
465 was inefficient for membrane cleaning. A second cleaning strategy was applied. The  
466 ceramic membrane was cleaned using base and acid solutions at 70 °C and 50 °C,  
467 respectively, leading to 60% recovery compared to the new membrane. The membrane  
468 could then be used for 20 days at low TMP (under 0.15 bar<sub>g</sub>). However, on day 70, the  
469 flux dropped to 6 L/h/m<sup>2</sup>. Thus, the TMP was increased. It was still possible to use the  
470 membrane for 36 days at 0.30 bar<sub>g</sub>. In total, the ceramic membrane was used for almost  
471 2 months without cleaning. The flux after cleaning was 18 L/h/m<sup>2</sup>, showing a high  
472 recovery of the flux. This showed that increasing the pressure did not cause irreversible  
473 fouling, suggesting that the pressure can be increased to maintain the flux for months.  
474 However, maintaining a moderate TMP for as long as possible is recommended to lower  
475 pore clogging.

476 A proper membrane cleaning procedure and bioreactor monitoring allowed the AnMBR  
477 to be run for almost 2 months (56 days) without cleaning. However, this performance was  
478 only possible with a high recirculation flow rate applied to the membrane (700 L/h). The

479 reduction of the recirculation rate to 300 L/h led to a 70% flux decrease within 2 days,  
480 leading to inability to maintain the desired HRT. This tremendous flux decrease was  
481 expected since this lower recirculation rate corresponded to a liquid speed of 0.5 m/s,  
482 which is deemed extremely low for an ultrafiltration membrane. A liquid speed of at least  
483 2 m/s is recommended for an ultrafiltration membrane [25]. Nevertheless, this high  
484 recirculation rate would lead to high operation costs on an industrial scale for anaerobic  
485 treatment.

486 In our study, no sludge discharge was applied during 8 months of operation. After the two  
487 cleaning cycle intervals described previously, the membrane had been used for 4 months  
488 without cleaning (data not shown). During this period, the membrane flux decreased  
489 progressively to 10 L/h/m<sup>2</sup> with a TMP enhanced to 1 bar. Moreover, channel clogging  
490 occurred. The soluble COD concentration inside the bioreactor was eightfold higher than  
491 the COD of the permeate. This showed the accumulation of organic content in the  
492 bioreactor after 8 months of operation without sludge discharge. This organic matter  
493 probably comprised SMP and EPS, which are known to cause membrane fouling [7].

494 Some studies have shown that a long SRT led to serious membrane fouling for long-term  
495 use [26]. A high SRT led to accumulation of organic content and increased the SMP  
496 production by microorganisms [10]. This suggests faster membrane fouling because of  
497 biomass attached to the membrane surface or trapped in pores. Huang et al. [27] almost  
498 doubled their cleaning interval by decreasing the SRT from 90 to 60 days; membrane  
499 cleaning intervals were changed from 25 to 40 days.

500 Due to the energy needed for the recirculation loop in the external configuration, it is  
501 necessary to investigate other filtration processes, such as submerged membranes, as they

502 are considered more economically feasible on an industrial scale. Moreover, it is  
503 necessary to investigate the optimal SRT to prevent membrane fouling.

#### 504 3.4 F/M ratio

505 In our study, the OLR was applied based on the FOS/TAC ratio. The MLVSS increase  
506 was observed over time, showing the microorganism growth. Thus, the OLR was also  
507 increased because the higher the microorganism concentration, the higher the COD that  
508 can be treated. The OLR and MLVSS values were used to follow the evolution of the  
509 F/M ratio over time.

510 During the two stable stages in Phases 1 and 3, two different F/M ratios were observed,  
511 respectively: 1.05 and 0.74  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$ . In Phase 2, the F/M ratio was 0.96  
512  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$ . In the first phase, anaerobic digestion performances were similar  
513 despite the higher F/M ratio.

514 At the end of the experiment, the MLVSS was stabilized at 5.6 g/L, showing a halt in  
515 microorganism growth, indicating that the F/M ratio of 0.7  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$  was too low  
516 to support the biomass growth. Mnif et al. [14] observed the same stabilization of the  
517 biomass concentration at 2.5 g/L of volatile suspended solids under their conditions with  
518 a high F/M ratio (2.5  $\text{g}_{\text{COD}}/\text{g}_{\text{VSS}}/\text{day}$ ).

