Treating anaerobic effluents using forward osmosis for combined water purification and biogas production

Schneider, Carina; Rajmohan, Rajath Sathyadev; Zarebska, Agata; Tsapekos, Panagiotis; Hélix-Nielsen, Claus

Published in:
Science of the Total Environment

Link to article, DOI:
[10.1016/j.scitotenv.2018.08.036](https://doi.org/10.1016/j.scitotenv.2018.08.036)

Publication date:
2019

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):
Schneider, C., Rajmohan, R. S., Zarebska, A., Tsapekos, P., & Hélix-Nielsen, C. (2019). Treating anaerobic effluents using forward osmosis for combined water purification and biogas production. *Science of the Total Environment*, 647, 1021-1030. <https://doi.org/10.1016/j.scitotenv.2018.08.036>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Treating anaerobic effluents using forward osmosis for combined water purification and biogas production

Carina [Schneider](#)^a
Rajath Sathyadev [Rajmohan](#)^a
Agata [Zarebska](#)^a
Panagiotis [Tsapekos](#)^a
Claus [Hélix-Nielsen](#)^{a, b, *}
clhe@env.dtu.dk

^aTechnical University of Denmark, Department of Environmental Engineering, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark
^bUniversity of Maribor, Faculty of Chemistry and Chemical Engineering, Smetanova ulica 17, 2000 Maribor, Slovenia
*Corresponding author at: Department of Environmental Engineering, Technical University of Denmark, Bygningstorvet Bygning 115, DK-2800 Kgs. Lyngby, Denmark.

Editor: Zhen (Jason) He

Abstract

Forward osmosis (FO) can be used to reclaim nutrients and high-quality water from wastewater streams. This could potentially contribute towards relieving global water scarcity. Here we investigated the feasibility of extracting water from four real and four synthetic anaerobically digested effluents, using FO membranes. The goal of this study was to 1) evaluate FO membrane performance in terms of water flux and nutrient rejection 2) examine the methane yield that can be achieved and 3) analyse FO membrane fouling. Out of the four tested real anaerobically digested effluents, swine manure and potato starch wastewater achieved the highest combined average FO water flux (>3 liter per square meter per hour (LMH) with 0.66 M MgCl₂ as initial draw solution concentration) and methane yield (>300 mL CH₄ per gram of organic waste expressed as volatile solids (VS)).

Rejection of total ammonia nitrogen (TAN), total Kjeldahl nitrogen (TKN) and total phosphorous (TP) was high (up to 96.95%, 95.87% and 99.83%, respectively), resulting in low nutrient concentrations in the recovered water. Membrane autopsy revealed presence of organic and biological fouling on the FO membrane. However, no direct correlation between feed properties and methane yield and fouling potential was found, indicating that there is no inherent trade-off between high water flux and high methane production.

Keywords: Water reclamation; [Biogas production](#); [Fouling](#); [Anaerobic digestion](#); [Wastewater treatment](#)

1.1 Introduction

Access to safe and clean drinking water as well as sustainable energy is a basic human necessity, contributing to human health, poverty reduction and environmental sustainability ([Luo et al., 2016](#); [WWAP, 2015](#)).

Despite intense efforts in recent years to increase water supply, sanitation and hygiene for people in water-stressed areas, 663 million people still remain without access to drinking water sources and the population growth outpaces the progress made ([WWAP, 2015](#)). Water scarcity is not limited to developing and third world countries, but affects industrialized nations as well. This makes it one of the major challenges of this century and raises the need to develop new sources of water ([Shannon et al., 2008](#)).

In addition, the quest to provide sustainable energy sources to satisfy a rapidly increasing global energy consumption while alleviating climate change has yet to be solved ([McGinnis and Elimelech, 2008](#)).

One possible solution is the combined reclamation of water and energy from municipal or industrial wastewater sources ([Shannon et al., 2008](#)). In recent years, the perception of wastewater has changed. It is no longer considered as waste, but a resource of nutrients (N, P and K), water and energy in form of biogas ([Ansari et al., 2017](#); [Lutchmiah et al., 2011](#)).

Biogas can be produced via anaerobic digestion (AD), which converts complex organic matter mainly to methane and carbon dioxide. AD is widely used for the treatment of wastewater because it generates less sludge than

conventional aerobic processes and is also more cost-efficient, since aeration is not required and energy can be partly recovered by utilizing the produced biogas (Ansari et al., 2017). In recent years anaerobic membrane bioreactors (AnMBR) that combine biogas production with low-energy wastewater treatment using porous microfiltration (MF) or ultrafiltration membranes (UF) have raised increasing interest. The advantages of using AnMBR systems include improved effluent quality, lower sludge production and improved biogas yields by increasing the retention time of anaerobic microorganisms in the bioreactor (Gu et al., 2015; Wang et al., 2017).

However, conventional AnMBRs face some challenges that are rooted in their reliance on pressure-driven membrane processes and porous membranes (Stuckey, 2012). Less-readily biodegradable soluble organics, dissolved solids (Lay et al., 2010) and trace organic pollutants, such as pharmaceuticals and endocrine disrupting compounds (EDC) (Clara et al., 2005) are washed out through the pores of MF and UF membranes, lowering the effluent quality and negatively affecting the biogas yield (Gu et al., 2015). Further on, fouling results in rapid flux decline and reduces the overall performance (X. Wang et al., 2016). These problems could potentially be solved by replacing MF/UF membranes with tight forward osmosis (FO) membranes.

In FO, a draw solution (DS) is used to induce a net flow of water through a semipermeable membrane into the DS from a feed solution (FS). The flow is driven by the transmembrane osmotic pressure gradient $\Delta\pi$ between the DS and FS and will occur as long as $\pi_{DS} > \pi_{FS}$. The π_{DS} arises from the DS osmolyte where seawater, by-products from industrial processes, and inorganic salts (e.g. NaCl, $MgCl_2$) all have been evaluated in previous studies. This emerging membrane technology can be used to extract water from wastewater streams while efficiently retaining organic matter and microorganisms (York et al., 1999). FO membrane systems are able to treat complex wastewater streams of varying composition (Lutchmiah et al., 2014), such as landfill leachate (York et al., 1999), municipal wastewater (Hey et al., 2017; Z. Wang et al., 2016), or wastewater from oil and gas separations (Hey et al., 2017). The diluted DS from FO can be re-concentrated by reverse osmosis (RO) (Holloway et al., 2007) or membrane distillation (MD) (Liu et al., 2016), while simultaneously producing high quality water.

Taking all of these considerations together, the integration of FO membranes into an osmotic anaerobic membrane bioreactor (FO-AnMBR) can be seen as a promising technology for wastewater treatment, water reclamation and simultaneous biogas production (Chen et al., 2014; Gu et al., 2015; Li et al., 2017; Tang and Ng, 2014). However, this concept has to our knowledge so far not been tested for high-strength wastewater sources, such as agricultural wastewater and cattle manure.

