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Sustainable organic loading rate and energy recovery potential of mesophilic anaerobic membrane bioreactor for municipal wastewater treatment

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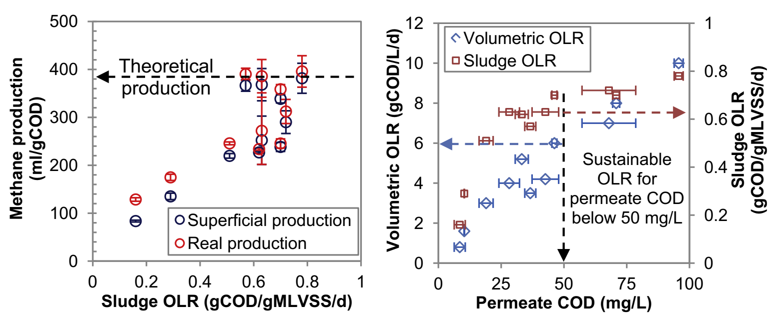
HIGHLIGHTS

- Sustainable volumetric and sludge OLR was 6 gCOD/L/d and 0.63 gCOD/gMLVSS/d.
- Sustainable sludge OLR resulted in high methane production up to theoretical value.
- A very low biomass production of 0.015–0.026 gMLVSS/gCOD was observed.
- A sustainable flux of 6 L/m^2/h maintained stable permeability for over 3 months.
- AnMBR coupling heat pump and forward osmosis was promising in temperate area.

GRAPHICAL ABSTRACT



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ABSTRACT

The overall performance of a mesophilic anaerobic membrane bioreactor (AnMBR) for synthetic municipal wastewater treatment was investigated under a range of organic loading rate (OLR). A very steady and high chemical oxygen demand (COD) removal (around 98%) was achieved over a broad range of volumetric OLR of 0.8–10 gCOD/L/d. The sustainable volumetric and sludge OLR satisfying a permeate COD below 50 mg/L for general reuse was 6 gCOD/L/d and 0.63 gCOD/gMLVSS (mixed liquor volatile suspended solids)/d, respectively. At a high sludge OLR of over 0.6 gCOD/gMLVSS/d, the AnMBR achieved high methane production of over 300 ml/gCOD (even approaching the theoretical value of 382 ml/gCOD). A low biomass production of 0.015–0.026 gMLVSS/gCOD and a sustainable flux of 6 L/m^2/h were observed. The integration of a heat pump and forward osmosis into the mesophilic AnMBR process would be a promising way for net energy recovery from typical municipal wastewater in a temperate area.

1. Introduction

Municipal wastewater can be regarded as a valuable resource rather than a simple waste (Mo and Zhang, 2013). Compared with the widely-used aerobic biological process for municipal wastewater treatment, the anaerobic biological process has some potential benefits including no energy input required for aeration, energy recovery through methane production, lower sludge production and most nutrients (nitrogen and phosphorus) remain in the effluent suitable for agricultural and landscaping irrigation reuse (McCarty et al., 2011). Despite these advantages, the perceived notion is that an anaerobic biological process is inappropriate for municipal wastewater treatment as there is relatively low concentration of organics (in terms of chemical oxygen demand (COD), usually less than 1000 mg/L) with a significant particulate fraction in

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municipal wastewater. This makes it technically challenging to achieve a high organic loading rate (OLR) to sustain a sufficient amount of slow-growing anaerobic biomass in the bioreactor. Furthermore, a conventional anaerobic activated sludge process is generally restricted by the sludge-water separation performance in a gravitational settler (McCarty et al., 2011), and may not be able to achieve high organics removal and good effluent quality.

An anaerobic membrane bioreactor (AnMBR) – a hybrid of anaerobic digestion and membrane separation – may overcome the drawbacks of the conventional anaerobic activated sludge process through the excellent sludge-water separation by a microfiltration (MF) or ultrafiltration (UF) membrane. The coupling of a membrane results in high biomass (and thus high OLR and organics removal), and produces a good quality effluent that has significantly lower amount of particles and pathogens (Liao et al., 2006). Several recent reviews on published AnMBR studies over the past two decades show that AnMBR can achieve high COD removal (more than 85%, up to 99%) with low permeate COD (less than 100 mg/L, down to below 10 mg/L) under a range of hydraulic retention time (HRT) of 3–120 h, sludge retention time (SRT) of 15 d to infinity, volumetric OLR of 0.2–5 gCOD/L/d and operating temperature of 10–37 °C for real and/or synthetic municipal wastewater treatment (Lin et al., 2013; Ozgun et al., 2013; Skouteris et al., 2012; Smith et al., 2012; Stuckey, 2012).

For the energy recovery (expressed as methane production) from municipal wastewater through an AnMBR, the observed methane production in terms of millilitres methane (at the standard conditions of 25 °C and 1 atm) per gram of removed COD (ml/gCOD) has been reported to be in the range of 110–320 ml/gCOD (Gimenez et al., 2011; Ho and Sung, 2009; Hu and Stuckey, 2006; Huang et al., 2013; Kim et al., 2011; Lin et al., 2011; Martinez-Sosa et al., 2011; Saddoud et al., 2007; Wen et al., 1999; Yoo et al., 2012). However, it is still substantially lower than the theoretical value of 382 ml/gCOD, even considering the maximum methane production of 320 ml/gCOD reported which was achieved under optimal conditions for a mesophilic AnMBR using synthetic municipal wastewater (Hu and Stuckey, 2006). Most of these mentioned studies have maintained the optimal conditions including mesophilic temperature, neutral pH, long SRT and strict anaerobic environment to favor methane production. In order to enhance methane production further, increasing the food to microorganism ratio (F/M), and thus the sludge OLR, may be a possible way as high sludge OLR would promote more microorganisms to convert more substrate into methane. However, due to both high biomass concentration in terms of mixed liquor (volatile) suspended solids (ML(V)SS) and low volumetric OLR in these studies, sludge OLR is generally in the low range of 0.03–0.5 gCOD/gMLVSS/d, which may limit the observed methane production. There are only a few studies (Saddoud et al., 2007) that showed increasing biogas production with OLR. Therefore, the comprehensive effects of OLR on methane production as well as organics removal and biomass production still need further investigation to optimize AnMBR operation.

