Submerged anaerobic membrane bioreactor (SAnMBR) performance on sewage treatment: removal efficiencies, biogas production and membrane fouling

Rong Chen, Yulun Nie, Jiayuan Ji, Tetsuya Utashiro, Qian Li, Daisuke Komori and Yu-You Li

ABSTRACT

A submerged anaerobic membrane reactor (SAnMBR) was employed for comprehensive evaluation of sewage treatment at 25°C and its performance in removal efficiency, biogas production and membrane fouling. Average 89% methanogenic degradation efficiency as well as 90%, 94% and 96% removal of total chemical oxygen demand (TCOD), biochemical oxygen demand (BOD) and nonionic surfactant were obtained, while nitrogen and phosphorus were only subjected to small removals. Results suggest that SAnMBRs can effectively decouple organic degradation and nutrients disposal, and reserve all the nitrogen and phosphorus in the effluent for further possible recovery. Small biomass yields of 0.11 g mixed liquor volatile suspended solids (MLVSS)/gCOD were achieved, coupled to excellent methane production efficiencies of 0.338 NLCH4/gCOD, making SAnMBR an attractive technology characterized by low excess sludge production and high bioenergy recovery. Batch tests revealed the SAnMBR appeared to have the potential to bear a high food-to-microorganism ratio (F/M) of 1.54 gCOD/gMLVSS without any inhibition effect, and maximum methane production rate occurred at F/M 0.7 gCOD/gMLVSS. Pore blocking dominated the membrane fouling behaviour at a relative long hydraulic retention time (HRT), i.e. >12 hours, while cake layer dominated significantly at shorter HRTs, i.e. <8 hours.

Key words | biomass yield, food to microorganism, fouling behaviour, methanogenic degradation, SAnMBR, sewage

INTRODUCTION

Sewage is the most abundant type of wastewater and a valuable resource containing water, nutrients and energy, worthy of recovery and reuse (Prieto et al. 2013). If recoverable, sewage has the potential to become a net supplier of renewable energy and reclaimed water (Ozgun et al. 2013). Consequently the selection of an appropriate technology that can convert sewage into high level renewable energy and high quality reclaimed water is very important.

Anaerobic digestion has drawn considerable attention for its ability to convert chemically bound energy in the organic pollutants to useful energy, namely biogas (Shizas & Bagley 2004). Basically anaerobic digestion involves four key steps including hydrolysis, acidogenesis, acetogenesis and methanogenesis, which are carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenotrophic methanogens and aceticlastic methanogens (Batstone et al. 2002). Hydrolysis and
methanoogenesis are widely accepted to be the two rate-limiting steps of anaerobic digestion, where hydrolysis, mainly depending on enzymatic activity, is normally rate-limiting if the substrate is in particulate form (Vavilin et al. 1996), and methanoogenesis, depending on slow-growth methanogens, is the most crucial stage in biogas production and also very sensitive to operational parameters (Finney & Evans 1975). For this reason, compared with activated sludge, anaerobic sludge tends to appear slow growing, especially under the condition of low organic strength feeding, which may become a main obstacle to applying anaerobic digestion directly to sewage treatment. If membrane technology is involved, the retention of slow growing anaerobic biomass is successfully realized by the membrane solid-liquid separation. Anaerobic membrane reactors combine both anaerobic digestion with membrane separation, thus decoupling the biomass retention of the slow-growth methanogens from the shortened hydraulic retention time (HRT), an essential process to feed appropriate organic loading rates (OLR) in case a reactor is supplied by low-strength substrates (Lin et al. 2013). For this reason, short HRT coupled with long solid retention time (SRT) in anaerobic membrane reactors can achieve high biomass concentrations, which are quite favourable for high efficiency of anaerobic digestion and biogas production (Huang et al. 2011).

Up to now, most of the studies related to anaerobic membrane reactors have focused on high-strength organic wastewater, while a few are related to low-strength wastewater like sewage. Normally, suspended solids (SS), organic matter, nitrogen and phosphorus are the main concerns in terms of common pollutants in sewage, and the treatment performances for these pollutants appear to have different fates in an anaerobic membrane reactor. Usually SS can be removed to a large extent by microfiltration membranes, which are commonly used in a membrane bioreactor. The organic matter in sewage is always composed of complex polymers, which can be categorized as carbohydrates, fats and proteins, and their removal efficiencies are highly dependent on the anaerobic digestion ability. Anaerobic membrane bioreactors are supposed to retain sufficient slow-growth methanogens and prevent biomass being washed out to deal with organic complex polymers in sewage. In contrast, nitrogen and phosphorus in sewage would become challenges for individual anaerobic digestion, and additional or coupled processes may be necessary to enhance their removal efficiency or recover them.

