



Psychrophilic anaerobic dynamic membrane bioreactor for domestic wastewater treatment: Effects of organic loading and sludge recycling

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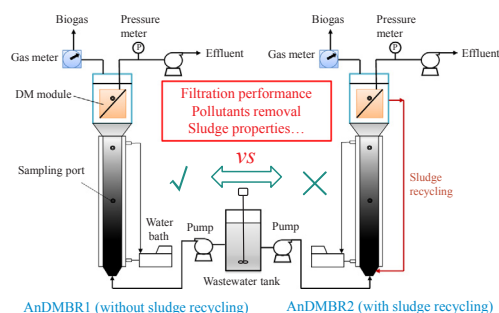
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GRAPHICAL ABSTRACT



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ABSTRACT

Two upflow anaerobic dynamic membrane bioreactors (AnDMBRs) with and without sludge recycling were operated in parallel at varied organic loadings and psychrophilic temperature for domestic wastewater treatment. A 75 μm nylon mesh, used as a supporting material, enabled quick and stable dynamic membrane formation. The AnDMBRs could operate continuously without relaxation at a high flux rate of 22.5 L/m²h; however, high organic loading accelerated the increasing rate of trans-membrane pressure (TMP). High chemical oxygen demand removal was achieved in both AnDMBRs with removal efficiencies of 70–90%. Sludge recycling enhanced the cross-flow velocity but negatively affected the effluent turbidity, sludge properties (particle size reduction and biopolymer release) and dynamic membrane filterability. Although increased organic loading enhanced biogas yield, the low biogas production was related to the dissolved methane loss in the effluent. Easy-operation, minimal maintenance and low-energy consumption makes the AnDMBR process cost-effective for practical wastewater treatment in temperate areas.

1. Introduction

Conventional activated sludge (CAS) processes are widely used for municipal wastewater treatment and result in excellent effluent quality,

which meets increasingly stringent discharge standards. However, high energy consumption for aeration, expensive sludge handling costs, high carbon emissions, and low resource recovery efficiency of the CAS system do not support the goals of sustainable development (Verstraete

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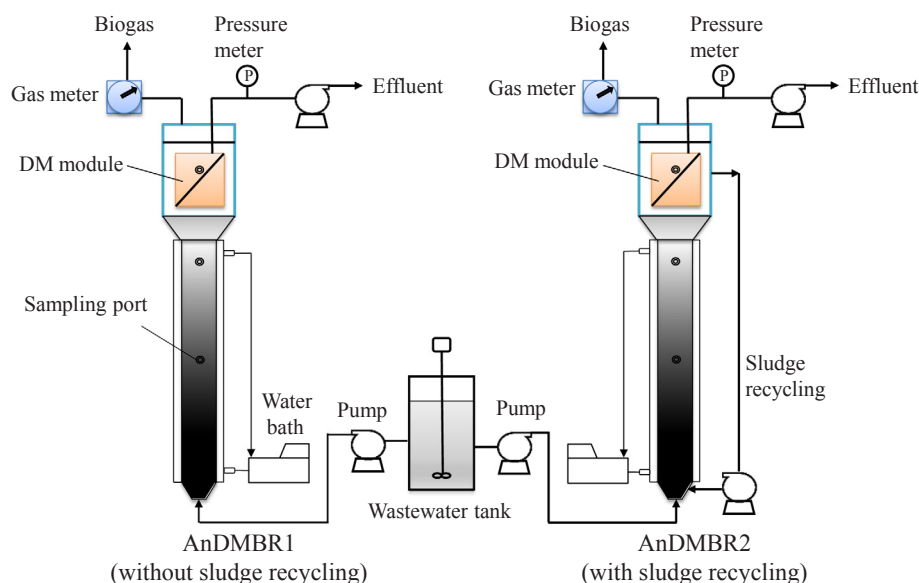


Fig. 1. Schematic diagram of the lab-scale AnDMBRs.

et al., 2009; Martinez-Sosa et al., 2011). Therefore, the application of anaerobic processes for municipal wastewater treatment have been gaining attention in recent years due to the advantages of lower energy demand, the ability to generate methane-rich biogas, and lower sludge production (McCarty et al., 2011). In particular, high rate anaerobic digesters (such as the upflow anaerobic sludge blanket process, (UASB)), commonly used for treating high-strength industrial wastewaters, have been increasingly used for the treatment of low-strength municipal wastewaters (Lettinga et al., 2001). Anaerobic processes have been successfully used for treating municipal wastewater in tropical countries due to high ambient temperature. In temperate or cold countries, municipal wastewater is usually characterized by low temperature, low organic strength and high particulate organic content, which means reduced hydrolysis rate of organic matter, low biomass activity, and low growth rate (Ozgun et al., 2013). Thus, the complexity and variability of municipal wastewater can often result in biomass washouts even in high-rate anaerobic digesters (Quek et al., 2017). Maintaining a long solid retention time (SRT) and high biomass concentration are critical for anaerobic treatment processes.