The overall performance (COD removal, permeate COD, methane/biomass production, membrane permeability) of a mesophilic AnMBR treating synthetic municipal wastewater at different OLR levels was intensively investigated to explore the sustainable OLR in this study. The energy recovery potential from typical municipal wastewater through AnMBR was also analyzed.

2. Methods

2.1. AnMBR set-up

A schematic diagram of the lab-scale AnMBR set-up used in this study is shown in Fig. 1. The set-up consisted of an commercial

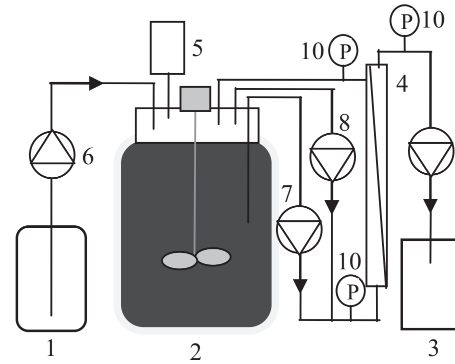

Fig. 1. Schematic diagram of the lab-scale AnMBR set-up.

anaerobic completely stirred tank reactor (CSTR, Applikon Biotechnology, Netherland) with an effective volume of 2 L and integrated control functions (including temperature, pH, level, mixing), a side-stream hollow fiber polyvinylidene fluoride (PVDF) UF membrane module with a nominal pore size of 30 nm and effective filtration area of 310 cm^2, pumps (for feeding influent, sludge recirculation, membrane permeation, biogas recirculation), influent/permeate tank, biogas collection system, pressure sensors, and connection tubing.

The synthetic municipal wastewater in the influent tank was pumped into the well-mixed anaerobic CSTR under the control of a level sensor to maintain the stable effective volume. The sludge mixture in the anaerobic CSTR was pumped at a pre-set flow (corresponding to superficial crossflow velocity (CFV) of 0.1–0.3 m/s along the membrane surface) into the membrane module, where one part passed through the membrane into the permeate tank while the other part was recycled back into the anaerobic CSTR (i.e., sludge recirculation). The pre-set permeate flow was controlled by the permeate pump with intermittent operating mode (on/off of 9/1 min) to achieve constant-flux operation. The biogas in the anaerobic CSTR was also continuously pumped at a pre-set flow (corresponding to superficial CFV of 0.1–0.2 m/s along the membrane surface) into the membrane module to enhance gas–liquid diphase crossflow scouring along the membrane surface for membrane fouling control. The pressure sensors connected in recirculation and permeate tubing were used to monitor the trans-membrane pressure (TMP) as the indicator for membrane fouling. A gas bag was connected to the headspace of the anaerobic CSTR to collect biogas.

2.2. AnMBR operational conditions

A synthetic recipe simulating municipal wastewater (COD 400 mg/L) in Table 1 was used in this study. It contained COD of 400 ± 10 mg/L and total organic carbon (TOC) of 115 ± 5 mg/L, which resulted in a mean oxidation number (MON) of −1.2 according to the equation (MON = 4–1.5 × COD/TOC). In order to explore the maximum sustainable OLR, concentrated wastewater (COD 750–5000 mg/L) from the original recipe was also used due to the limit of reducing HRT caused by the small membrane area. A concentrated stock (20 times) was prepared weekly using tap water and stored in a refrigerator (4 °C), which was diluted to the target concentration for daily use as influent.

Table 1
Recipe of synthetic municipal wastewater.

Chemical compounds	Concentration (mg/L)	Food ingredients	Concentration (mg/L)	Trace metals	Concentration (mg/L)
Urea	91.7	Starch	122	$Cr(NO_3)_3 \cdot 9H_2O$	0.770
NH_4Cl	12.8	Milk powder	116	$CuCl_2 \cdot 2H_2O$	0.536
Na-acetate	79.4	Yeast	52.2	$MnSO_4 \cdot H_2O$	0.108
Peptone	17.4			$NiSO_4 \cdot 6H_2O$	0.336
$MgHPO_4 \cdot 3H_2O$	29.0			$PbCl_2$	0.100
KH_2PO_4	23.4			$ZnCl_2$	0.208
$FeSO_4 \cdot 7H_2O$	5.8				

The seed sludge was a pre-screened (0.5 mm mesh size) mixture of camel manure from Jeddah and anaerobic digester sludge from the Riyadh wastewater treatment plant in Saudi Arabia. Before starting the AnMBR experiment, the seed sludge was acclimated with synthetic municipal wastewater for 4 months. No sludge wasting except regular sampling for biomass measurements (weekly, about 10–15 ml sampling) and intensive sampling to seed another anaerobic reactor (200 ml sampling in Run 5 listed in Table 2) was done. According to the general calculation method of total sludge volume in CSTR divided by daily wasting sludge volume from CSTR, the SRT was approximate to 1000 d in most runs. During the whole operation, the anaerobic bioreactor was well controlled at a temperature of 35 ± 1 °C (mesophilic), stirrer speed of 200 ± 2 rpm and pH of 7 ± 0.1 through the built-in control systems of this commercial CSTR. Oxidation reduction potential (ORP) was also monitored and stabilized at around −440 mV. The operational parameters in all runs are shown in Table 2. For membrane cleaning, periodic (1–3 times per week) enhanced biogas-sludge scouring (double normal CFV in Table 2 for 5–15 min) along the membrane surface without permeation was applied. No chemical cleaning was done during the whole operation.