In this study the membrane performance of a novel biomimetic FO flat sheet membrane is evaluated for the treatment of AD effluents. Eight types of effluents were selected: potato starch wastewater, swine manure and two types of cattle manure (thermophilic and mesophilic), as well as four effluents based on basal anaerobic medium (BA): synthetic sugars, synthetic lipids, synthetic proteins and synthetic mixture. The synthetic effluents were chosen in addition to the real effluents due to their known composition. This should help to find possible correlations between the effluent composition, biogas potential and fouling propensity.

The objective of the present study is to answer the following questions:

- 1) What is the FO membrane performance of the selected AD effluents, with regards to water flux and nutrient rejection?
- 2) What is the methane yield achieved by these wastewaters during AD? This aspect is especially important with respects to reduction of operational expenditure.
- 3) What is the extent and the nature of the fouling and how does the composition of the AD effluents affect the membrane fouling?

Taken together, the results from this study can be used towards the development of an integrated FO-AnMBR-MD/RO process, and will help to improve understanding regarding which types of wastewater can be treated successfully, providing a compromise between high biogas production, good FO-based water extraction and low fouling potential. The scope of the article is depicted in Figure 1.

Figure 1 Scope, process outcome and analyses in this study. Effluents from anaerobic digestion (AD) reactors were treated using forward osmosis (FO). The FO membrane performance of the effluents and the methane yield during anaerobic digestion were evaluated. Furthermore, fouled membranes were analysed to gain insight into fouling properties.

alt-text: Fig. 1

2.2 Material and methods

2.1.2.1 FO membrane

The thin film composite (TFC) flat sheet FO membranes used herein are Aquaporin Inside™ membranes provided by Aquaporin A/S, Denmark. They are composed of a polyethersulfone (PES) support layer and a polyamide active (PA) layer with incorporated Aquaporin proteins reconstituted in spherical polymer vesicles. (Habel et al., 2015; Zhao et al., 2012). Membrane thickness is 110 µm (±15 µm). The isoelectric point lies at approximately pH 2.9 and the zeta potential is between -80 mV and -90 mV at pH 7 (Singh et al., 2018).

2.2.2.2 Anaerobic digestion effluents

Effluents were collected from eight lab-scale bioreactors at steady state conditions and frozen immediately at -20 °C until used. The total amount of effluents collected, as well as additional information about the bioreactors, can be found in the Supplementary data. The anaerobic reactors were fed with four types of real wastewater (potato starch wastewater, swine manure, cattle manure from a thermophilic reactor and cattle manure from a mesophilic reactor) and four types of synthetic wastewater, based on BA medium (Angelidaki et al., 1990) supplemented with sugars (glucose), proteins (casein), lipids (glycerol trioleate, GTO) and a mixture of the aforementioned. To ensure that the collection of effluents was taken from a process operated at steady state conditions, the biogas production of bioreactors was measured daily via a water-displacement gas meter (Kougias et al., 2013) and other biochemical parameters (i.e. pH, volatile fatty acids (VFA), CH₄ content in the biogas (Tsapekos et al., 2017)) were monitored at least twice a week. The methane yield per gram volatile solids (VS) [mL CH₄/g VS] was daily calculated taking into consideration the biogas production [mL], the organic loading rate (OLR) [g VS/L] and CH₄ content [%] in the biogas:

$$\text{methane yield} = \frac{\text{biogas production} * \text{CH}_4 \text{ content}}{\text{OLR} * V_L} \tag{1}$$

where V_L is the reactor volume.

Further information about the bioreactors can be found in the Supplementary data and in previously published studies (Bassani et al., 2016; Mahdy et al., 2017; Tsapekos et al., 2017).

Prior to use, the effluents were unfrosted and sieved in three steps, using sieves with a mesh size of 1 mm, 250 µm and 125 µm. The characteristics of the effluents are given in Table 1.

Table 1 Effluent characteristics.

alt-text: Table 1

Real effluents		Synthetic effluents	

	Swine manure	Potato starch wastewater	Cattle manure, thermophilic	Cattle manure, mesophilic	BA medium + GTO	BA medium + glucose	BA medium + mixture	BA medium + casein
TOC [g/L]	1.51±0.01	1.37±0.12	4.55±0.00	7.11±0.05	1.95±0.15	1.42±0.15	1.18±0.01	0.90±0.48
TS [g/L]	7.73±0.00	8.68±0.35	6.51±3.18	8.27±0.26	7.06±0.02	18.55±0.04	6.72±0.30	21.81±0.07
VS [g/L]	3.21±0.33	1.82±1.57	4.26±2.84	3.7±0.25	3.53±1.83	10.91±0.99	4.15±2.92	15.14±6.81
TSS [g/L]	11.92±0.33	13.70±1.57	23.17±2.84	23.02±0.25	21.03±1.83	14.12±0.99	19.35±2.92	12.79±6.81
TKN [g/L]	1.68±0.01	1.19±0.09	3.34±0.00	2.20±0.00	5.97±0.01	4.84±0.01	3.80±0.00	2.69±0.32
TAN [g/L]	2.08±0.00	0.91±0.06	3.41±0.00	1.66±0.00	5.80±0.00	4.46±0.01	3.24±0.00	1.79±0.02
Ortho-P [g/L]	0.12±0.00	0.19±0.06	0.31±0.00	0.28±0.01	0.59±0.00	0.67±0.00	0.57±0.00	0.83±0.00
Density [kg/L]	1.00±0.00	1.00±0.00	1.02±0.00	1.01±0.00	1.00±0.00	1.00±0.00	1.00±0.00	1.01±0.00
Dynamic viscosity μ [mPa·s]	1.35±0.00	1.15±0.00	2.00±0.00		1.39±0.00	1.44±0.00	1.24±0.00	1.45±0.00
Kinematic viscosity [mm²/s]	1.35±0.00	1.15±0.00	2.00±0.00		1.37±0.02	1.44±0.00	1.23±0.02	1.43±0.00

^a Viscosity could not be measured due to high TS content.

2.3.2.3 Forward osmosis experimental setup

All FO experiments were conducted in a lab-scale FO cross-flow setup with the active layer facing the feed solution (AL-FS). The membranes were pre-soaked in MilliQ water for 45 min before each FO experiment.

A schematic representation of the setup can be found in the Supplementary material.

The flat sheet module was manufactured by Mikrolab Aarhus A/S (Denmark) and consisted of two symmetrical flow chambers with the dimensions 175 mm (length), 80 mm (width) and 1.3 mm (height) and an effective membrane area of 140 cm².

Feed and draw solution were recirculated by gearing pumps (Longer Precision Pump Co., Ltd^a, China) at 500 mL/min, corresponding to a crossflow velocity of 8 cm/s. A draw-side mesh spacer was used to facilitate stirring close to the membrane surface.