Anaerobic membrane reactors are generally implemented based on two configurations: external-stream and submerged. Commonly, the external configuration expresses the advantages of easier membrane cleaning/replacement and higher flux but higher energy consumption, while the submerged configuration, called the submerged anaerobic membrane reactor (SAnMBR), offers an obvious advantage of much lower energy consumption by placing the membrane into the mixed liquid, which avoids the transfer from the anaerobic digestion unit to the membrane filtration unit by pumps. The objective of this study is to provide a comprehensive understanding of SAnMBR performance in removal efficiency, biogas production and membrane fouling towards sewage treatment.

MATERIAL AND METHODS

Reactor set up

Figure 1 shows the laboratory-scale reactor set up. The working liquid volume of the SAnMBR was 6 L and it was operated at 25 ± 1°C. The flat sheet membrane module (Kubota Membrane Cartridge, Japan) was equipped inside the reactor and its characteristics were as follows: mean pore size, 0.2 μm; filtration area, 0.116 m²; material, chlorinated polyethylene. The produced biogas was continuously returned by a gas pump (APN-085 LV-1, Iwaki, Japan) with a flow rate of 5 L/min, to scour the membrane surface to prevent solids fouling, through a bubble diffuser equipped at the bottom of the membrane module. Filtration is driven by applying negative pressure to the permeate side of the membrane with a peristaltic pump (Model 7518-10, Cole-Parmer, USA). Trans-membrane pressure (TMP) was measured by a digital pressure meter (Keyence, AP-V85) installed between the membrane and the permeate pump. A gas holder was used to measure biogas production.

Seed sludge and feed sewage

The SAnMBR was inoculated with anaerobic digested sludge from the wastewater treatment plant in Sendai, Japan. The main characteristics of the sewage fed to SAnMBR are listed in Table 1, which was synthesized by 150 mg/L toilet paper, 64.6 mg/L beef extract, 56.8 mg/L peptone, 56.8 mg/L yeast extract, 88.2 mg/L NaHCO₃, 0.125 mg/L CuSO₄·5H₂O, 88.2 mg/L H₃BO₃. The recipe for synthetic sewage referred to a previous published research (Chen et al. 2017).

Table 1. Synthetic sewage.

<table>
<thead>
<tr>
<th>Component</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toilet paper</td>
<td>150 mg/L</td>
</tr>
<tr>
<td>Beef extract</td>
<td>64.6 mg/L</td>
</tr>
<tr>
<td>Peptone</td>
<td>56.8 mg/L</td>
</tr>
<tr>
<td>Yeast extract</td>
<td>56.8 mg/L</td>
</tr>
<tr>
<td>NaHCO₃</td>
<td>88.2 mg/L</td>
</tr>
<tr>
<td>H₃BO₃</td>
<td>0.125 mg/L</td>
</tr>
</tbody>
</table>

Figure 1: Laboratory-scale reactor set up. The working liquid volume of the SAnMBR was 6 L and it was operated at 25 ± 1°C. The flat sheet membrane module (Kubota Membrane Cartridge, Japan) was equipped inside the reactor and its characteristics were as follows: mean pore size, 0.2 μm; filtration area, 0.116 m²; material, chlorinated polyethylene. The produced biogas was continuously returned by a gas pump (APN-085 LV-1, Iwaki, Japan) with a flow rate of 5 L/min, to scour the membrane surface to prevent solids fouling, through a bubble diffuser equipped at the bottom of the membrane module. Filtration is driven by applying negative pressure to the permeate side of the membrane with a peristaltic pump (Model 7518-10, Cole-Parmer, USA). Trans-membrane pressure (TMP) was measured by a digital pressure meter (Keyence, AP-V85) installed between the membrane and the permeate pump. A gas holder was used to measure biogas production.