The overall performance including organics removal, biogas production, biomass evolution, microbial community dynamics, and membrane permeability were monitored during all runs. Commercial COD kits (TNT series, Hach Company) were used to measure the COD of influent and permeate based on the method of rapid digestion (150 °C, 2 h) followed by colorimetric measurement. Biogas composition and volume were measured according to the gas bag method (Ambler and Logan, 2011) based on gas chromatography (GC). One GC (SGI 301C) equipped with a molecular sieve column (argon as carrier gas) followed by a thermal conductivity detector (TCD) was used to detect hydrogen, nitrogen and methane. Another GC (SGI 301C) equipped with a silica column (helium as carrier gas) followed by a TCD was used to detect carbon dioxide. After the first measurement of the original biogas composition, pure nitrogen with a fixed volume (10–200 ml, at least 10% of the original nitrogen volume) was injected into the gas bag and then nitrogen content was measured again. According to the nitrogen content increase after the injection of a fixed-volume pure nitrogen, the original biogas volume under the ambient temperature of 25 °C and pressure of 1 atm could be calculated based on a protocol described previously (Ambler and Logan, 2011). Biomass concentration in terms of ML(V)SS was measured according to the standard method of glass fiber membrane filtration followed by sequential drying at 105 °C and 550 °C. Microbial community analysis consisted of a series of procedures including deoxyribonucleic acid (DNA) extraction, polymerase chain reaction (PCR) amplification, next-generation sequencing and bioinformatics analysis according to the reported methods (Hong et al., 2012). Membrane permeability in terms of TMP and flux was directly measured via monitoring of pressure sensors and permeate flow, respectively.

Table 2
Operational parameters in all runs.

Run	Time (d)	Influent COD (mg/L)	HRT (h)	Volumetric OLR (gCOD/L/d)	Sludge CFV (m/s)	Biogas CFV (m/s)
1	0–21	400	12	0.8	0.2	0.1
2	21–35	400	6	1.6	0.2	0.1
3	35–54	750	6	3.0	0.2	0.2
4	54–63	1000	6	4.0	0.3	0.2
5	63–70	1750	12	3.5	0.2	0.2
6	70–89	2100	12	4.2	0.2	0.1
7	89–96	2600	12	5.2	0.2	0.1
8	96–102	3000	12	6.0	0.2	0.1
9	102–110	3500	12	7.0	0.2	0.1
10	110–116	4000	12	8.0	0.2	0.1
11	116–120	5000	12	10.0	0.2	0.1

3. Results and Discussion

3.1. Organics removal

From Fig. 2, a very steady and high COD removal (more than 95%, mostly around 98%) was achieved in all runs including Run 11 with the maximum volumetric OLR of 10 gCOD/L/d (i.e., equivalent HRT as short as 0.96 h). The permeate COD in most runs (Run 1–8) was below 50 mg/L, a common regulation for non-potable reused water (EPA and AID, 2012; MEP and AQSIQ, 2002).

From Table 3, permeate COD showed a gradual increase with both volumetric and sludge OLR while COD removal was not significantly affected. The maximum volumetric and sludge OLR for satisfying the permeate COD below 50 mg/L was 6 gCOD/L/d (i.e., the equivalent HRT of 1.6 h) and 0.63 gCOD/gMLVSS/d, respectively, which could be regarded as the sustainable OLR for the AnMBR system used in this study for treating synthetic municipal wastewater.

3.2. Biogas production

From Fig. 3a, the methane content in the biogas increased from 30% to 70% in Run 1, further increasing to 80% and 90% in Run 2 and 3, and was maintained at 80–90% from Run 4 and on, indicating change in the metabolic activity of methanogens. The CO_2 content in the biogas was very low (below 5%, even below 1%) in Runs 1–4 under low OLR (below 3–4 gCOD/L/d) and gradually increased with OLR in Runs 5–11, reaching a maximum of 22% under the maximum OLR of 10 gCOD/L/d. According to the MON (−1.2) of the synthetic municipal wastewater, the theoretical biogas composition should be 65% CH_4 and 35% CO_2 (Lier et al., 2008). Thus, the high CH_4 and low CO_2 content observed in the biogas may indicate a significant contribution from the pathway of CO_2 to CH_4 (i.e., $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$) in this study.

Furthermore, archaeal community analyses shown in Fig. 3b support this hypothesis. *Methanobacterium* made up, on average, over 75% of the archaeal community during all sampled runs, and exhibited an apparent negative correlation with the CO_2 fraction. Most of the currently known *Methanobacterium* species use H_2 and CO_2 as electron donor and acceptor, respectively, to produce

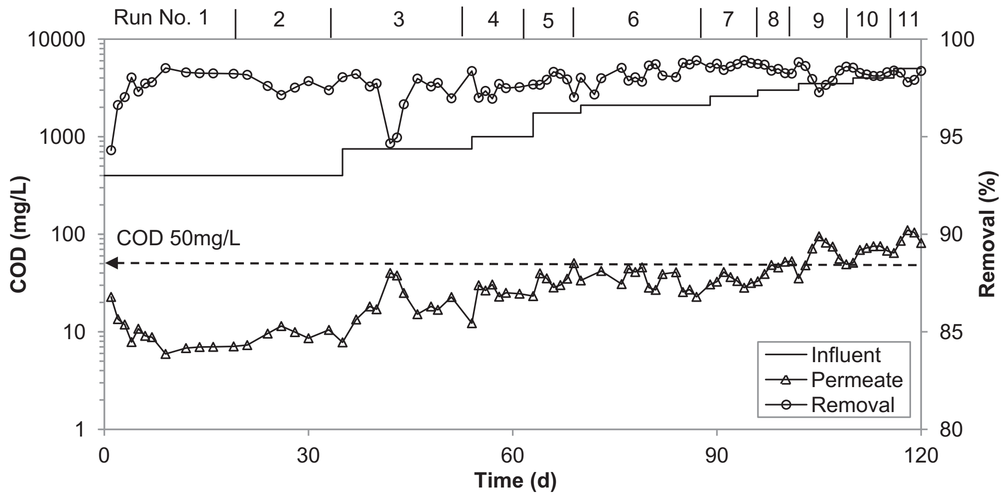

Fig. 2. COD removal in all runs.