As a feed solution, 2 L of anaerobic digestion effluents were used. The pH was adjusted to 6.7 with H₂SO₄ (6 M) to improve total ammonia nitrogen (TAN) retention by reducing the fraction of TAN present as NH₃ (Camilleri-Rumbau et al., 2015). For the draw solution, 4 L of 0.66 M MgCl₂ (analytical grade, Sigma-Aldrich, USA), was prepared by dissolving the salt in MilliQ water.

The concentration of the draw solute was chosen using the van't Hoff equation for dilute and ideal solutions in order to yield a theoretical osmotic pressure of 49 bar.

The feed solution bottle was placed on a balance (Kern, Germany) and the draw solution was mixed continuously to avoid the formation of a concentration gradient inside the draw solution bottle due to returning the diluted draw solution.

The water flux J_w [L m⁻² h⁻¹] was determined by recording the weight decrease of the feed solution, based on [equation Eq. \(2\)](#)

$$J_w = \frac{(m_{t2} - m_{t1})}{A_m \Delta t \rho}$$

(2)

where A_m is the membrane surface in m², ρ the density of the feed [kg m⁻³] and m the mass of the feed solution [kg] recorded at the times $t1$ and $t2$ (Bowden et al., 2012). Weight data was logged automatically every [five minutes5 min](#) throughout the entire duration of the experiments.

The concentration of the draw solution was not kept constant, meaning it decreased over time due to the water transport from feed to draw solution.

The reverse salt flux J_s was calculated using [equation Eq. \(3\)](#):

$$J_{s,Cl}/J_{s,Mg} = \frac{(V_{t2}\beta_{t2} - V_{t1}\beta_{t1})}{A\Delta t} \tag{3}$$

With β_{t1} and β_{t2} as the respective Mg^{2+} and Cl^{-} concentrations in the feed solution [g/L] and V_{t1} and V_{t2} as the volume of the feed solution [L] at $t1$ (beginning of the experiment) and $t2$ (end of the experiment).

Nutrient rejection is calculated based on the feed side mass balance as

$$R = \left(\frac{V_{t1}c_{t1} - V_{t2}c_{t2}}{V_{t1}c_{t1}} \right) \times 100\% \tag{4}$$

With c_{t2} and c_{t1} as the respective TAN, TN and TP^{-} concentrations in the feed solution [mol/L] at $t1$ (beginning of the experiment) and $t2$ (end of the experiment).

The duration of the experiments was 24 h to allow formation of a fouling layer.

2.4.2.4 Analytical methods

The AD effluents and the draw solution were analysed at the beginning and at the end of each FO experiment, namely: Total solids (TS), volatile solids (VS) and total suspended solids (TSS) were determined according to the [APHA standard methods \(2005\)](#). Total Kjeldahl Nitrogen (TKN) and total ammonia nitrogen (TAN) were analysed using a Kjeldahl distillation unit (FOSS, Denmark). Total organic carbon (TOC) was determined with a TOC analyser (LECO, USA). Orthophosphate-P and Cl^{-} concentrations in the feed were analysed using ion chromatography (IC) (Thermo Fisher Scientific, USA). Mg^{2+} concentrations in the feed were analysed with, inductively coupled plasma optical emission spectrometry (ICP-OES). Viscosity and density of the samples was measured using a viscosity meter (Anton Paar, Austria).

2.5.2.5 Membrane autopsy for analysis of the fouled membrane

Membrane autopsy methods include scanning electron microscopy (SEM) to determine the overall morphology of the fouling layer, energy-dispersive X-ray spectroscopy (SEM-EDS) to analyse the elemental composition of the fouling layer, Fourier-transform infrared spectroscopy (FTIR) to identify organic foulants, Ion chromatography (IC) for the determination anionic foulants, Inductively coupled plasma optical emission spectrometry (ICP-OES) for the determination cationic foulants and Adenosine Triphosphate (ATP) analysis to analyse biological fouling.

Before SEM and SEM-EDS (FEI, USA), fouled membrane coupons were soaked in 2.5% glutaraldehyde for 1 h (Sigma-Aldrich, USA) to fixate bacteria on the membrane surface, followed by dehydration in an ethanol dilution series (50% (v/v), 60% (v/v), 70% (v/v), 80% (v/v), 100% (v/v)) by immersion in for 5 min each. The samples were kept at 4 °C until analysis. Both fouled and virgin membrane coupons were sputter coated with gold ([Morrow et al., 2017](#)). SEM samples were analysed at a 10 mm working distance, at an acceleration voltage of 10.0 kV and a spot size of 3.0.

For FTIR, fouled and virgin membrane coupons were analysed in an attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectrometer (PerkinElmer, USA), equipped with a diamond crystal. Four scans per measurement point were collected.

To determine the inorganic components on the fouling layer, the foulants were extracted from a membrane coupon of the size 3.5 cm ~~✖~~ 3.5 cm. The fouled membrane coupons were immersed in MilliQ water (for IC) or 0.8 M HNO_3 (for ICP-OES) for 24 h followed by ultrasonification for 180 min. The extracts were kept at 4 °C until analysis. An ICP-OES scan included Ba, Ca, Fe, K, Mg, Mn, Na, P, S, Si, Sr and Zn was carried out using a Vista MPX spectrometer (Varian, USA). IC analysis included chloride, bromide, nitrate and sulfate was analysed on a Thermoscientific DIONEX AS-AP (Thermo Fisher Scientific, USA).

Prior to ATP analysis, the biomass was extracted from membrane coupons of the size 5cmx5cm defined size by immersing them in MilliQ water, followed by ultrasonification for 5 min in order to keep the microbial cells intact. The samples were kept at ~~-~~80 °C until analysis. To determine the ATP concentration, a luciferase assay was used, following the procedure described by [Vang et al. \(2014\)](#). In short, to determine the total ATP, the samples were lysed before adding luciferase. For free ATP, no cell lysis was conducted and luciferase was added directly. Microbial ATP was calculated as the difference between free ATP concentrations and total ATP concentrations. Bioluminescence was measured in a luminometer (Charles River, USA) and microbial ATP was determined indirectly by subtracting free (extracellular) ATP from total ATP.

3.3 Results and Discussion

3.1.3.1 FO performance and methane yield of AD effluents

J_w for all tested AD effluents is displayed in [Figure 2](#). Since the draw solution concentration was diluted over time, data is displayed as a function of feed recovery. This way, it is possible to compare flux decline at a given level of draw solution dilution ([Blandin et al., 2016](#)) The duration of the experiments was 24 h each.