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<td>88.2 mg/L</td>
</tr>
<tr>
<td>H₃BO₃</td>
<td>0.125 mg/L</td>
</tr>
</tbody>
</table>
Operating conditions

Table 2 shows the operating conditions. The HRT was decreased from 48 to 8 hours by adjusting the suction cycle of the permeate pump. The pH was not controlled but ranged from 6.8 to 7.5, which imposed no negative effect on anaerobic degradation. When TMP reached 20 kPa, the membrane was taken out of the SAnMBR for cleaning. Firstly, tap water was used to wash the membrane surface, then 0.1% NaClO solution and 10 g/L citric acid solution were used to soak the membrane sequentially. Filtration test was carried out for the membrane before any cleaning, and after water cleaning, NaClO cleaning and citric acid cleaning. Tap water permeability was tested. No excess sludge was regularly discharged except for periodical sampling (20 ml per week) for analysis, mainly due to small mixed liquor volatile suspended solids (MLVSS) yields, determined as 0.06–0.09 over the whole duration. Thus the SRT can be considered to approach infinity in this study.

Batch test

Batch tests of methane production were carried out in 120 mL serum bottles placed and incubated in a water bath at 25 ± 1°C. For each serum bottle, 40 mL of sludge taken from the SAnMBR were inoculated, in which the MLVSS concentration was determined as 10 g/L. Then a synthetic solution, with a volume of 2.4, 6.4, 8.0, 10.4, 14.4 and 17.6 ml respectively, was added to realize different food-to-microorganism ratios (F/M) of 0.21, 0.56, 0.70, 0.91, 1.26 and 1.54 gCOD/g MLVSS. The synthetic solution, with a chemical oxygen demand (COD) concentration of 35 g/L, was prepared according to the chemical

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Concentrations (mg/L)</th>
</tr>
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<tbody>
<tr>
<td>SS</td>
<td>150 ± 50</td>
</tr>
<tr>
<td>TCOD</td>
<td>700 ± 100</td>
</tr>
<tr>
<td>SCOD</td>
<td>500 ± 100</td>
</tr>
<tr>
<td>BOD</td>
<td>400 ± 50</td>
</tr>
<tr>
<td>NH₄-N</td>
<td>40 ± 15</td>
</tr>
<tr>
<td>TP</td>
<td>5.0 ± 2.0</td>
</tr>
<tr>
<td>Anionic surfactant (LAS)</td>
<td>2.5 ± 1.0</td>
</tr>
<tr>
<td>Nonionic surfactant (AE)</td>
<td>5.0 ± 2.0</td>
</tr>
<tr>
<td>Trace elements</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>HRT (hours)</th>
<th>48</th>
<th>24</th>
<th>16</th>
<th>12</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operating duration (days)</td>
<td>1–30</td>
<td>31–70</td>
<td>71–90</td>
<td>91–121</td>
<td>121–151</td>
</tr>
<tr>
<td>Flux (L/m²·h)</td>
<td>1.08</td>
<td>2.15</td>
<td>3.23</td>
<td>4.31</td>
<td>6.47</td>
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<tr>
<td>Suction cycle</td>
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<td>1 min on</td>
<td>1 min on</td>
<td>1 min on</td>
</tr>
<tr>
<td></td>
<td>9 min off</td>
<td>5 min off</td>
<td>3 min off</td>
<td>2 min off</td>
<td>1 min off</td>
</tr>
</tbody>
</table>
composition in the recipe of synthetic sewage. After that, each bottle was set at the same liquid volume of 80 ml by adding nutrient solution, which was boiled for 2 h to remove any dissolved oxygen and cooled down to room temperature under a nitrogen atmosphere before use. The headspace of all bottles was purged with nitrogen gas for 2 min to remove oxygen. After each bottle reached the set temperature in the water bath, the headspace was vented using a syringe to release the pressure caused by the thermal expansion. Biogas production and composition were measured regularly according to the biogas volume, and normalized to the value at standard state. Each sample was conducted in two replicates to ensure its reliability. The obtained results were simulated by the modified Gompertz equation (Li et al. 2015):

\[
P = P_0 \cdot \exp \left\{ -\exp \left[ \frac{R_{\text{max}} \cdot e}{P_0} \cdot (t_0 - t) + 1 \right] \right\}
\]

where \(P\) is cumulative methane production (CMP) (mL), \(P_0\) is methane production potential (mL), \(R_{\text{max}}\) is the maximum methane production rate (mL/d), and \(t_0\) is the lag time (days). The constants \(P_0\) and \(t_0\) were estimated by a nonlinear fitting program using an Origin Software.