Table 3
Summarized results of sludge OLR, permeate COD and COD removal in all runs.

Run	Sludge OLR (gCOD/gMLVSS/d)	Permeate COD (mg/L)	COD removal (%)
1	0.16 ± 0.012	8.5 ± 2.2	97.9 ± 0.6
2	0.29 ± 0.003	10.3 ± 0.1	97.4 ± 0.1
3	0.51 ± 0.018	19.0 ± 2.8	97.5 ± 0.4
4	0.63 ± 0.014	28.2 ± 4.1	97.2 ± 0.4
5	0.57 ± 0.019	36.6 ± 2.2	97.9 ± 0.1
6	0.63 ± 0.088	42.6 ± 5.3	98.2 ± 0.3
7	0.62 ± 0.001	33.2 ± 2.6	98.7 ± 0.1
8	0.70 ± 0.014	46.2 ± 1.0	98.5 ± 0.1
9	0.72 ± 0.072	67.9 ± 10.7	98.1 ± 0.3
10	0.70 ± 0.05	70.8 ± 1.4	98.2 ± 0.1
11	0.78 ± 0.028	95.7 ± 1.8	98.1 ± 0.1

methane gas (Whitman et al., 2001). Their high relative abundance within the archaeal community could account for the production of up to 90% methane content with very little CO_2 production for the majority of the operation period. The increase in the OLR from Run 5 to Run 11 also showed a strong negative correlation (R^2 of about 0.90, data shown in Fig. S1 in Supplementary Data) with the relative abundance of *Methanobacterium*. It would appear that the increase in the OLR was the primary cause for the decline in their relative abundance.

For each run in this study, the superficial methane production was defined as methane volume in biogas divided by total removed COD in liquid phase (both were from direct measurements) without considering COD conversion from biomass proliferation/decay and methane loss in permeate. According to COD mass balance, total removed COD consisted of two parts: part I converted to methane and part II converted to/from biomass proliferation/decay. Assuming a biomass COD conversion coefficient of 1.42 gCOD/gMLVSS (Rittmann and McCarty, 2001), the above-mentioned two parts could be calculated based on the MLVSS changes in each run. Thus, methane production in biogas could be defined as methane volume in biogas divided by part I of total removed COD in each run. Assuming a methane concentration in permeate of 15 ml/L (Hu and Stuckey, 2006), methane volume in permeate could be simply calculated in each run. Thus, real methane production could be defined as total methane volume (i.e., the sum of methane volume in biogas and permeate) divided by p part I of total removed COD in each run. Table 4 shows the methane production in all runs. From Runs 1 to 5 with stable relative abundance of *Methanobacterium* (75–95%), superficial methane production (84–368 ml/gCOD) showed good linearity (R^2 of about 0.90, data shown in Fig. S2 in Supplementary Data) with sludge OLR (0.16–0.63 gCOD/gMLVSS/d), indicating the enhanced potential of methanogens' methane conversion capacity with sludge OLR. A moderate linearity (R^2 of about 0.67, data shown in Fig. S2 in Supplementary Data) between superficial methane production (84–382 ml/gCOD) with sludge OLR (0.16–0.78 gCOD/gMLVSS/d) during all runs might be mainly due to the significant changes of archaeal community (i.e., decreasing abundance of the predominant *Methanobacterium* and increasing abundance of unclassified archaea) from Runs 6 to 11 (Fig. 3b). The methane production in biogas was 1–6% higher than the superficial methane production in all runs due to the positive contribution from the gradual biomass increase (i.e., one part of removed COD converted to biomass) during the whole operation (data shown in Fig. 4). The real methane production was further 1–3% higher than the methane production in biogas in most runs due to the positive contribution from methane dissolved in permeate. From the superficial (or real) methane production, the obtained values of 368 ± 34 (386 ± 35), 366 ± 12 (390 ± 12) and 382 ± 31 (396 ± 33) ml/gCOD in Runs 4, 5 and 11 were comparable to the theoretical value of 382 ml/gCOD, and were among the highest achievable methane production values demonstrated in all published data to the best of our knowledge. This high methane production could be due to high sludge OLR (>0.6 gCOD/gMLVSS/d) and a persistent presence of methanogenic community throughout the runs (i.e., either dominant hydrogen-utilizing methanogens in Run 4 and 5 or hydrogen-utilizing methanogens and unclassified archaea in Run 11). Based on the results in Table 4, a methane production in biogas (i.e., collectable methane for post-utilization) of around 300 ml/gCOD can be achieved under the sustainable OLR in this study.