The anaerobic membrane bioreactor (AnMBR) is considered to be a promising technology for mainstream treatment of municipal wastewater by decoupling hydraulic retention time (HRT) from SRT, which allows maintenance of high sludge concentrations and decreased reactor size (Smith et al., 2014; Shin and Bae, 2018). Moreover, previous work indicated that AnMBR could produce high quality effluent even at extreme conditions, including for low strength wastewater, at low temperatures ($< 20\text{ }^{\circ}\text{C}$), high salinity, and short HRTs (2–8 h), largely due to the efficient retention of suspended solids, colloids, and part of the soluble substances, by using microfiltration/ultrafiltration membranes (Liao et al., 2006; Stuckey, 2012; Lin et al., 2013). However, membrane fouling, low filtration flux, and high membrane cost are still the main challenges limiting the widespread application of the AnMBR process.

Recently, dynamic membrane (DM) filtration technology, which used coarse-pore mesh (10–200 μm) as the inner support material for an outer cake layer formation and achieved excellent solid-liquid separation during suspended solids filtration, was integrated with an anaerobic process to form various AnDMBR configurations (Loderer et al., 2012; Ersahin et al., 2012; Hu et al., 2018). As reported, AnDMBR had similar performance to the conventional AnMBR but had a lower membrane cost and filtration resistance, and easier fouling control (Hu et al., 2018), thus enabling AnMBR applications at a much lower capital

expenditure. As an emerging technology, researchers have attempted to use the AnDMBR for treating real municipal wastewater (An et al., 2009; Ma et al., 2013; Quek et al., 2017), synthetic high-strength wastewater (Ersahin et al., 2014; Alibardi et al., 2016), landfill leachate (Xie et al., 2014), and solid wastes (Liu et al., 2016; Tang et al., 2017a).

To date, the performance of the AnDMBR is influenced by the supporting material, operational conditions, and bioreactor configurations. In an upflow anaerobic sludge blanket and dynamic membrane-coupled process treating municipal wastewater, polyethylene terephthalate mesh with a larger pore size (46 μm) showed similar pollutant removal efficiency but a filtration duration four times longer than mesh with a smaller pore size (28 μm), implying the importance of pore blocking during long-term DM filtration (Quek et al., 2017). Another AnDMBR study indicated that for simulated high strength municipal wastewater, lower HRT ($< 0.5\text{ d}$) decreased average chemical oxygen demand (COD) removal from 80% to 50–60% and also caused more methane loss through the reactor effluent (Alibardi et al., 2016). Moreover, in an immersed AnDMBR, SRT was found to have a significant effect on soluble microbial products (SMP) and extracellular polymeric substances (EPS) production, sludge filterability, DM layer formation and consolidation (Ersahin et al., 2014). Regarding the effect of AnDMBR configuration, submerged AnDMBR requires a shorter start-up period, slightly better permeate quality, and higher biogas production compared to an external AnDMBR (Ersahin et al., 2017). The results deepened the understanding of the AnDMBR process, but previous studies mainly paid attention to synthetic wastewater and were conducted under well-controlled operational conditions (such as mesophilic temperature) rather than the extreme conditions often encountered during real municipal wastewater treatment.

As previously stated, the applicability and performance of AnDMBR for low-strength wastewater treatment at challenging conditions (such as low temperature and HRT) has rarely been studied. Therefore, the main objective of this study was to investigate the viability of AnDMBR for treating domestic wastewater with varied organic loading (COD concentrations ranging from 300 to 1000 mg/L) at psychrophilic temperature (22–25 $^{\circ}\text{C}$) and a short HRT (8 h).

2. Materials and methods

2.1. Lab-scale AnDMBR setup

Two lab-scale AnDMBRs were operated in parallel with an effective

working volume of 3.5 L each. AnDMBR2 had sludge recycling and AnDMBR1 did not. The schematic diagram of the AnDMBRs is shown in Fig. 1. The AnDMBR made of plexi-glass was comprised of an upflow bioreactor and a submerged dynamic membrane module located at the top of the bioreactor. A flat-sheet DM module with a total filtration area of 0.02 m² was used in each bioreactor, and the supporting material used for DM formation was nylon mesh (75 µm pore size) (Hu et al., 2017a). In each AnDMBR system, two peristaltic pumps (Longer BT-100, China) were separately used to feed substrate into the bioreactor and to draw permeate from the DM module. The influent pump was controlled by a water level sensor to maintain a constant water level in the bioreactor. Trans-membrane pressure (TMP) was recorded by an online pressure sensor (SIN-P400, China) located at the permeate line. In AnDMBR2, the sludge mixture was recycled from the top to the bottom of the upflow reactor with the assistance of a peristaltic pump (Longer BT-100, China), which had a flow rate of 1.1 L/h, resulting in a cross-flow velocity (CFV) of 0.83 m/h compared to that of 0.24 m/h in AnDMBR1 without sludge recycling.