**Analytic methods**

Total COD (TCOD), biochemical oxygen demand (BOD), SS, total phosphorus (TP), MLSS and MLVSS concentrations were determined as per Standard Methods (APHA/AWWA/WEF 2005). To measure soluble COD (SCOD) in the influent, the sample was filtered through a 0.45 μm syringe filter. The NH_4^+-N concentration was analysed by a capillary electrophoresis device (Agilent 7100) after being filtered through a 0.45 μm syringe filter. The determination of LAS and AE concentrations was conducted with iron (II) chelate (1,10-phenanthroline) and a coloured iodine-micelle complex by absorbometry, respectively (Ross & Olivier 1959). Removal efficiencies for all pollutants were calculated by the differences of influent and effluent concentrations, and that of COD in particular was based on TCOD. The composition of N\(_2\), CH\(_4\) and CO\(_2\) in the collected biogas was measured using a gas chromatograph (Shimadzu, GC-8A, Japan) equipped with a thermal conductivity detector. Dissolved methane in the permeate was determined by the headspace technique (Hatamoto et al. 2010). All the measurements of the methane amount were normalized to a standard state (0 °C, 1 atm) by using Equation (2).

After that, for the purpose of the COD mass balance, the normalized CH\(_4\) amount can be further converted to a COD amount by 1 gCOD = 0.35 NLCH\(_4\) derived from the stoichiometric reaction: \(\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}\).

\[
M = \frac{BP \cdot MC}{273.15 + T}
\]

where \(M\) is the normalized CH\(_4\) amount (NL CH\(_4\)), \(BP\) is the biogas production (L), \(MC\) is the methane composition in biogas (%), and \(T\) is the biogas temperature (°C).

**RESULTS AND DISCUSSION**

**Removal efficiencies**

Figure 2 shows the concentrations of COD, BOD, SS, NH\(_4^+\)-N, TP and surfactant in the influent and effluent. It showed that almost all the removal efficiencies were similar under all the HRTs over the whole process. Most of the effluent CODs were less than 40 mg/L and the average removal efficiencies of TCOD and BOD were higher than 90% (Figure 2(b) and 2(c)), which meant the organic matter was highly degraded. Nearly 100% removal efficiencies for SS were achieved due to membrane microfiltration, because almost no SS can be detected most of the time (Figure 2(d)). The results obtained indicated that SAnMBRs treating sewage can achieve high organic anaerobic digestion efficiency and qualified effluent with low COD concentrations at 25 °C. By contrast, NH\(_4^+\)-N concentration in the permeate averaged 58% higher than that in the influent (Figure 2(e)), and this phenomenon was attributed to the anaerobic ammonization converting organic nitrogen in protein and urea to inorganic NH\(_4^+\)-N. TP concentration appeared to show no significant difference between the influent and permeate (Figure 2(f)). These results were also observed in related research (Gouveia et al. 2015). Low removal efficiency of phosphorus and increased effluent ammonia nitrogen in the SAnMBR might suggest that the individual anaerobic membrane reactor is unsuitable to be applied to treat sewage if the effluent is finally discharged into a receiving water body, which may cause algal blooms due to the sufficient nutrient inflows. But on the other hand, it can effectively separate organic matter from nutrients in sewage and keep all the influent nitrogen and phosphorus to the effluent, which will be beneficial for nutrient recovery, as well as being used for agricultural or forest irrigation. Regarding the surfactant pollutants, AE was largely removed and the efficiencies were stably more than 95%, while LAS was only removed by on average 44% and its removal...
efficiencies seemed to decrease with HRT shortening. Wagener and Schink (Wagener & Schink 1987), working with anaerobic degradation of nonionic and anionic surfactants, reported nonionic surfactants like AE enhanced methane production and most of them were converted to methane; in contrast, anionic surfactants like LAS always impaired methane formation, associated with accumulation of acetate.