3.3. Biomass evolution

Fig. 4 shows biomass changes during the whole operation. The ratio of MLVSS/MLSS was maintained at a stable range of 81–90% in all runs, indicating no apparent accumulation of inorganic solids, which is assumed to be mainly due to the little inorganic solids in the synthetic wastewater recipe. MLVSS showed a relatively low increase with time (0.027 g/L/d, R^2 = 0.90) from 4.9 g/L in Run 1 to 6.4 g/L in Run 4 under the low OLR (<4 gCOD/L/d). The decrease in Run 5 was due to biomass extraction for seeding another

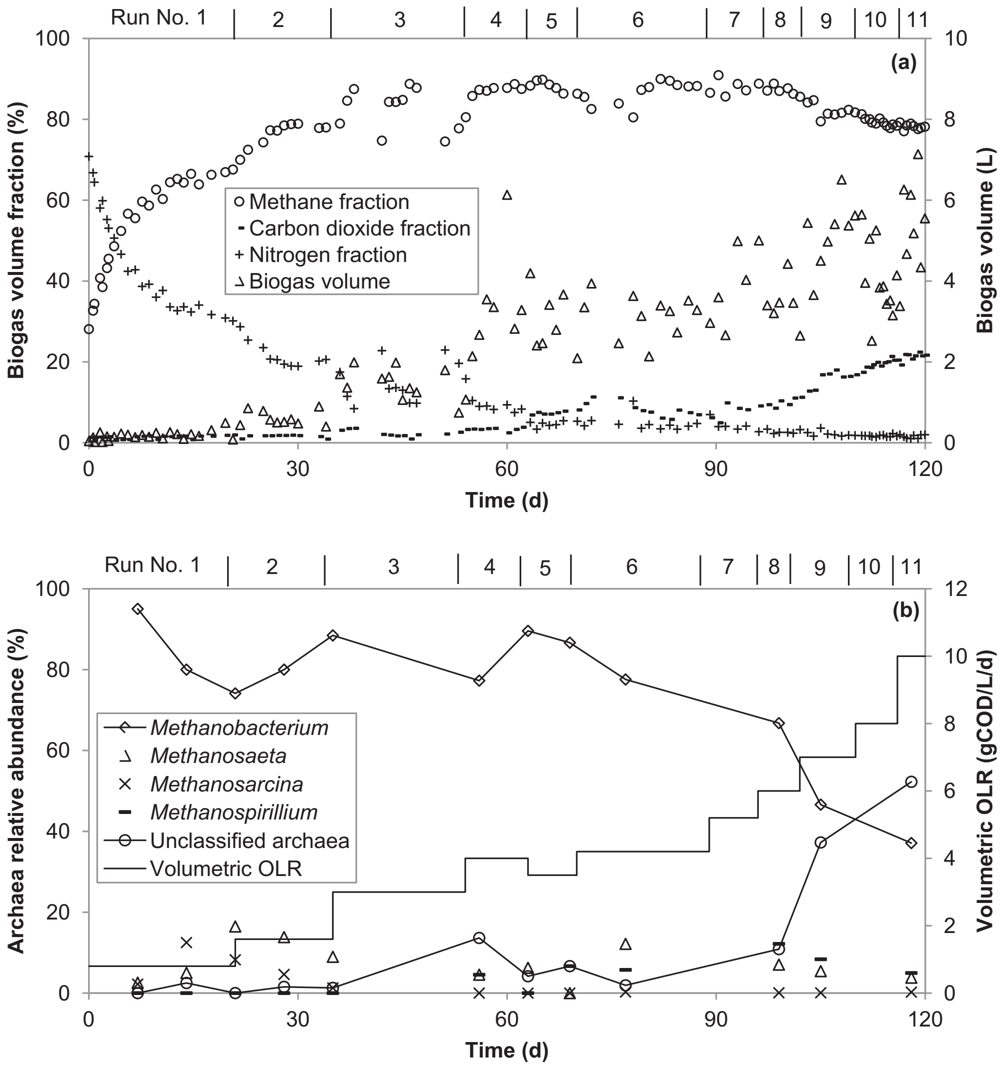

Fig. 3. Biogas production (a) and archaeal community (b) in all runs.

anaerobic reactor. A higher increase rate (0.16 g/L/d, $R^2 = 0.95$) from 5.9 g/L in Run 6 to 13.3 g/L in Run 11 was observed under the middle-high OLR of 5–10 gCOD/L/d. Based on MLVSS change and total COD removal from Runs 1–4 and Runs 6–11, the observed biomass yield (Y_{obs}) was calculated as 0.015 and 0.026 gMLVSS/gCOD, respectively, which is assumed related to the above-mentioned methanogenic community changes (Fig. 3b). These values are one order of magnitude lower than aerobic processes and also lower than values reported in other AnMBR studies (Hu and Stuckey, 2006; Yoo et al., 2012). In addition, there was nearly a constant MLVSS (5.6 g/L) maintained in Run 2 and thus the corresponding sludge OLR (around 0.29 gCOD/gMLVSS/d) might be regarded as the maintenance energy, which resulted in the balance of microbial growth and decay (i.e., zero biomass production).

According to the Monod equation describing the kinetics of substrate utilization (Eq. (1) and its variant Eq. (1')) and microbial growth (Eq. (2)), the kinetic constants (including maximum substrate utilization rate, k, half-maximum-rate concentration, K_s, true biomass yield, Y, decay rate, k_d, and maintenance energy, m) can be calculated through the linear correlation of Eq. (1') (Fig. S3a in Supplementary Data, for k and K_s) followed by Eq. (2) (Fig. S3b in Supplementary Data, for Y, k_d, m) using the data from each run (Rittmann and McCarty, 2001).

$$\frac{S_f - S_e}{\mathrm{HRT}} = \frac{kS_e}{K_s + S_e}\mathrm{MLVSS_a} \tag{1}$$

$$\frac{(\mathrm{HRT})\ (\mathrm{MLVSS})_a}{S_f - S_e} = \frac{K_s}{k}\frac{1}{S_e} + \frac{1}{k} \tag{1'}$$

$$\mu = \frac{(\mathrm{MLVSS_f} - \mathrm{MLVSS_i})}{(\mathrm{MLVSS_i})T} = Y\frac{kS_e}{K_s + S_e} - k_d = Y\left(\frac{kS_e}{K_s + S_e} - m\right) \tag{2}$$

where for each run, S_f = influent COD (mg/L), S_e = effluent COD (mg/L), HRT = hydraulic retention time (d), $\mathrm{MLVSS_a}$ = average MLVSS (g/L), $\mathrm{MLVSS_f}$ = final MLVSS (g/L), $\mathrm{MLVSS_i}$ = initial MLVSS (g/L), T = operation time (d), μ = net specific biomass growth rate (1/d).