Figure 2 Fig. 2 Development of J_w as a function of feed recovery of anaerobic digestion effluents. Each sample point in the curves represents the mean value of two experiments.

alt-text: Fig. 2

The initial J_w ranges from 4.3–4.8 LMH for the real effluents and from 4.3–5.1 LMH for the synthetic effluents. Even though the initial J_w for all effluents was within a comparable range, decline over time differs substantially. While potato starch wastewater and swine manure reached 30% feed recovery within 12 h, thermophilic cattle manure and mesophilic cattle manure required 16 h and 21 h, respectively. As the impact of the draw solution dilution is the same at this point, the differences can be attributed to fouling and ICP effects (Blandin et al., 2016). Further on, differences in reverse salt flux of draw solution ions, reducing the osmotic pressure difference over the membrane, could influence the results as well. However, there seems to be no direct correlation between flux decline and $J_{s,Mg}$ or $J_{s,Cl}$.

As can be seen in Table 2, $J_{s,Cl}$ exceeds $J_{s,Mg}$ substantially. This is in agreement with previous studies, that found that is mainly governed by the anion transport rather than the paired cations (Achilli et al., 2010; Coday et al., 2013).

Table 2 FO performance. $J_{w,ave}$ was calculated as a mean value of J_w , during the entire duration of the experiment. Flux decline is calculated over 24 h.

alt-text: Table 2

	Real effluents				Synthetic effluents			
	Swine manure	Potato starch wastewater	Cattle manure, thermophilic	Cattle manure, mesophilic	BA medium + GTO	BA medium + glucose	BA medium + mixture	BA medium + casein
$J_{w,ave}$ [L/(m ² ·h)]	3.3±0.10	3.0±0.22	2.4±0.01	1.9±0.17	3.1±0.07	2.9±0.38	2.9±0.15	2.4±0.63
Flux decline [%]	40.67±10.37	61.58±2.91	59.92±10.50	76.16±4.85	66.12±4.03	56.09±5.53	71.03±0.94	74.88±19.81
$J_{s,Mg}$ [g/(m ² ·h)]	0.09±0.02	0.31±0.06	0.62±0.02	0.41±0.05	-0.08±0.00	-0.10±0.01	0.10±0.00	0.01±0.01
$J_{s,Cl}$ [g/(m ² ·h)]	2.41±1.02	1.09±0.06	2.46±0.15	0.55±0.26	2.17±0.42	1.03±0.78	2.25±1.50	1.26±1.24

The differences in FO performance are listed in Table 2 and compared with effluent properties, such TOC, TS and dynamic viscosity (see Table 1). An increase in viscosity of the feed solution can lead to decreased water diffusivity over the membrane and thus decreased J_w (McCutcheon and Elimelech, 2006; Phuntsho et al., 2012).

Flux decline can also be caused by the formation of an organic fouling layer on the membrane, leading to increased hydraulic resistance (Lee et al., 2010; Parida and Ng, 2013). Organic foulants are often measured as TOC, therefore the organic matter content of each tested effluents was assessed.

In these experiments, $J_{w,ave}$ of the real effluent, except for potato starch wastewater, generally declined with increasing dynamic viscosity and TOC content. The synthetic effluents, on the other hand, showed the opposite trend. The highest average J_w of 3.3 LMH was achieved for swine manure, which has a dynamic viscosity of 1.35 mPa·s, closely followed by potato starch wastewater with an average J_w of 3.0 and a dynamic viscosity of 1.1 mPa·s. Overall, no clear correlation between feed characteristics and FO performance was found.

In order to decide on a suitable wastewater source for the use in an osmotic anaerobic bioreactor, both membrane performance (in terms of J_w) and methane yield must be considered to yield an economically viable process (Figure 3). Overall, the synthetic BA medium effluents achieve a higher methane yield, because their macro- and micronutrients have been optimised for biogas production. GTO gives a high methane yield, since lipids are very energy-

rich, while glucose is easily processed by AD, making it a very efficient substrate.

Figure 3 Fig. 3 Methane yield in comparison to the average J_w over a time period of 24 h.

alt-text: Fig. 3

Cattle manure and swine manure, on the other hand, contain lignocellulosic material which is recalcitrant to AD (Tsapekos et al., 2015). The swine manure has been sieved before AD, thus removing large parts of the lignocellulosic material and achieving a higher methane yield than cattle manure. However, the methane yield is also influenced by differences in the operation conditions of the bioreactors, such as HRT and OLR (see Supplementary data).

Out of the real effluents, potato starch wastewater and swine manure achieve both a high J_w and higher methane yield, combined with a moderate flux decline, making them interesting wastewaters candidates for the use in an osmotic anaerobic bioreactor to produce clean water and energy in the form of methane. Unsurprisingly, the methane yield of the synthetic effluents exceeds that of the real effluents, as BA medium has been specifically optimised for AD.

3.2.3.2 Nutrient rejection

The TFC membrane achieves remarkable results in terms of TAN, TKN and orthophosphate rejection, as shown in Table 3. Rejections were calculated using the initial and final concentrations in the feed solution.

Table 3 TAN, TKN and orthophosphate rejection.

alt-text: Table 3

	Rejection [%]		
	TAN	TKN	Orthophosphate-P
Swine manure	92.34	88.62	98.74
Potato starch wastewater	85.53	82.95	99.18
Cattle manure, thermophilic	96.95	93.83	99.56
Cattle manure, mesophilic	95.34	95.87	99.52
BA medium + GTO	81.25	81.08	99.74
BA medium + glucose	96.55	82.84	99.78
BA medium + mixture	80.76	68.69	99.74
BA medium + casein	85.38	81.84	99.83

The orthophosphate rejection is consistently high for all effluents (>99%), which is consistent with previous results for TFC membranes (Xue et al., 2015). Xue et al. (2015) found that TAN was not effectively retained. This was not observed in this study. Although rejection of TAN and TKN is lower than the orthophosphate rejection, TAN rejection ranges from 80.76% (in BA medium + mixture) up to 96.95% (in thermophilic cattle manure) and TKN rejection ranges from 68.69% (in BA medium + mixture) up to 95.87% (in thermophilic cattle manure). The similar TKN and TAN rejections can be explained by the fact that TAN made up between 66 and 100% of the measured TKN content in

all effluents. In semipermeable membranes, ion rejection is governed by electrostatic repulsion between ions and the membrane as well as steric hindrance depending on the hydration radii of the ions (Cornelissen et al., 2008; Xue et al., 2015). Since the experiments were carried out at pH 6.7 and the isoelectric point of the membrane is at pH 2.9, the membrane is negatively charged. The higher orthophosphate rejection could therefore be explained by the negative charge of the ion being repulsed by the membrane, while the positively charged ammonia ion faces no such repulsion. Additionally, the dynamic hydrated radius of orthophosphate is larger than that of ammonia (0.3 nm and 0.1 nm, respectively) (Kiriukhin and Collins, 2002).