The produced biogas that was collected in the head space of the reactor flowed through a gas-liquid separator and was measured by a wet-type gas flowmeter (TC-2, China). The flux was set to a constant rate of 22.5 L/m²h. During the stable operation period, TMP varied in the range of 0–30 kPa. In both systems, the temperature in the reactor was controlled at approximately at 20 °C by using a water bath.

2.2. Experimental procedure

The operation period of 150 d can be divided into Phase I, Phase II, and Phase III according to the varied organic loading. During the three phases, the average COD concentration in the domestic wastewater fed into the AnDMBRs increased from 292 mg/L and 516 mg/L to 1028 mg/L. This resulted in average organic loading rates of 0.88, 1.55, and 3.01 kg COD/m³d, respectively. The HRT was constant at 8 h. The SRT was infinite as no sludge was wasted except that collected for sludge property measurement, given the low biomass yield under psychrophilic temperature.

In order to promote quick DM layer formation in each filtration cycle, sludge recycling was set at a high rate from the top to the bottom of the bioreactor and permeation was set at a high flux during the initial DM formation period of about 15 s. Afterwards, the AnDMBR1 was operated without sludge recycling, thus making anaerobic sludge settle to form a sludge layer and a supernatant layer. In the AnDMBR2, sludge recycling was constant at 1.1 L/h, with no stopping. The DM membrane filtered continuously without relaxation under a vacuum caused by the effluent pump, and no additional methods (such as backwashing and biogas sparging) were used for fouling control. At the end of one phase, the DM module was taken out and hydraulically cleaned to restore permeability, and then the module was reused again for the next phase. This kind of operation mode necessitated minimal maintenance during one filtration cycle.

2.3. Inoculum and wastewater

The AnDMBRs were inoculated with anaerobic sludge from a full-scale anaerobic digester treating brewery wastewater. The concentration of the seed sludge was 5600 mg/L. Two weeks of acclimation using real domestic wastewater was conducted before the stable operation. In Phase I, real domestic wastewater was used as substrate, while in Phase II and Phase III, synthetic wastewater, containing glucose and other micronutrients, was prepared and added to the real domestic wastewater to increase influent COD concentration. The recipe for preparing synthetic wastewater was per the reference (Ersahin et al., 2014). Wastewater treatment plant influent is relatively stable, as noted in this study (influent COD ranging from 200 to 400 mg/L), thus adding synthetic wastewater to real wastewater to improve organic loading rates (OLR) is reasonable in order to simulate real wastewater as much as

possible.

2.4. Analytical methods

Measurements of COD, total phosphorus (TP), total nitrogen (TN), ammonia (NH₃-N) in the influent and effluent, and mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) in the bioreactor were performed according to Chinese National Environmental Policy Act (NEPA) standard methods (Chinese NEPA, 2002). The pH and ORP were measured using respective meters (Hach HQ30, USA). Turbidity measurement was conducted with a Hach 2100N turbidimeter. Soluble COD samples were filtered through a 0.45 µm filter before the analysis.

The DM layer on about 0.005 m² of the membrane surface (one quarter of the total membrane area) was scraped off with a plastic sheet at the end of each phase. The collected sludge was diluted with deionized water to a volume of 15–20 mL, and then the diluted sample was gently mixed using a magnetic blender. Afterwards, the sludge samples were subjected to total suspended solids (TSS) and volatile suspended solid (VSS) measurement. The total TSS and VSS per unit membrane area could be calculated based on the measured TSS/VSS concentrations and known membrane area.

SMP and EPS were extracted from sludge samples at different phases (Hu et al., 2017b), while the main components (proteins and polysaccharides) were also measured according to the reported methods (Hartree, 1972; Dubois et al., 1956). The biogas composition (such as CH₄, CO₂, N₂, and H₂) was measured using a gas chromatograph (PerkinElmer clarus680, USA) equipped with a thermal conductivity detector (TCD) and a 2 m carbon molecular sieve TDX-01 column. Volatile fatty acids in the influent, effluent, and bioreactors was measured using a gas chromatograph (GC2014, Shimadzu, Japan) with a flame ionization detector, according to a previous study (Tang et al., 2017b). Particle size distribution (PSD) of the anaerobic sludge was measured using a laser granularity distribution analyzer (LS 230/SVM +, Beckman Coulter Corporation, USA). The molecular weight distributions (MWD) of samples were determined using a Gel Filtration Chromatogram (GFC) analyzer (LC-2010A, Shimadzu Corporation, Japan) installed with a Zenix SEC-100 type gel column (Sepax Technologies Corporation, USA) and an ultraviolet (UV) detector (SPD-10, Shimadzu Corporation, Japan) at 40 °C.