519 The F/M ratio decrease over time for the same bioprocess stabilization suggests that the  
520 biomass activity with a long SRT (>100 days) tends to decrease, probably because of  
521 accumulation of dead microorganisms or suspended organic matter that did not contain  
522 microorganisms. This shows that applying a shorter SRT is necessary for process  
523 performance and membrane fouling. Working with an F/M ratio as high as 1.05  
524  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$  in Phase 1 did not affect the process performance. Thus, the SRT must  
525 be controlled to maintain this ratio around 1  $\text{g}_{\text{COD}}/\text{g}_{\text{MLVSS}}/\text{day}$  for a constant OLR. A ratio

526 of 1.05 g<sub>COD</sub>/g<sub>M<sub>LVSS</sub></sub>/day was relatively high compared to other studies. Basset et al. [5]  
527 reported an F/M of 0.30 g<sub>COD</sub>/g<sub>M<sub>LVSS</sub></sub>/day and Kunacheva et al. [28] used an F/M of 0.5  
528 g<sub>COD</sub>/g<sub>M<sub>LVSS</sub></sub>/day. Our results showed a relatively high biological activity of the  
529 consortium compared to other studies. However, the ratio in our study was determined  
530 during the start-up phase. Further investigations with a stabilized F/M ratio during  
531 multiple cleaning cycle intervals are required to confirm this ratio, which depends on the  
532 OLR, HRT, and membrane characteristics and surface.

#### 533 4 Conclusion

534 The results obtained in this study confirm that the membrane bioreactor is a promising  
535 technology for wastewater treatment by anaerobic digestion. Excellent digestion  
536 performances were reached: a high methane production of 0.26 L<sub>CH<sub>4</sub></sub>/g<sub>CODinput</sub>, a high  
537 COD removal efficiency of 96.9%, and a short start-up period (21 days). All these  
538 performances exceeded the performances of the APBR, which is the currently used  
539 industrial-scale process for this wastewater.

540 Moreover, a long interval between cleaning of at least 44 days was reached, confirming  
541 its interest for industrial-scale development. However, this technology still requires  
542 improvement for membrane fouling control, as a high energy demand was observed in  
543 our study for membrane filtration. Further study with a submerged membrane could  
544 limit the energy demand for this process.

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Wastewater	Configuration	Membrane surface (m <sup>2</sup> /m <sup>3</sup> )	HRT (days)	Temperature	pH	Wastewater COD content (g <sub>COD</sub> /L)	OLR (g <sub>COD</sub> /L/day)	Biogas production (NL/L/day)	Methane production (NL CH <sub>4</sub> /g <sub>COD</sub> input)	CH <sub>4</sub> (%) – CO <sub>2</sub> (%)	COD Removal efficiency (%)	VFA (eq CH <sub>3</sub> COOH mg/L)	FOS/TAC	Reference
Malting plant Wastewater	Submerged flat sheet	6 *	1.5	18 °C	7.4	3.2	2.1	0.6 *	0.21	80.95 – 13.51	92	-	-	[20]
Textile effluent	Submerged flat sheet	2.9	2	30 °C–40 °C	-	0.7–1.2	0.20–0.35	-	-	-	82	-	-	[21]
Confectionery wastewater	Submerged flat sheet	4	4.1–2.3	35 °C	7	18–18.9	4.4–7.9	1.45–2.42	0.31–0.26	-	99	-	-	[22]
Brewing wastewater	Submerged flat sheet	7	5	37 °C	7.3–7.7	4.9	0.98	0.213 LCH <sub>4</sub> /L/day	-	-	-	-	-	[23]
Distillery Wastewater	External tubular ceramic	1.7	2.9 ± 0.1	37 °C	7.36 ± 0.09	12.2	3.97 ± 0.15	1.36 ± 0.05	0.26 ± 0.01	74.7 ± 0.4 – 25.3 ± 0.4	95.4 ± 0.7	476 ± 40	0.25 ± 0.03	This study
Distillery Wastewater	APBR	na	2.8 ± 0.4	37 °C	7.50 ± 0.19	12.2	4.48 ± 0.94	0.94 ± 0.23	0.15 ± 0.04	71.6 ± 1.0 – 28.4 ± 1.0	93.0 ± 2.8	826 ± 40	0.51 ± 0.02	This study

652 Table 2: Performance of AnMBR compared to other recent studies on AnMBR treating real industrial wastewater (2020–2021) and to  
653 APBR technology. na: not applicable. \*Volume of the reactor was estimated from membrane flux and HRT