Calculated values were k = 1.02 gCOD/gMLVSS/d and K_s = 25.4 mg/L ($R^2 = 0.94$, data shown in Fig. S3a in Supplementary Data). The k in this study was four times higher than reported value from an AnMBR treating synthetic municipal wastewater under 25 °C (Ho and Sung, 2009), which might explain the high COD removal (98%) under high OLR (up to 10 gCOD/L/d) in this study. The Y, k_d and m were 0.037 gMLVSS/gCOD, 0.009 d^{-1}, and 0.23 gCOD/gMLVSS/d ($R^2 = 0.65$, data shown in Fig. S3b in Supplementary Data), respectively. The relative low linear correlation was mainly due to the calculation errors from the left side of Eq. (2) and the

Table 4
Summarized results of methane production in all runs.

Run	Superficial methane production (without biomass related COD conversion and methane loss) (ml/gCOD)	Methane production in biogas (with biomass related COD conversion) (ml/gCOD)	Real methane production (with biomass related COD conversion and methane loss) (ml/gCOD)
1	84 ± 2	88 ± 3	129 ± 4
2	135 ± 7	136 ± 7	175 ± 6
3	220 ± 5	226 ± 3	246 ± 3
4	368 ± 34	373 ± 34	386 ± 35
5	366 ± 12	382 ± 12	390 ± 12
6	252 ± 50	265 ± 71	272 ± 70
7	227 ± 2	229 ± 2	234 ± 2
8	238 ± 9	241 ± 10	245 ± 10
9	290 ± 24	308 ± 25	312 ± 25
10	339 ± 9	355 ± 10	359 ± 10
11	382 ± 31	393 ± 33	396 ± 33

second substitution of k and K_s into the right side of Eq. (2). Y was close to the typical value for methanogens, while k_d was lower than typical values reported (Rittmann and McCarty, 2001). The m was also close to the experimental observation, indicating these kinetic constants were reasonable to describe the kinetics of the AnMBR in this study.

3.4. Membrane permeability

Fig. 5 shows membrane permeability measured during the whole operation. From Runs 1 and 5–11 with a flux of 6 L/m^2/h under combined sludge (0.2 m/s) and biogas (0.1 m/s) crossflow scouring, a consistent low TMP (<5 kPa) was maintained in these runs (about 3 months totally), with only a small rate of increase (<0.05 kPa/d), showing the characteristics of sub-critical (or sustainable) flux operation (e.g., critical flux was estimated around 10–12.5 L/m^2/h, data shown in Fig. S4 in Supplementary Data). For Runs 2–4 with a higher flux of around 10–12 L/m^2/h under combined sludge (0.2–0.3 m/s) and biogas (0.1–0.2 m/s) crossflow scouring, a more rapid rate of TMP increase was observed (>1.5 kPa/d) between two cleanings (i.e., enhanced crossflow scouring without permeation) and a gradual TMP increase from below 10 kPa in Run 2 to above 20 kPa in Run 4, indicating the characteristics of operation around critical flux. In addition, the good permeability recovery observed after enhanced crossflow scouring in Run 5 indicated that hydraulically reversible fouling (mainly cake layer) dominated in Run 2–4. Membrane fouling characteristics under sub-critical and around critical flux operation in this study were in agreement with both previous AnMBR (Martinez-Sosa et al., 2011; Robles et al., 2013) and aerobic MBR (Guglielmi et al., 2007; Wei et al., 2011) studies. The sub-critical flux of 6 L/m^2/h achieved in this lab-scale study was close to the reported value of 7 L/m^2/h in a pilot-scale AnMBR study (Martinez-Sosa et al., 2011) but lower than the reported value of 9–13 L/m^2/h in another pilot-scale AnMBR study (Robles et al., 2013). Although still lower than 20–30 L/m^2/h in aerobic MBR (Guglielmi et al., 2007; Wei et al., 2011), it would be possible to improve the sustainable flux in AnMBR through the further studies on enhancing crossflow scouring (Prieto et al., 2013), improving sludge filterability (Buntner et al., 2013), modifying membrane material and so on.

3.5. Energy recovery potential from municipal wastewater

Based on the results from this study, the energy recovery potential from low-strength municipal wastewater (e.g., COD 500 mg/L) through the AnMBR process may be dependent on several factors such as wastewater temperature, AnMBR operating conditions, and alternative pre-treatment processes. Wastewater temperature is normally the key factor determining the energy recovery due to the optimal mesophilic (35 °C) operation of AnMBR.

For a tropical arid area like Saudi Arabia, there is generally no need for pre-heating feed wastewater to maintain mesophilic AnMBR operation. Thus, the results generated from this study can be regarded as a reference value, which implies a methane production of 300 ml/gCOD under a sustainable sludge OLR of 0.6 gCOD/gMLVSS/d achieved by flexible control of HRT and MLVSS (e.g., HRT of 2 h and MLVSS of 10 g/L). The corresponding energy recovery is calculated as 1.57 kWh/m^3 based on a COD removal of 95% and a methane energy potential of 11 kWh/m^3 (McCarty et al., 2011).