3. Results and discussion

3.1. Filtration performance

In this study, constant flux filtration mode was used in the AnDMBRs at a flux of 22.5 L/m²h. The evolution profiles of TMP over operation time were plotted to reflect the DM filtration behaviors as shown in Fig. 2. Influent and effluent turbidity were also measured to verify the retention effect of the DM layer and sustainable filtration. In order to assess the viability of the AnDMBR process for treating wastewater with varied organic strength, as is commonly encountered in practical municipal wastewater treatment, different organic loadings were examined during long-term operation (from Phase I to Phase III). It was noted that for treating low-strength real domestic wastewater (average COD = 292 mg/L) in Phase I, AnDMBRs with and without sludge recycling showed little difference in turbidity removal. Effluent turbidity was lower than 30 NTU, indicating effective retention of particulate substances by the stable DM layer. Similar turbidity removal performance was reported in previous AnDMBR studies (Ersahin et al., 2014, 2017). Slightly higher TMP values were always observed in AnDMBR2, in which sludge recycling was conducted to enhance CFV along the DM surface and to promote sludge mixture. After a gradual increase, at the end of Phase I (45 d), TMP in the AnDMBRs was less than 15 kPa and 15–18 kPa, respectively. It was worth noting that continuous permeate extraction was used without periodic relaxation or

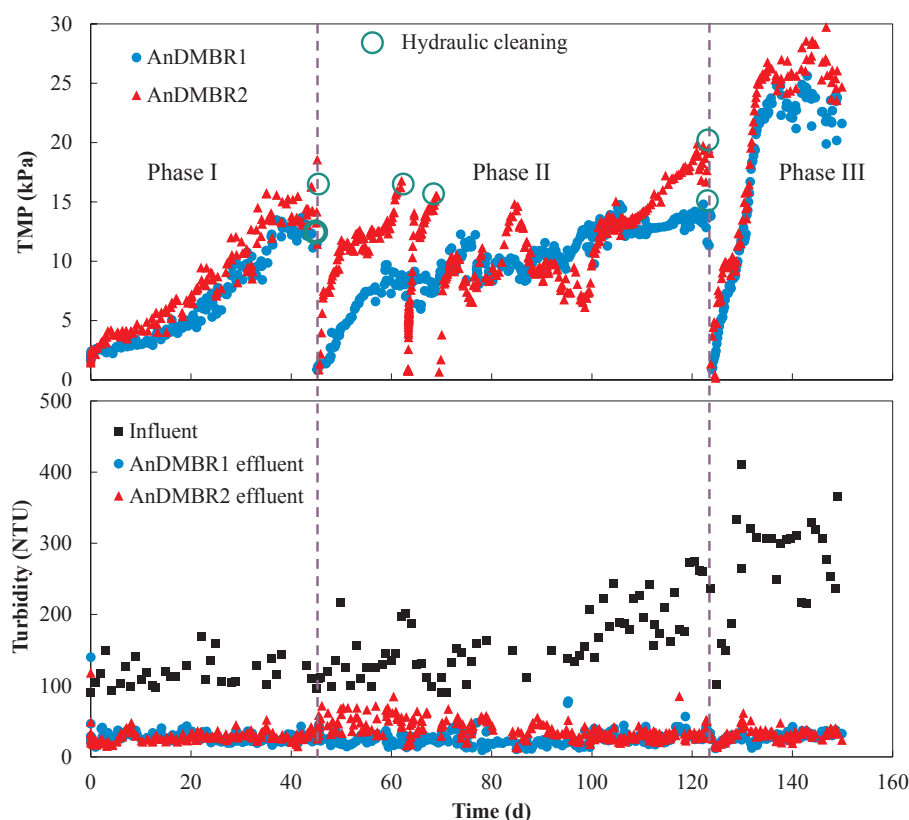


Fig. 2. Profiles of TMP and turbidity in the AnDMBRs at different phases.

back-flushing and no any other fouling mitigation methods were adopted, which was different from the conventional aerobic MBRs and AnMBRs (Meng et al., 2017; Lei et al., 2018). As reported, periodic relaxation and back-flushing, biogas sparging, vibration, and various types of chemical cleaning were commonly used for fouling control in conventional AnMBRs (Giménez et al., 2011; Robles et al., 2012; Lin et al., 2013), which would affect the net permeability and maintenance costs. Notably, a large portion (over 70%) of energy demand is due to fouling control when biogas sparging is used. The energy demands are 0.69–3.41 kWh/m³ with specific gas demands (SGD) ranging from 0.4 to 3.0 m³/m²h in lab-scale and pilot-scale AnMBRs (Mei et al., 2016; Shin and Bae, 2018; Lei et al., 2018). However, the AnDMBRs were only hydraulically cleaned using tap water once at the end of Phase I to restore DM filterability. This easy operation and minimal maintenance as well as low energy consumption enabled the AnDMBR to be an attractive process for treating low-strength wastewater compared to conventional AnMBRs.