Figure 3 shows the sludge/biomass growth in the SAnMBR. The following results were found: (1) the MLVSS concentration kept very slowly growing from HRT 48 h to 16 h, possibly due to the relatively low OLR feeding at these long HRTs, while the MLSS and MLVSS presented an observed increase from HRT 16 h to 12 h and then to 8 h, and reached 9.3 and 8.1 g/L respectively at HRT 8 h, mainly due to the enhanced biomass yield at a short HRT, namely high OLR feeding; (2) the ratios of MLVSS/MLSS were maintained in a stable range of on average 0.84–0.91 over the whole experimental period, which may indicate there was no obvious accumulation of solid organic or inorganic matter in the bioreactor.

The biomass concentration in a biological reactor was deemed to depend on both microbial growth and endogenous decay. For this reason, the following kinetic model (Equations (3)–(5)) provided by Horan (Horan 1989; Huang et al. 2001) can be used to determine biomass yields under different HRTs. The calculation of Equations (3) and (4) can be implemented directly from the experimental

![Figure 2](https://iwaponline.com/wst/article-pdf/76/6/1308/449165/wst076061308.pdf)  
**Figure 2** | The SAnMBR removal efficiencies: (a) OLR; (b) COD; (c) BOD; (d) SS; (e) NH\(_4^+\); (f) TP; (g) AE; and (h) LAS.

![Figure 3](https://iwaponline.com/wst/article-pdf/76/6/1308/449165/wst076061308.pdf)  
**Figure 3** | MLSS and MLVSS concentrations at various operating HRTs.
data, while that of Equation (5), based on the results of Equations (3) and (4), was to obtain the theoretical biomass yield and the endogenous decay coefficient.

\[
Y = \frac{R_{MLVSS}}{R_{COD}}
\]  

(3)

\[
\mu = \frac{R_{MLVSS}}{X_{MLVSS}}
\]  

(4)

\[
\frac{1}{Y} = \frac{1}{Y_0} + \frac{b}{Y_0} \frac{1}{\mu}
\]  

(5)

where \( Y \) is the observed MLVSS yield (gMLVSS/gCOD), \( R_{MLVSS} \) is the MLVSS growth rate (gMLVSS/L/d), \( R_{COD} \) is the organic removal rate (gCOD/L/d), \( \mu \) is the specific MLVSS growth rate (d/COD), \( X_{MLVSS} \) is the MLVSS concentration in the SAnMBR (g/L), \( Y_0 \) is the theoretical MLVSS yield (gMLVSS/gCOD), and \( b \) is the endogenous decay coefficient (d

Table 3 shows the results of the calculation. It can be found that the biomass yields (\( Y \)) at various operating HRTs ranged from 0.06 to 0.09 gMLVSS/gCOD, which was within an experiential range of 0.03–0.18 gMLVSS/gCOD in anaerobic digestion processes (Henze 2008). The theoretical biomass yield (\( Y_0 \)) and endogenous decay coefficient (\( b \)) were calculated as 0.11 gMLVSS/gCOD and 0.004 d

Table 3 | Biomass yields at various operating HRTs

| HRT (hours) | \( Y \) (gMLVSS/gCOD) | \( \mu \) (d

| 48 | 0.09 | 0.002 |
| 24 | 0.07 | 0.001 |
| 16 | 0.08 | 0.006 | 0.11 | 0.004 |
| 12 | 0.06 | 0.005 |
| 8  | 0.06 | 0.006 |

Table 3 shows the results of the calculation. It can be found that the biomass yields (\( Y \)) at various operating HRTs ranged from 0.06 to 0.09 gMLVSS/gCOD, which was within an experiential range of 0.03–0.18 gMLVSS/gCOD in anaerobic digestion processes (Henze 2008). The theoretical biomass yield (\( Y_0 \)) and endogenous decay coefficient (\( b \)) were calculated as 0.11 gMLVSS/gCOD and 0.004 d