However, for most temperate areas, where the temperature of raw municipal wastewater is usually 10–30 °C, pre-heating for a 5–25 °C increment is required for mesophilic AnMBR operation. Due to the large water specific heat of 1.16 kWh/m^3/°C and low COD of 500 mg/L in raw municipal wastewater, the energy recovery from methane production is usually far below the energy input for pre-heating. To overcome this problem, two solutions may be feasible.

One is to use heat pump technology for transfer of the heat energy from the mesophilic AnMBR permeate (35 °C) into the feed

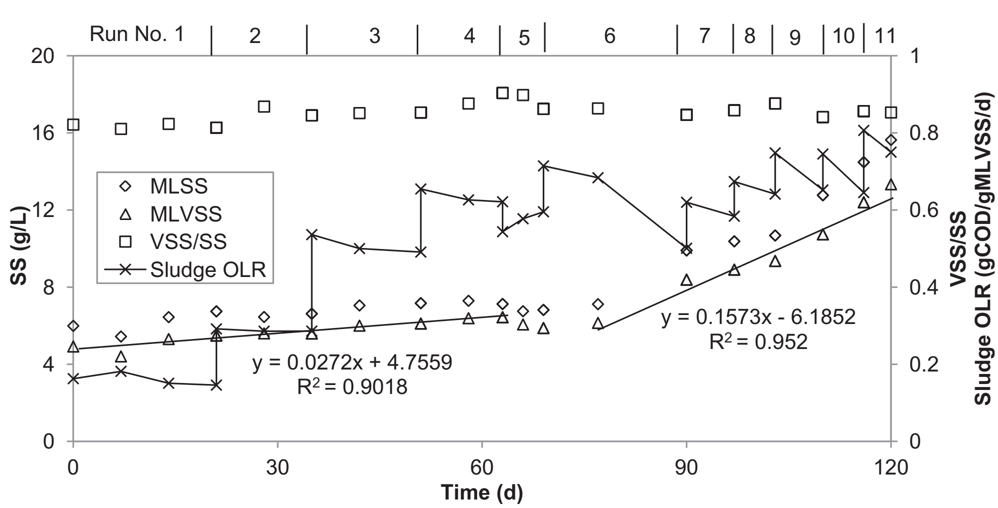

Fig. 4. Biomass characteristics in all runs.

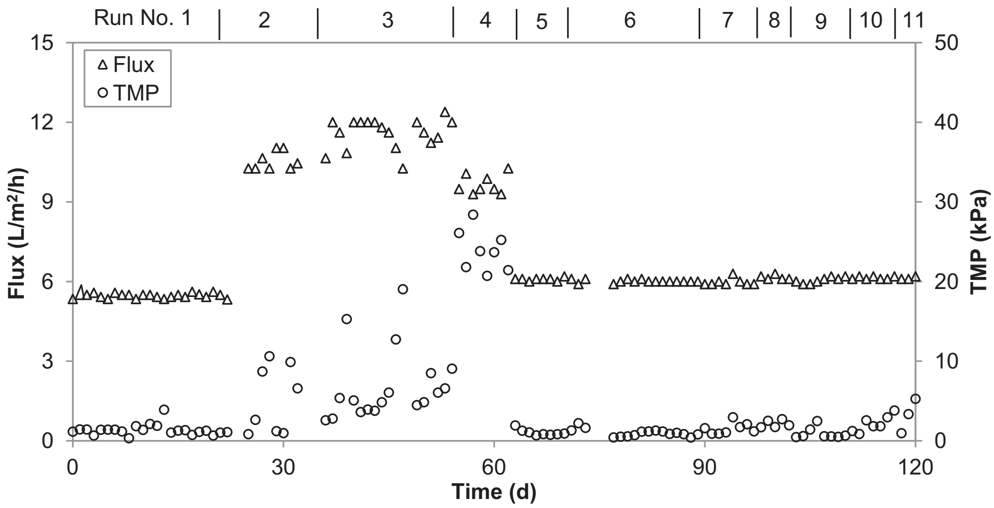

Fig. 5. Flux and TMP in all runs.

wastewater thus reducing net energy input for pre-heating, which has been used in WWTPs to recover heat energy from raw and treated wastewater for heating or cooling (Funamizu et al., 2001; McCarty et al., 2011; Mo and Zhang, 2013). A heat pump has a typical coefficient of performance of around 4, which means 4 units of transferred heat energy per unit of electrical energy consumed by the heat pump (Chua et al., 2010; Funamizu et al., 2001; Liu et al., 2014; McCarty et al., 2011; Mo and Zhang, 2013). Thus, the net energy input for pre-heating would be reduced by 75% using heat pump technology compared to direct pre-heating. In addition, the high AnMBR permeate quality (i.e., no suspended solids, low reductive organics concentration, very low microbial concentration) would greatly reduce the potential operational issues (e.g., blocking, scaling, corrosion) and the associated maintenance costs of a heat pump (Chua et al., 2010). In the proposed hybrid system (Fig. 6), a heat pump would use fresh mesophilic AnMBR permeate with a temperature of 35 °C as a heat source to preheat the feed wastewater with ambient temperature (e.g., 10–30 °C) to the target temperature of 35 °C. Under the ideal conditions, the permeate temperature would drop to the original feed temperature (e.g., 10–30 °C) while the feed temperature would rise to the target of 35 °C through the heat energy transfer from permeate into feed by the heat pump. In practice, the heat loss would be reduced to the minimum level through the shortest connection among permeate line, feed line, heat pump and mesophilic bioreactor.