In Phase II, influent organic loading (average COD = 515.6 mg/L) was improved by adding synthetic substrate to real domestic water. AnDMBR2 showed a quick TMP climb during the first 20 d and had to be physically cleaned twice in order to sustain long-term operation. Meanwhile, effluent turbidity also fluctuated substantially. In the following 60 d, the TMP increase rate was much lower though some fluctuations were noted. However, AnDMBR1 still showed a stable filtration behavior, with a quick TMP rise followed by a gradual increase until the end of the operation period. The differing performance could be due to the accumulation of anaerobic biomass on an altered substrate (Xie et al., 2014). Moreover, sludge recycling seemed to prolong this adaption process and had a negative effect on sludge properties.

In Phase III, mixed wastewater with an even higher organic loading was fed into the AnDMBRs. TMP climbed in both reactors almost linearly and quickly exceeded 20 kPa over a short time (less than 20 d), and then the TMP increase rate slowed down. At higher organic loading

and low temperature, the accumulation of metabolic products and the change in sludge properties (production of biopolymers) could occur, which would deteriorate DM filtration behavior (Lettinga et al., 2001; Martín García et al., 2013; Alibardi et al., 2016), but the effluent turbidity did not seem to be affected. During the stable operation period, suspended solids (SS) in the mixed liquor near the membrane in AnDMBR1 and AnDMBR2 were negligible compared to the MLSS concentration in the sludge layer (6–16 g/L) and the supernatant turbidity was measured at 10–140 NTU and 130–270 NTU in the AnDMBRs. Thus, the effects of SS on fouling behavior and DM evolution were not significant. The applied sludge recycling in AnDMBR2 was expected to scour the membrane surface and modify sludge properties, thus affecting DM filterability. This is discussed later. The effluent SS concentration ranged from 10 to 20 mg/L and 15 to 25 mg/L for AnDMBR1 and AnDMBR2, respectively, which did not meet the most rigorous wastewater discharge limits in China, but did meet the water quality standards for irrigation in agriculture and other reuse purposes.

The initial formation process of the DM layer is also discussed herein. As mentioned in Section 2.2, after a short time (15 s) of high-rate sludge recycling and high-flux filtration in both AnDMBRs, the effluent turbidity decreased dramatically from above 100 NTU to a low level within 5 min, and declined gradually afterwards in the first filtration cycle. In subsequent cycles after physical cleaning, effluent turbidity at the DM formation stage showed a low initial value (< 50 NTU), and decreased to a lower value afterwards. The results indicated quick DM layer formation within 5 min, which was not affected by physical cleaning, and enabled effective retention of sludge particles.

Overall, the results indicated that for treating municipal wastewater with varied strength, the AnDMBR1 without sludge recycling showed stable filtration behavior and satisfactory turbidity removal with low energy consumption and maintenance requirements. For higher strength wastewater treatment (municipal or industrial wastewater), further investigations were still needed to optimize operating

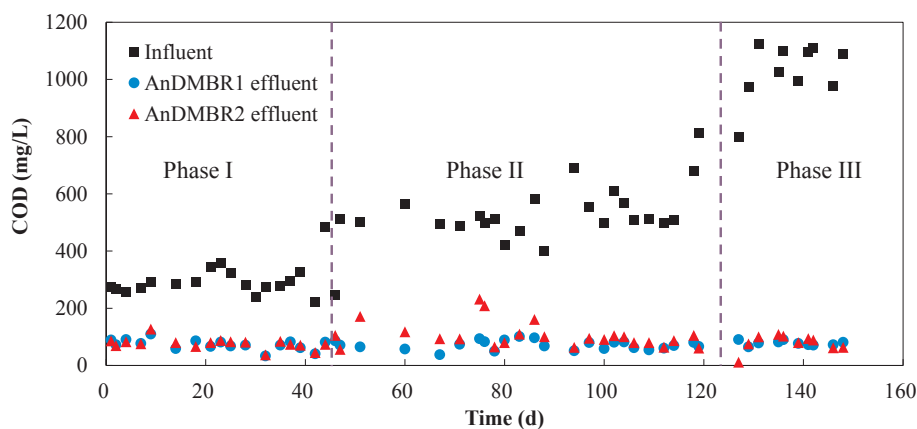


Fig. 3. Variation of COD in the influent and effluent of AnDMBRs at different phases.

conditions to alleviate the rapid increase in TMP.

3.2. Treatment performance

Influent and effluent COD concentrations over the operation period are shown in Fig. 3. As shown, regardless of the increase in average influent COD concentration from 292 to 1028 mg/L, the effluent COD was always less than 100 mg/L in AnDMBR1, with the average values of 73.2, 73.6, and 78.1 mg/L during Phase I, Phase II, and Phase III, respectively. The corresponding COD concentration of AnDMBR2 effluent was 77.5, 108.4, and 82.5 mg/L in Phase I, Phase II, and Phase III, respectively, which was consistently higher than those detected in AnDMBR1. The results were similar to the variation in effluent turbidity previously discussed. It was worth noting that although influent organic loading increased by nearly four times from Phase I to Phase III, the COD removal efficiency was not affected. Instead, it increased from 75% to 92%, and 73% to 92% in the two AnDMBRs. The added synthetic substrate, consisting of biodegradable organics and other micronutrients, could be easily biodegraded by anaerobic biomass. The effective biodegradation, DM retention and in-situ biodegradation all contributed to the stable COD removal as noted in previous DMBR studies (Ersahin et al., 2014; Alibardi et al., 2016; Hu et al., 2016).