Another possible explanation for the high TAN rejection is the use of $MgCl_2$ as a draw solution: Hu et al. attributed this to the Donnan equilibrium (Hu et al., 2017): Reverse salt diffusion of Cl^- ions exceeds that of Mg^{2+} ions (Table 2). This causes a charge imbalance and is prompting anions to diffuse from the feed solution to the draw solution in order to restore the charge equilibrium and leads to an accumulation of NH_4-N in the feed.

3.3.3.3 Characterisation of FO fouling layer

Organic fouling was analysed by comparing ATR-FTIR spectra of the clean and fouled active layers of the membranes (Figure 4). The spectrum of the clean active layer shows characteristic bands of polyamide at 1658 and 1578 cm^{-1} , which can be associated with amide I (C

O stretching) and amide II (N

H) respectively (Hey et al., 2016; Melián-Martel et al., 2012), whereas the bands at 1486 cm^{-1} (CH_3

C

CH_3 stretching), 1322 cm^{-1} (C

SO_2

C asymmetric stretching), 1298 cm^{-1} (S

O stretching), 1242 cm^{-1} (C

O

C stretching), 1151 cm^{-1} (C

SO_2

C symmetric stretching), as well as 1106 cm^{-1} and 872 cm^{-1} (skeletal aliphatic C

C/aromatic hydrogen bending, rocking), 836 cm^{-1} and 719 cm^{-1} (aliphatic C

H rocking) can be ascribed to the porous PES support layer, because the penetration depth samples both active and support layer (Singh et al., 2006).

Figure 4 Fig. 4 FTIR of FO membranes fouled with effluents from anaerobic digestion over a time period of 24 h

alt-text: Fig. 4

The distinct peaks of the clean membrane attenuated probably due to the presence of organic and/or biological fouling deposits on the membrane surface. All fouled membranes except the one exposed to potato starch wastewater sample exhibit a broad, distinctive peak at 3241–3290 cm⁻¹, which can be attributed to O–H stretching of hydroxyl groups or N–H stretching of amide A (Xu et al., 2016; Zarebska et al., 2014). The broadness of this peak suggests presence of polysaccharides, i.e. CH and OH groups.

Furthermore, there are smaller C–H peaks at 2917–2920 and 2847–2858 cm⁻¹. These peaks can be assigned to aliphatic compounds, lipids and aldehydes (Bell et al., 2017; Chang et al., 2011; Provenzano et al., 2014).

The peaks at 1627–1638 cm⁻¹ is present in all fouled membrane samples and can be attributed to amide I C=O stretching vibrations, indicating the presence of proteins in the fouling layer (Bell et al., 2016; Hashim et al., 2010). Controversially, Bell and colleagues attributed a peak at 1625 cm⁻¹ to a C–C stretch of aromatics (Bell et al., 2017). In principle, aromatic groups in manure-based effluents could stem from lignin breakdown of lignocellulosic materials in the animals feed. However, the BA medium based effluents contain no lignin-based material, thus the band is likely to be associated with amide I.

The small peak at 1429–1432 cm⁻¹ can be attributed to amide II (N–H stretching), while another small peak at 981–1022 cm⁻¹ due to C–O stretching corresponds to polysaccharides or polysaccharide-like substances (Chang et al., 2011; Melián-Martel et al., 2012; Provenzano et al., 2014).

ATP concentrations are often used as a tool in membrane autopsy as an indicator of biofouling and to assess the cellular viability of the biomass. After 24 h of FO experiment duration, there was a visible biofilm formation on the feed side of the membrane, while no sign of fouling was visible on the draw side. The ATP concentrations of the extracted foulants are shown in Table 4. Some attempts have been made to link ATP concentrations to total cell numbers (Hammes et al., 2010; van der Wielen and van der Kooij, 2010). However as the amount of ATP per cell may vary (Liu et al., 2013), the reliability of these methods is still debated and therefore, the results will be presented as microbial ATP concentrations.

Table 4 ATP concentration of biomass extracted from membrane surface

alt-text: Table 4

	Total ATP	Free ATP	Microbial ATP
	[ng] ATP/[cm ²] membrane	[ng] ATP/[cm ²] membrane	[ng] ATP/[cm ²] membrane
Swine manure	34.30 ± 9.56	1.08 ± 1.19	33.22 ± 8.37
Potato starch wastewater	1.40 ± 0.34	0.13 ± 0.08	1.46 ± 0.15
Cattle manure (thermophilic)	17.18 ± 11.56	0.15 ± 0.02	17.03 ± 11.54
Cattle manure (mesophilic)	6.54 ± 0.75	0.70 ± 0.03	5.84 ± 0.72
BA medium + GTO	149.96 ± 23.92	1.11 ± 0.14	148.85 ± 24.06
BA medium + glucose	N/A	N/A	N/A
BA medium + mixture	10.52 ± 0.47	0.34 ± 0.21	10.37 ± 0.96

BA medium + casein	1.91±1.54	0.17±0.17	1.74±1.37
--------------------	-----------	-----------	-----------

Free ATP is one to two orders of magnitude lower in comparison to total ATP, which is in agreement with previous results (van der Kooij, 1992; van der Wielen and van der Kooij, 2010). Overall, the variation of microbial ATP concentrations on the membrane surface differs significantly between the AD effluents. The highest microbial ATP concentration is found with high-lipid medium (BA medium + GTO) with 165.87 ng ATP/cm² membrane, which also had the highest methane yield, while high-protein medium (BA medium + casein) has the lowest microbial ATP concentrations with 0.77 ng ATP/cm² membrane. The differences in microbial ATP concentrations also reflect differences in the microbial viabilities in the bioreactors, which were operated at different conditions in terms of reactor type, OLR and HRT (see Supplementary data), containing different microbial communities with different amounts of ATP/cell. Furthermore, effluents were collected over a period of several weeks in order to have sufficient feed volumes, which may have affected the microbial viability as well.

The fouled and clean FO membranes were visually examined using SEM (Figure 5). The results show that, after 24 h of FO operation, the active layer of the pristine membrane (Figure 5-I) is entirely covered by a deposited cake layer in all fouled samples, which as revealed by FTIR contains proteins and carbohydrates.

Figure 5 SEM of FO membranes active layer fouled with effluents from anaerobic digestion over a time period of 24 h (A) swine manure, (B) potato starch wastewater, (C) cattle manure (thermophilic), (D) cattle manure (mesophilic), (E) BA medium + GTO, (F) BA medium + casein, (G) BA medium + glucose, (H) BA medium + mix, (I) clean membrane (AL)

alt-text: Fig. 5

In some cases, singular (Figure 5-A, C, E, F, G) or clustered (Figure 5-H) rod-shaped microbial structures can be seen incorporated in the fouling layer, suggesting that biological fouling occurred in agreement with ATP measurements. Both ICP-OES and IC analysis (Table 5) reveal the presence of Mg and Cl ions in the fouling layer. The difference between findings from SEM and ICP-OES and IC can be attributed to the fact the first method is surface sensitive, whereas the latter method analyses the foulants extract. Although Mg was already present in the effluents (see Supplementary data), the relative concentration in comparison with other divalent cations seemed to increase. This can be explained by reverse salt flux deposits on the membrane active layer surface. Divalent cations like Mg²⁺ and Ca²⁺ have been shown to promote organic fouling by forming complexes carboxylic groups of natural organic matter, thus helping to form intermolecular bridges (Mi and Elimelech, 2008).