Biogas production

Figure 4 shows the general conditions of biogas production including daily biogas production and composition of CH\(_4\), CO\(_2\), and N\(_2\) in the produced biogas at various HRTs. As Figure 4(a) shows, the biogas production rate averaged 0.14, 0.28, 0.42, 0.56 and 0.89 L/L/d respectively at HRTs of 48 h, 24 h, 16 h, 12 h and 8 h, and it presented significantly closely dependent on the HRT shortening. The CH\(_4\) content in the biogas tended to be stable at more than 70% when the reactor started up successfully at HRT 48 h, and it reached approximately 80% at HRT 12 h and 8 h. Relatively high solubility of CO\(_2\) to the liquid at room temperature resulted in a relatively low CO\(_2\) content of 5–10% in the biogas (Lettinga et al. 2001). It should be noted that the biogas contained 10–15% N\(_2\), which was higher than expected, and this may result from the effect of soluble N\(_2\) in the feed wastewater and denitrification from nitrate. Besides, the permeate side being exposed to the atmosphere may also result in increasing the N\(_2\) content, because a small amount of atmosphere may enter into the reactor during the relax period within the intermittent filtration cycle, which is carried out by automatically switching on and off the permeate pump. The operation was stopped at the end of the operating duration of HRT 12 h, to take out the fully-fouled membrane to be cleaned for recovery of permeability. The anaerobic environment inside the reactor was broken when withdrawing the membrane, but the biogas production and CH\(_4\) content recovered in a short time, 5 days after the cleaned membrane was reinstalled. Hence, it can inferred that ex-situ membrane cleaning/replacement imposes a slight negative effect on the performance of a SAnMBR.

Figure 4 | Biogas production: (a) biogas production rate; (b) composition of CH\(_4\), CO\(_2\), N\(_2\) in biogas.
Figure 5(a) provides the TCOD mass balance of the SAnMBR at different HRTs. The TCOD mass in the system consisted of the effluent COD in the permeate, the COD converted to CH₄ in the biogas and dissolved in the permeate, and the COD used for MLVSS multiplication. The effluent COD in the permeate accounted for 7.2% of the total TCOD input at HRT 48 h and then decreased with HRT shortening and reached 3.0% at HRT 8 h, which revealed that the degradation efficiency of organic matter tended to be promoted smoothly with decreasing HRT. The COD levels of dissolved CH₄ in the permeate were kept stable in a small range of 6.0 to 6.2%, possibly due to the saturated solubility of CH₄ at 25°C. The COD used for MLVSS multiplication was also at a low level due to the mentioned small biomass yield. Most importantly, TCOD mass conversion to net CH₄ gas was sustained at a high proportion of more than 83% and reached 93.8% at HRT 8 h. The high methane yield can also be reflected in the normalized CH₄ production per gram removal COD shown in Figure 5(b), which was maintained at a value of more than 0.3 NL CH₄/gCOD and reached 0.338 NL CH₄/gCOD, closely approaching the theoretical value of 0.35 NL CH₄/gCOD under the standard state. The in-situ specific methanogenic rate of the biomass in the SAnMBR shown in Figure 5(c) revealed that the biomass capacity of organic degradation/CH₄ production kept obviously increasing with the shortening of operating HRT. Based on the above analysis, this meant that, following the HRT shortening, namely OLR increasing, methanogenic efficiency remained at a high level (>80% most of the time) and tended to increase with shortening of the operated HRT, and furthermore the OLR of 2.1 gCOD/L/d at the shortest HRT, 8 h, was possibly still below the maximum potential of the anaerobic digestion system in the SAnMBR.

Batch tests were implemented to investigate the biochemical methane potential by using different F/M ratios, and the results are provided in Figure 6. An increase in F/M ratio resulted in an increased final CMP. The expected inhibition effect did not occur even when the F/M ratio was increased to a high value of 1.54 gCOD/gMLVSS. The results of batch tests suggested that the mixed liquor in the SAnMBR was able to bear an F/M ratio of more than 1.54 gCOD/gMLVSS without any inhibition effect on methane production. Table 4 shows the results of the Gompertz equation (Equation (1)) during the simulation of batch tests. It can be found that methane production occurred under all F/M ratios with a short lag time, and the maximum methanogenic production rate (Rmax) occurred as 41.9 mL/d when the F/M ratio was 0.7 gCOD/gMLVSS.