Table 1 shows detailed data in terms of average influent COD, supernatant COD, effluent COD, and biogas yield. Adding synthetic substrate to domestic wastewater in Phase II and Phase III obviously enhanced the soluble COD concentration. However, after anaerobic digestion in the bioreactor, the average sludge supernatant COD was 96.8–110.3 mg/L and 105.7–179.3 mg/L in AnDMBRs 1 and 2, respectively, which was still higher than their corresponding effluent COD (73.2–78.1 mg/L and 77.5–108.4 mg/L). It was attributed to the rejection of particulate substances and the degradation of organic matter by active biomass (or biofilm) in the DM layer (Alibardi et al., 2016). Comparing the total COD and soluble COD in the effluent, it was

found that soluble COD concentrations of 43.2–49.8 mg/L and 41.7–73.2 mg/L still contributed to 59–67% and 54–70% of the total effluent COD in the AnDMBRs, which was largely due to the production of SMP in the bioreactor that could permeate into the effluent as residual organic matter. In a submerged AnDMBR for synthetic concentrated wastewater treatment, the total and soluble effluent COD were 100 and 85 mg/L (Ersahin et al., 2017). On the other hand, it should be noted that a substantial amount of particulate COD existed in the AnDMBR permeate, indicating insufficient retention of fine particles by the DM filtration process compared to UF/MF membranes used in AnMBRs. Thus, further efforts to enhance filtration performance of the DM layer were needed. This was done by regulating DM structure using additives (Chu et al., 2010) or modifying sludge properties to lower the contents of SMP and fine particles (Stuckey, 2012).

In addition, after stable biogas production was obtained, the average biogas yield in a gaseous state during Phase I was 0.30 L/day and 0.25 L/day in AnDMBR1 and AnDMBR2, respectively. For low-strength domestic wastewater treatment at psychrophilic temperature, the biogas yield was low compared to other reported results obtained for synthetic and industrial wastewater treatment at high temperature conditions (Lin et al., 2013; Ersahin et al., 2014; Jeong et al., 2017), and similar to the results reported for practical municipal wastewater treatment at low temperatures (Smith et al., 2013; Alibardi et al., 2016). It was explained that in municipal wastewater, particulate organic substances accounted for more than 50% of the total organic matter and a substantial part of particulate organics were slow-biodegradable and even non-biodegradable substances (Lettinga et al., 2001; Zhang et al., 2018). Thus, low conversion of organics into biogas could be expected. Furthermore, as previously documented, at low temperatures, the activities of anaerobic microorganisms were inhibited to some extent, especially for the hydrolysis process and methanogenesis process (McKeown et al., 2012; Bandara et al., 2012). The dissolved biogas lost in the AnDMBR effluent could also contribute to the low biogas production observed. However, in Phase II and Phase III, the addition of synthetic substances (glucose as a soluble organic) enhanced the organic loading and significantly increased biogas yield from approximately 0.4 to 1.6 L/d. In spite of organic loading and sludge recycling, methane content in the generated biogas ranged from 70% to 80%. The results indicated that when using AnDMBR for practical wastewater treatment at low temperature, biogas production and recovery would be a great challenge. However, as explored by previous researchers, the co-digestion with readily biodegradable substances or the addition of a treatment unit for accumulated particle hydrolysis could be feasible options to enhance biogas generation (Mahmoud et al., 2004; Zhang et al., 2013).

If the produced biogas was converted into electrical energy, it could be used to supplement energy requirements for energy-neutral or even energy-positive wastewater treatment processes (McCarty et al., 2011;

Table 1
Important parameters of operation of the AnDMBRs (average values).

Items	Phase I	Phase II	Phase III
Influent COD (mg/L)	292.0 ± 62.2	515.6 ± 63.4	1027.7 ± 107.9
Influent SCOD (mg/L)	83.0 ± 20.7	282.7 ± 105.0	581.32 ± 131.4
^a Supernatant COD (mg/L)	96.8 ± 17.0	110.3 ± 39.6	101.4 ± 8.3
^b Supernatant COD (mg/L)	105.7 ± 21.1	179.3 ± 70.9	114.5 ± 21.2
^a Effluent COD (mg/L)	73.2 ± 17.6	73.6 ± 17.8	78.1 ± 9.1
^b Effluent COD (mg/L)	77.5 ± 19.2	108.4 ± 45.9	82.5 ± 30.9
^a Effluent SCOD (mg/L)	43.2 ± 17.6	49.8 ± 15.1	47.9 ± 9.1
^b Effluent SCOD (mg/L)	41.7 ± 11.5	73.2 ± 34.9	57.9 ± 15.1
^a Biogas yield (L/d)	0.30 ± 0.14	0.41 ± 0.19	1.56 ± 0.39
^b Biogas yield (L/d)	0.25 ± 0.08	0.37 ± 0.13	1.65 ± 0.45

Note: ^aAnDMBR1; ^bAnDMBR2.