Table 5 ICP-OES and IC analysis cation concentration in foulants extracted from membrane surface in [g* kg⁻¹ membrane]. For ICP-OES, results of Ba, Mn and Sr are omitted due to very low concentrations. For IC, results for nitrate and bromide are not displayed, as they could not be detected in the samples.

alt-text: Table 5

	Concentration [g* kg ⁻¹ membrane]	Swine manure	Potato starch wastewater	Cattle manure (thermophilic)	Cattle manure (mesophilic)	BA medium + GTO	BA medium + glucose	BA medium + mixture	BA medium + casein
ICP-OES	Ca	3.93±2.62	1.25±1.08	4.26±0.06	4.22±1.76	2.84±0.86	4.84±2.11	2.07±0.34	2.08±1.21
	Fe	0.76±0.53	0.18±0.13	0.45±0.02	0.44±0.24	0.84±0.13	0.60±0.19	0.35±0.04	0.32±0.05
	K	2.97±3.07	13.62±0.52	3.51±0.98	2.82±1.51	3.92±1.72	4.87±1.93	3.35±2.14	1.17±0.62
	Mg	19.75±20.44	34.11±2.59	12.82±4.94	9.3±5.23	36.04±10.32	28.89±8.37	33.54±14.34	32.52±5.85
	Na	1.16±1.20	1.69±0.18	1.43±0.36	1.17±0.56	1.75±1.15	2.87±1.28	2.18±1.69	1.89±1.56
	P	25.10±27.72	32.83±13.58	15.83±6.19	11.46±8.83	59.11±16.41	32.40±11.11	33.13±11.39	43.12±23.09
	S	2.59±2.63	2.91±0.87	0.62±0.08	0.56±0.06	1.05±0.63	2.12±1.66	1.71±1.30	2.75±2.08
	Si	0.29±0.19	0.18±0.04	0.24±0.04	0.22±0.16	0.30±0.06	0.30±0.11	0.18±0.03	0.12±0.06
	Zn	2.38±1.57	0.16±0.10	0.13±0.03	0.10±0.09	0.16±0.04	0.16±0.07	0.11±0.01	0.07±0.04
IC	Cl ⁻	2.97±2.70	5.19±3.45	7.97±0.53	5.37±0.22	3.49±2.10	3.65±0.01	6.03±0.38	4.96±0.24
	SO ₄ ²⁻	0.14±0.09	0.15±0.07	1.07±0.08	0.30±0.04	0.09±0.05	0.13±0.05	0.23±0.04	0.43±0.11

SEM-EDS was used to analyse the overall elemental composition of the fouling layer (Table 6). Asides from C, N and O, the main elements in the fouling layers are Mg, S, P and Ca. These results are in agreement with the results from ICP-OES analysis and indicate the presence of magnesium ions on the active layer due to the reverse salt flux. The P could stem from the rejected orthophosphate in the feed accumulating on the membrane surface. The presence of N and P on the fouled membrane suggests biofouling and the presence of extracellular polymeric substances (EPS), which is confirmed by ATP analysis.

Table 6 Representative SEM-EDS results of FO membranes fouled with effluents from anaerobic digestion in weight %.

alt-text: Table 6

Weight %	Swine manure	Potato starch wastewater	Cattle manure thermophilic	Cattle manure mesophilic	BA medium + GTO	BA medium + glucose	BA medium + mix	BA medium + casein
C			66.20	64.99	42.91	54.63	69.61	59.51
N	12.83	11.89	7.42	7.11	0.91	4.85	6.67	5.49
O	62.15	35.54	25.16	24.05	40.51	25.63	13.11	19.84
Na								0.09
Mg	5.03	16.21	1.21	0.72	6.09	6.29	0.49	1.59
Al	0.69			0.34		0.09	0.14	0.59
Si	1.73	1.14		1.44		0.15	0.41	2.48
P	4.89	20.80		0.38	6.45	7.35	0.60	2.16
S	2.11	5.48		0.58	2.85	0.17	8.07	2.17
K		1.58		0.05	0.11	0.09		0.12
Ca	5.74	1.98		0.32	0.06	0.56	0.48	4.56

Fe					0.10	0.19	0.15	0.89
Cu							0.25	0.50

The trace amounts of Cu, Al, and Zn found in the membranes fouled with synthetic effluents can be attributed to the trace element solution used in the preparation of the BA medium, while the S could stem from the thiamine and ~~thiotic~~ ~~thioctic~~ acid in the vitamin solution (Angelidaki et al., 1990).

The small amounts of gold found in some samples originate from sputter coating the samples before SEM/SEM-EDS. The SEM-EDS results are not directly comparable with IC and ICP-OES analysis, since it's possible that the foulant layer composition is inconsistent over the membrane area. The complete SEM-EDS spectra can be found in the Supplementary data.

4.4 Conclusions

This study provides an initial feasibility assessment for the treatment of various types of anaerobic digestion effluents by FO membranes, resulting in reclaimed water and methane production. Overall, the membranes showed reasonable initial J_w (4.3–5.1 LMH) and high nutrient rejection, with TAN rejection ranging from 80.8–97.0% and orthophosphate rejection from 98.7–99.8%.

Although effluent properties (TOC and viscosity) influenced J_w , no clear correlation between the methane yield, fouling potential and FO performance of an effluent was found. This shows that no compromise between high methane yield and J_w has to be made, when choosing a wastewater candidate.

Specifically, swine manure and potato starch achieved the highest methane yield and highest water flux out of four tested real effluents and could therefore be suitable wastewater candidates for an FO-AnMBR for simultaneous water purification and energy production.

During FO water extraction from anaerobic digested effluents the prevailing fouling is of biological and organic origin.

Taken together, these results indicate that FO membranes are suitable to be used in an FO-AnMBR-RO application, where high nutrient rejection and low reverse flux are essential to sustain simultaneous high product water quality and stable biogas production. In order to optimize the process, further work is warranted with regards to the re-concentration of the FO draw solution, wastewater pre-treatment and membrane cleaning strategies.

Acknowledgements

This work was supported by the Innovation Fund Denmark (Innovationsfonden) under the MEMENTO project with grant number 4106-00021B. The laboratory technicians at DTU Environment are thanked for conducting the IC and ICP-OES analyses. The authors would further like to acknowledge the support from Aquaporin A/S for providing the FO membranes.