Membrane fouling

Compared to the satisfied efficiencies of methanogenic degradation indicated by long-term operational performances and
batch tests, membrane fouling was believed to be the bottleneck for the SAnMBR to be operated at a relatively short HRT, i.e. less than 8 hours. Figure 7(a) shows the fouling performance over the whole experimental process. With the target of maintaining a determined flux at each HRT, TMP increased continuously during the operating process. At the end of HRT 12 h when the TMP reached 20 kPa, the membrane module was taken out of the reactor to be cleaned following the aforementioned process, and was installed back to be operated at HRT 8 h. When the HRT changed to 8 h, the TMP presented a slow increase in the first 15 days but appeared to experience a significant increase after that. This phenomenon was called TMP jump, which has also happened in some other researches and is mainly attributed to the sudden increase in the EPS concentration inside the cake layer attached on the membrane surface (Hwang et al. 2008). TMP increased sharply in the later phase of HRT 8 h and the membrane was fully fouled again on the 31st day of the duration of HRT 8 h.

Common fouling behaviours can be divided into cake fouling and pore fouling (Trussell et al. 2007), membrane cleaning and specific filtration tests are helpful to understand the fouling behaviours at different operating HRTs. As for the purposes of the different cleaning processes, water cleaning is mainly intended to wash away the cake layer on the membrane surface. NaClO solution soaking is mainly used to remove soluble or micro foulants including organic matter and microorganisms, which cause narrowing and blocking of membrane pores. Citric acid solution soaking is mainly used to clear the inorganic matter, especially ions in the membrane pores (Wang et al. 2014). The results of membrane cleaning and serial filtration tests at HRT 12 are shown in Figure 7(b); the flux recovered by about 41.1% of the total recovered amount after water cleaning. It recovered by 52.4% after NaClO solution cleaning from that of water cleaning, and 6.5% after citric acid solution.

<table>
<thead>
<tr>
<th>F/M (gCOD/gMLVSS)</th>
<th>Equivalent HRT (hours)</th>
<th>Maximum methane production rate $R_{max}$ (mL/d)</th>
<th>Lag time $t_0$ (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.21</td>
<td>8.0</td>
<td>31.6</td>
<td>0.044</td>
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<tr>
<td>0.56</td>
<td>3.0</td>
<td>39.0</td>
<td>0.098</td>
</tr>
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<td>0.70</td>
<td>2.4</td>
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<td>0.91</td>
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<td>1.26</td>
<td>1.3</td>
<td>21.7</td>
<td>0.044</td>
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<tr>
<td>1.54</td>
<td>1.1</td>
<td>21.7</td>
<td>0.219</td>
</tr>
</tbody>
</table>

Table 4 | Simulation results of batch tests by Gompertz equation

Figure 7 | Membrane fouling performance at various HRTs (a) and ex-situ filtration tests for the membrane at the end of operating HRT 12 h (b) and 8 h (c).
cleaning, which indicated that the predominant form of fouling could have been pore blocking. Differently, at HRT 8 h (Figure 7(c)), the flux recovered by about 74.4% of the total after water cleaning but it only recovered by 17.2% after NaClO solution cleaning. Hence it can be suggested that pore blocking/narrowing may have dominated the membrane fouling when the HRT was relatively longer than 12 h, while cake formation was the main process when the HRT was short i.e. less than 8 h.

CONCLUSIONS

Comprehensive performances of removal efficiencies, biogas production and membrane fouling were investigated for a SAnMBR treating sewage at 25 °C. Our main findings are as follows.

(1) The SAnMBR showed desirable organic digestion efficiencies as well as high removal of COD, BOD, SS and nonionic surfactants. Small sludge yields of 0.11 gMLVSS/gCOD and high methane production efficiency of 0.538 NLCH4/gCOD make SAnMBRs an attractive technology for sewage treatment.

(2) Advantages of the SAnMBR include small excess sludge production and high bioenergy recovery efficiency, easy adaption to increasing F/M ratios and no apparent microbial inhibition under an F/M ratio less than 1.54 gCOD/gMLVSS.

(3) Because nitrogen and phosphorus observed small removal efficiencies, it can be inferred that SAnMBRs can effectively decouple treating organic matter with nutrients, and reserve all the nitrogen and phosphorus in the effluent for further recovery. Thus application of the effluent for agricultural purposes or forest irrigation is possible.

(4) Despite significant fouling development at 12 and 8 h HRTs, the reversible character in both cases suggests that pore blocking (at HRT > 12 h) and cake layer formation (at HRT < 8 h) are the main processes in the TMP increase.

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REFERENCES


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