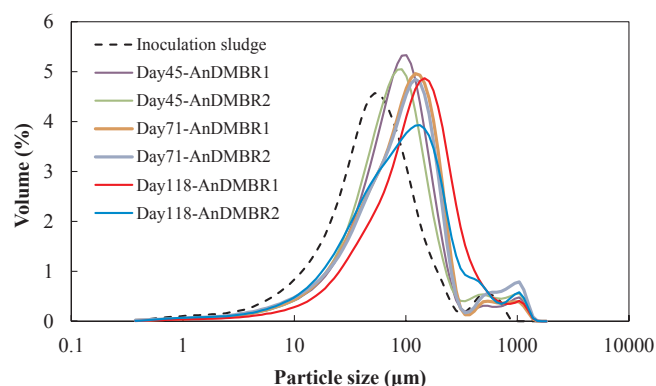


Fig. 4. Particle size distribution of different sludge samples.

Mei et al., 2016). Through the detailed energy balance calculation, it was observed that if operated under ambient temperature (without heating), the AnDMBRs can achieve net energy production of 0.06 and 0.05 kWh/m³ in Phase I considering overall energy consumption in terms of wastewater feeding, effluent extraction, sludge recycling, and electrical energy generation from produced methane (Kim et al., 2011; Aslam et al., 2017; Lei et al., 2018). In Phase II and Phase III, much higher net energy production could be expected due to identical energy consumption under the same operating conditions but greater methane production as biogas yield. The analysis further verified the potential of net energy production using a membrane-based anaerobic process for efficient wastewater treatment.

3.3. Sludge properties

PSD analysis of inoculation sludge and different sludge samples collected from the two AnDMBRs is shown in Fig. 4. All of the PSD profiles have a multimodal curve, with the particle size of most suspended solids between 0.4 and 400 μm and the others between 400 and 2000 μm. The mean particle size of inoculation sludge was 48.6 μm. With the increase in operation time, in Day 45, Day 71, and Day 118, the mean particle size of sludge samples from AnDMBR1 was 76.2, 81.4, and 109.7 μm, respectively, while in AnDMBR2 the corresponding values were 76.0, 83.2, and 83.7 μm, respectively. Thus, regardless of sludge recycling, sludge particle size increased with operation time, which was consistent with studies regarding submerged aerobic DMBRs and anaerobic DMBRs (Hu et al., 2017b; Ersahin et al., 2017). DM has a lower effective retention efficiency than UF/MF membranes, so part of the fine particles and colloids could pass through the DM layer, making the PSD move towards the larger size. It was also noted that sludge PSD of AnDMBR1 improved constantly with time, which differed from stable PSD observed in AnDMBR2. This might be because sludge recycling negatively affected sludge floc structure and enhanced the release of fine particles and colloids as potential foulants, which would affect the effluent quality (such as turbidity and COD) and also DM filtration performance (quicker TMP increase always observed in AnDMBR2). In an external AnDMBR system with sludge recycling, the negative effects of the higher shear force impacted DM formation, PSD, and microbial activity (Ersahin et al., 2017).

SMP and EPS were considered the main contributors to membrane fouling in various MBRs and DMBRs (Yue et al., 2015; Kunacheva et al., 2017; Saleem et al., 2016); thus, more attention was paid to the properties of these biopolymers. As shown in Table 2, SMP amounts in both AnDMBRs constantly increased from Phase I to Phase II, while EPS had the highest concentrations in Phase II. Proteins were the main components of SMP and EPS, accounting for 80% of the total amount in most cases. In AnDMBR2, the increase in SMP was more obvious, increasing from 2.67 mg/gMLSS to 8.85 mg/gMLSS, among which proteins increased from 2.1 mg/gMLSS to 8.5 mg/gMLSS. The increase in

Table 2
SMP and EPS concentrations in the AnDMBRs.

Items		SMP (mg/gMLSS)			EPS (mg/gMLSS)		
		PS	PN	Total	PS	PN	Total
AnDMBR1	Phase I	0.26	0.80	1.06	1.73	11.85	13.58
	Phase II	1.04	2.92	3.96	12.29	30.73	43.02
	Phase III	0.30	5.58	5.89	5.07	20.16	25.22
AnDMBR2	Phase I	0.60	2.07	2.67	3.71	23.79	27.50
	Phase II	1.15	5.16	6.31	18.86	43.38	62.24
	Phase III	0.34	8.50	8.85	7.71	31.07	38.79

Note: SMP and EPS were collected after well-mixing of sludge mixture at the end of Phase I, II, and III; polysaccharides (PS) and proteins (PN) were measured with average values reported.