~~Appendix A.~~Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.08.036>.

References

Achilli A., Cath T.Y. and Childress A.E., Selection of inorganic-based draw solutions for forward osmosis applications, ~~Journal of Membrane Science~~~~J. Membr. Sci.~~ **364** (1), 2010, 233–241.

American Public Health, A, Eaton, A.D, American Water Works, A and Water Environment, F, Standard ~~m~~M~~e~~thods for the ~~e~~E~~x~~amination of ~~w~~Water and ~~w~~Wastewater, 2005, APHA-AWWA-WEF; Washington, D.C..

Angelidaki I., Petersen S.P. and Ahring B.K., Effects of lipids on thermophilic anaerobic digestion and reduction of lipid inhibition upon addition of bentonite, ~~Applied microbiology and biotechnology~~~~Appl. Microbiol. Biotechnol.~~ **33** (4), 1990, 469–472.

Ansari A.J., Hai F.I., Price W.E., Drewes J.E. and Nghiem L.D., Forward osmosis as a platform for resource recovery from municipal wastewater - ~~A~~a critical assessment of the literature, ~~Journal of Membrane Science~~~~J. Membr. Sci.~~ **529**, 2017, 195–206.

Bassani I., Kougias P.G. and Angelidaki I., In-situ biogas upgrading in thermophilic granular UASB reactor: key factors affecting the hydrogen mass transfer rate, *Bioresour. Technol.* **221** (Supplement C), 2016, 485–491.

Bell E.A., Holloway R.W. and Cath T.Y., Evaluation of forward osmosis membrane performance and fouling during long-term osmotic membrane bioreactor study, ~~Journal of Membrane Science~~~~J. Membr. Sci.~~ 2016.

- Bell E.A., Poynor T.E., Newhart K.B., Regnery J., Coday B.D. and Cath T.Y., Produced water treatment using forward osmosis membranes: Evaluation of extended-time performance and fouling, *Journal of Membrane Science*, *J. Membr. Sci.* **525**, 2017, 77-88.
- Blandin G., Vervoort H., Le-Clech P. and Verliefde A.R.D., Fouling and cleaning of high permeability forward osmosis membranes, *J. Water Process Eng.* **9**, 2016, 161-169.
- Bowden K.S., Achilli A. and Childress A.E., Organic ionic salt draw solutions for osmotic membrane bioreactors, *Bioresource technology*, *Bioresour. Technol.* **122**, 2012, 207-216.
- Camilleri-Rumbau M.S., Masse L., Dubreuil J., Mondor M., Christensen K.V. and Norddahl B., Fouling of a Spiral Wound Reverse Osmosis Membrane processing Swine Wastewater: Effect of Cleaning Procedure on Fouling Resistance, *Environ. Technol.* 2015, 1-38, (just-accepted).
- Chang E.E., Yang S.-Y., Huang C.-P., Liang C.-H. and Chiang P.-C., Assessing the fouling mechanisms of high-pressure nanofiltration membrane using the modified Hermia model and the resistance-in-series model, *Separation and Purification Technology*, *Sep. Purif. Technol.* **79** (3), 2011, 329-336.
- Chen L., Gu Y., Cao C., Zhang J., Ng J.-W. and Tang C., Performance of a submerged anaerobic membrane bioreactor with forward osmosis membrane for low-strength wastewater treatment, *Water research*, *Water Res.* **50**, 2014 114-123.
- Clara M., Kreuzinger N., Strenn B., Gans O. and Kroiss H., The solids retention time—a suitable design parameter to evaluate the capacity of wastewater treatment plants to remove micropollutants, *Water research*, *Water Res.* **39** (1), 2005, 97-106.
- Coday B.D., Heil D.M., Xu P. and Cath T.Y., Effects of transmembrane hydraulic pressure on performance of forward osmosis membranes, *Environ. Sci. Technol.* **47** (5), 2013, 2386-2393.
- Cornelissen E., Harmsen D., De Korte K., Ruiken C., Qin J.-J., Oo H. and Wessels L., Membrane fouling and process performance of forward osmosis membranes on activated sludge, *Journal of Membrane Science*, *J. Membr. Sci.* **319** (1), 2008, 158-168.
- van der Kooij D., Assimilable organic carbon as an indicator of bacterial regrowth, *J. Am. Water Works Assoc.* 1992, 57-65.
- van der Wielen P.W. and van der Kooij D., Effect of water composition, distance and season on the adenosine triphosphate concentration in unchlorinated drinking water in the Netherlands, *Water research*, *Water Res.* **44** (17), 2010, 4860-4867.
- Gu Y.S., Chen L., Ng J.W., Lee C., Chang V.W.C. and Tang C.Y.Y., Development of anaerobic osmotic membrane bioreactor for low-strength wastewater treatment at mesophilic condition, *Journal of Membrane Science*, *J. Membr. Sci.* **490**, 2015, 197-208.
- Habel J., Hansen M., Kynde S., Larsen N., Midtgaard S.R., Jensen G.V., Bomholt J., Ogbonna A., Almdal K. and Schulz A., Aquaporin-Based Biomimetic Polymeric Membranes: Approaches and Challenges, *Membranes* **5** (3), 2015, 307-351.
- Hammes F., Goldschmidt F., Vital M., Wang Y. and Egli T., Measurement and interpretation of microbial adenosine tri-phosphate (ATP) in aquatic environments, *Water research*, *Water Res.* **44** (13), 2010, 3915-3923.
- Hashim D.M., Man Y.B.C., Norakasha R., Shuhaimi M., Salmah Y. and Syahariza Z.A., Potential use of Fourier transform infrared spectroscopy for differentiation of bovine and porcine gelatins, *Food Chemistry*, *Food Chem.* **118** (3), 2010, 856-860.
- Hey T., Zarebska A., Bajraktari N., Vogel J., Hélix-Nielsen C., la Cour Jansen J. and Jönsson K., Influences of mechanical pretreatment on the non-biological treatment of municipal wastewater by forward osmosis, *Environmental technology*, *Environ. Technol.* 2016, 1-10.
- Hey T., Bajraktari N., Davidsson Å., Vogel J., Madsen H.T., Hélix-Nielsen C., Jansen J.L.C. and Jönsson K., Evaluation of direct membrane filtration and direct forward osmosis as concepts for compact and energy-positive municipal wastewater treatment, *Environmental technology*, *Environ. Technol.* 2017, 1-13.
- Holloway R.W., Childress A.E., Dennett K.E. and Cath T.Y., Forward osmosis for concentration of anaerobic digester centrate, *Water research*, *Water Res.* **41** (17), 2007, 4005-4014.
- Hu T., Wang X., Wang C., Li X. and Ren Y., Impacts of inorganic draw solutes on the performance of thin-film composite forward osmosis membrane in a microfiltration assisted anaerobic osmotic membrane bioreactor,

RSC Advances *RSC Adv.* **7** (26), 2017, 16057-16063.