The other is to use the emerging forward osmosis (FO) technology to concentrate raw municipal wastewater before feeding the AnMBR, which can increase feed COD and energy recovery per unit volume of wastewater as well as decrease the feed volume and pre-heating energy input. Ideally, FO extracts only water into a draw solution with a high osmotic gradient (e.g., seawater) from a feed solution (i.e., wastewater) while retaining all organics in the concentrated wastewater (Linares et al., 2013). The diluted draw solution may be discharged or reused for some purposes (e.g., salt-tolerant crops irrigation or some industrial use) and even for fresh water production through appropriate post-treatment such as low pressure reverse osmosis (LPRO). The concentrated wastewater would be the preferred feed for AnMBR to achieve the maximum net energy recovery.

Based on the integration of a heat pump and FO technology into mesophilic AnMBR for municipal wastewater treatment (Fig. 6), the projections of energy balance are presented in Table 5. Due to the large water specific heat capacity, direct pre-heating is not available to achieve a net energy recovery until the COD is up to

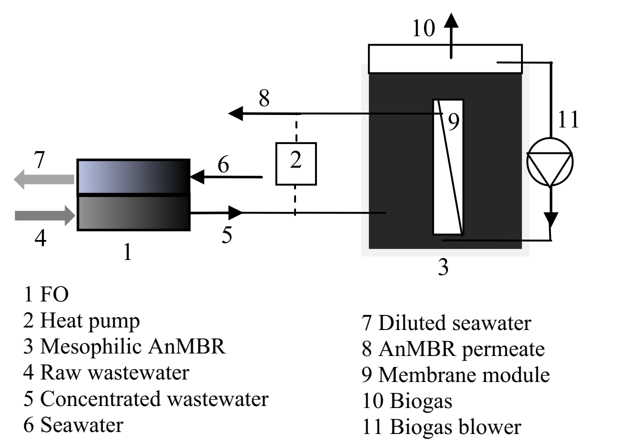

Fig. 6. Schematic diagram of proposed mesophilic AnMBR integrating heat pump and FO technology.

9500 mg/L (i.e., concentrating 19 times by FO) under the lowest temperature of 10 °C. After recycling pre-heating energy by a heat pump, the minimum COD to achieve net energy recovery under the lowest temperature of 10 °C is decreased to around 2500 mg/L (i.e., concentrating 5 times by FO), indicating the significant contribution to energy saving by a heat pump. The net energy recovery would be theoretically proportional to the feed COD (i.e., FO concentrating factor). Assuming the utilization of a heat pump and an average wastewater temperature of 10 °C (i.e., cold area), 20 °C (i.e., temperate area), and 30 °C (i.e., tropical area), the average net energy recovery from the mesophilic AnMBR process for a concentrated feed of 2500 mgCOD/L (i.e., FO concentrating factor of 5) is 0.55/3.46/6.38 kWh/m^3 and for a feed of 5000 mgCOD/L (i.e., FO concentrating factor of 10) is 8.38/11.30/14.22 kWh/m^3. The FO concentrating factor of 5–10 may be achievable because there is no theoretical limit due to the nature of the FO process. In any event, the optimal FO concentrating factor should be evaluated systemically for various parameters including salt leakage to feed solution, pollutant leakage to draw solution, membrane fouling, operational cost, etc.

From the above analysis, the pre-heating energy demand to maintain mesophilic AnMBR operation is the key obstacle limiting energy recovery potential. Besides the above-mentioned two approaches (heat pump and FO), another promising approach

Table 5
Energy balance projection of mesophilic (35 °C) AnMBR integrating heat pump and FO technology (raw COD 500 mg/L, COD removal 95%, methane production 300 ml/gCOD, methane energy potential 11 kWh/m^3, water specific heat 1.16 kWh/m^3/°C, coefficient of performance of heat pump 4).

Feed temperature (°C)	Heat energy consumption (kWh/m^3)		FO concentrating factor	Concentrated COD (mg/L)	Equivalent methane energy production (kWh/m^3)	Net energy recovery (kWh/m^3)	
	Direct heating	Heat pump utilization				Direct heating	Heat pump utilization
10	29.17	7.29	1	500	1.57	−27.60	−5.72
			5	2500	7.84	−21.33	0.55
			10	5000	15.68	−13.49	8.38
			20	10,000	31.35	2.183	24.06
20	17.50	4.38	1	500	1.57	−15.93	−2.81
			5	2500	7.84	−9.66	3.46
			10	5000	15.68	−1.83	11.30
			20	10,000	31.35	13.85	26.98
30	5.83	1.46	1	500	1.57	−4.27	0.11
			5	2500	7.84	2.00	6.38
			10	5000	15.68	9.84	14.22
			20	10,000	31.35	25.52	29.89

would be to adapt AnMBR to non-ideal fluctuating ambient temperature to remove the heating requirement (Shin et al., 2014). Although there would be a possible decrease of COD removal and methane production under ambient temperature, it is still possible to achieve good permeate and methane production through the optimal AnMBR operation, which should be further studied.

4. Conclusions

This study has demonstrated that mesophilic AnMBR is a promising technology for municipal wastewater treatment and reuse due to its good performance of high sustainable volumetric and sludge OLR (6 gCOD/L/d and 0.63 gCOD/gMLVSS/d), high methane production over 300 ml/gCOD (even approximate to the theoretical 382 ml/gCOD) and very low biomass production (0.015–0.026 gMLVSS/gCOD). Under the sustainable OLR, the equivalent energy recovery of 1.57 kWh per m^3 wastewater containing 500 mg/L COD could be achieved. Mesophilic AnMBR integrating heat pump and FO technology would be a promising alternative for net energy recovery from municipal wastewater, especially in a temperate area.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.biortech.2014.05.053.

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