SMP (especially for proteins) concentrations was in accordance with the rising tendency of TMP as discussed in Section 3.1. In several studies, polysaccharides in SMP were also regarded as critical foulants (Meng et al., 2017). However, in this study it was noted that polysaccharides in SMP were much lower in concentration than proteins, and the variation of polysaccharides (rising from Phase I to Phase II and lowering in Phase III) were not related to TMP evolutions. No such relationship was detected between EPS and the main components as TMP increased. Thus, proteins in SMP were considered to be the main foulants responsible for the quick TMP increase and high filtration resistance during long-term AnDMBR operation, especially for the high organic loading encountered in Phase III.

To further illustrate the properties of dissolved organic matters (DOM) from influent, sludge supernatant, and effluent, GFC was used to detect molecular weight distribution (MWD). Typical MWD curves of different water samples in Phase I were taken for analysis. It was noted that DOM in the influent showed a broad MWD with the liquid chromatography elute time from 7.5 min to 25 min, indicating the coverage of micro, high, intermediate, and low MW organics (Hu et al., 2017a). It was interesting to find out that the elute peaks around 13 min (high MW organics) showed much higher intensities in sludge supernatant samples than those in the influent, indicating the generation of SMP during biodegradation or biomass decay. While comparing other peaks between sludge supernatant and influent, substantial reduction was noted, due to the effective anaerobic digestion by active biomass. Furthermore, the comparison of MWD curves between sludge supernatant and effluent samples, showed that DOM concentrations in all the molecular weight ranges decreased, indicating that the retention of a DM layer contributed to DOM removal. It was also noted that part of the DOM (especially high to low MW substances with elute time longer than 13 min) could pass through the DM layer. The retained organics (such as SMP) would affect the evolution of the structure and various properties of the DM layer and contribute to the long-term filterability deterioration.

3.4. Characteristics of the DM layer

At the end of one operation period, the DM modules were also taken out for observation and characterization. As noted, after an operation period, the surface of new mesh was covered with a stable, black DM layer. The DM layer was reported to be formed by biomass and other accumulated materials such as EPS-like material and various kinds of inorganic compounds (Ersahin et al., 2016). The thickness of DM layers was measured to be 0.76, 0.90, and 1.1 mm in AnDMBR1, and 0.54, 0.63, and 0.75 mm in AnDMBR2 from Phase I to Phase III, respectively. This was due to the fact that with the increase in organic loading, more SMP, fine particles, and colloids accumulated in the DM layer. Moreover, the sludge recycling implemented in AnDMBR2 showed positive effects on controlling DM layer thickness. As shown in Table 3, the attached TSS and VSS in the AnDMBRs also increased from Phase I to

Table 3
Properties of the DM layer in the AnDMBRs.

Items		Thickness (mm)	VSS (g/m ²)	TSS (g/m ²)
AnDMBR1	Phase I	0.76	23.76	32.23
	Phase II	0.90	29.62	39.54
	Phase III	1.27	64.13	93.73
AnDMBR2	Phase I	0.54	20.36	30.15
	Phase II	0.63	30.20	38.86
	Phase III	0.95	44.83	74.45

Note: DM layers were collected at the end of Phase I, II, and III with average values reported.

Phase III, especially in Phase III. The results verified the production and release of biopolymers, which contributed to thicker DM layer formation. More TSS and VSS accumulated, and greater DM layer thickness was detected in AnDMBR1 compared to AnDMBR2. This did not mean higher TMP increase rates (mentioned in Section 3.1). Thus, the composition (such as fine particles and biopolymers) and structure of the formed DM layer determined DM filterability; although too much accumulation of foulants could lead to thick DM formation as noted in Phase III.

In aerobic and anaerobic MBRs, it was reported that fine particles in the sludge suspension had a tendency to deposit on the membrane surface, but the large particles could easily detach from the membrane surface due to gas scouring (Ersahin et al., 2017). Thus, it was expected that the higher CFV in AnDMBR2 was effective in preventing the accumulation of larger particles, and scouring of the surface of the DM layer was verified by the lower DM thickness. On the other hand, under higher CFV, the attached particles would be smaller in particle size, which might reduce the porosity of the DM layer, resulting in a compact DM structure and increased filtration resistance compared to the AnDMBR1, which did not have sludge recycling-induced CFV.

4. Conclusions

The AnDMBRs could successfully operate under high flux (22.5 L/m²h) and short HRT (8 h) with different organic loadings (0.88–3.01 kg COD/m³d) at psychrophilic temperature. Based on the DM filtration behavior and pollutant removal performance, it did not seem necessary to recycle sludge when treating low strength domestic wastewater, due to the negative effects of sludge recycling induced CFV on sludge properties. Increasing organic loading showed little influence on COD and turbidity removal but substantially enhanced methane production. Strategies enabling efficient biogas production and dissolved methane recovery will further improve net energy production in AnDMBRs.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.biortech.2018.08.128>.

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