Kiriukhin M.Y. and Collins K.D., Dynamic hydration numbers for biologically important ions, *Biophysical Chemistry* *Biophys. Chem.* **99** (2), 2002, 155-168.

Kougias P.G., Boe K. and Angelidaki I., Effect of organic loading rate and feedstock composition on foaming in manure-based biogas reactors, *Bioresource technology* *Bioresour. Technol.* **144**, 2013, 1-7.

Lay W.C.L., Liu Y. and Fane A.G., Impacts of salinity on the performance of high retention membrane bioreactors for water reclamation: A review, *Water research* *Water Res.* **44** (1), 2010, 21-40.

Lee S., Boo C., Elimelech M. and Hong S., Comparison of fouling behavior in forward osmosis (FO) and reverse osmosis (RO), *Journal of Membrane Science* *J. Membr. Sci.* **365** (1), 2010, 34-39.

Li S., Kim Y., Phuntsho S., Chekli L., Shon H.K., Leiknes T. and Ghaffour N., Methane production in an anaerobic osmotic membrane bioreactor using forward osmosis: Effect of reverse salt flux, *Bioresource technology* *Bioresour. Technol.* **239**, 2017, 285-293.

Liu G., Van der Mark E., Verberk J. and Van Dijk J., Flow cytometry total cell counts: a field study assessing microbiological water quality and growth in unchlorinated drinking water distribution systems, *BioMed. Res. Int.* **2013**.

Liu Q., Liu C., Zhao L., Ma W., Liu H. and Ma J., Integrated forward osmosis-membrane distillation process for human urine treatment, *Water research* *Water Res.* **91**, 2016, 45-54.

Luo W., Hai F.I., Price W.E., Elimelech M. and Nghiem L.D., Evaluating ionic organic draw solutes in osmotic membrane bioreactors for water reuse, *Journal of Membrane Science* *J. Membr. Sci.* **514**, 2016, 636-645.

Lutchmiah K., Cornelissen E.R., Harmsen D.J., Post J.W., Lampi K., Ramaekers H., Rietveld L.C. and Roest K., Water recovery from sewage using forward osmosis, *Water Sci. Technol.* **64** (7), 2011, 1443-1449.

Lutchmiah K., Verliefe A., Roest K., Rietveld L.C. and Cornelissen E.R., Forward osmosis for application in wastewater treatment: a review, *Water research* *Water Res.* **58**, 2014, 179-197.

Mahdy A., Fotidis I.A., Mancini E., Ballesteros M., González-Fernández C. and Angelidaki I., Ammonia tolerant inocula provide a good base for anaerobic digestion of microalgae in third generation biogas process, *Bioresour. Technol.* **225** (Supplement C), 2017, 272-278.

McCutcheon J.R. and Elimelech M., Influence of concentrative and dilutive internal concentration polarization on flux behavior in forward osmosis, *Journal of Membrane Science* *J. Membr. Sci.* **284** (1-2), 2006, 237-247.

McGinnis R.L. and Elimelech M., Global challenges in energy and water supply: the promise of engineered osmosis, *Environ. Sci. Technol.* **42** (23), 2008, 8625-8629.

Melián-Martel N., Sadhwani J.J., Malamis S. and Ochsenkühn-Petropoulou M., Structural and chemical characterization of long-term reverse osmosis membrane fouling in a full scale desalination plant, *Desalination* **305**, 2012, 44-53.

Mi B. and Elimelech M., Chemical and physical aspects of organic fouling of forward osmosis membranes, *Journal of Membrane Science* *J. Membr. Sci.* **320** (1), 2008, 292-302.

Morrow C.P., McGaughey A.L., Hiibel S.R. and Childress A.E., Submerged or sidestream? The influence of module configuration on fouling and salinity in osmotic membrane bioreactors, *J. Membr. Sci.* **2017**.

Parida V. and Ng H.Y., Forward osmosis organic fouling: Effects of organic loading, calcium and membrane orientation, *Desalination* **312**, 2013, 88-98.

Phuntsho S., Vigneswaran S., Kandasamy J., Hong S., Lee S. and Shon H.K., Influence of temperature and temperature difference in the performance of forward osmosis desalination process, *J. Membr. Sci.* **415** (Supplement C), 2012, 734-744.

Provenzano M.R., Malerba A.D., Pezzolla D. and Gigliotti G., Chemical and spectroscopic characterization of organic matter during the anaerobic digestion and successive composting of pig slurry, *Waste Management* *Waste Manag.* **34** (3), 2014, 653-660.

Shannon M.A., Bohn P.W., Elimelech M., Georgiadis J.G., Marinas B.J. and Mayes A.M., Science and technology for water purification in the coming decades, *Nature* **452** (7185), 2008, 301-310.

Singh P.S., Joshi S.V., Trivedi J.J., Devmurari C.V., Rao A.P. and Ghosh P.K., Probing the structural variations of thin film composite RO membranes obtained by coating polyamide over polysulfone membranes of different pore dimensions, *Journal of Membrane Science* *J. Membr. Sci.* **278** (1), 2006, 19-25.

Singh N., Petrinic I., Hélix-Nielsen C., Basu S. and Balakrishnan M., Concentrating molasses distillery wastewater using biomimetic forward osmosis (FO) membranes, *Water research* *Water Res.* **130**, 2018, 271-280.

Stuckey D.C., Recent developments in anaerobic membrane reactors, *~~Bioresource technology~~Bioresour. Technol.* **122**, 2012, 137-148.

Tang M.K.Y. and Ng H.Y., Impacts of Different Draw Solutions on a Novel Anaerobic Forward Osmosis Membrane Bioreactor (AnFOMBR), 2014.

Tsapekos P., Kougias P.G. and Angelidaki I., Biogas production from ensiled meadow grass; effect of mechanical pretreatments and rapid determination of substrate biodegradability via physicochemical methods, *~~Bioresource technology~~Bioresour. Technol.* **182**, 2015, 329-335.

Tsapekos P., Kougias P.G., Treu L., Campanaro S. and Angelidaki I., Process performance and comparative metagenomic analysis during co-digestion of manure and lignocellulosic biomass for biogas production, *Appl. Energy* **185** (Part 1), 2017, 126-135.

Vang Ó.K., Corfitzen C.B., Smith C. and Albrechtsen H.-J., Evaluation of ATP measurements to detect microbial ingress by wastewater and surface water in drinking water, *Water Res.* **64** (Supplement C), 2014, 309-320.

Wang X., Chang V.W.C. and Tang C.Y., Osmotic membrane bioreactor (OMBR) technology for wastewater treatment and reclamation: ~~A~~advances, challenges, and prospects for the future, *~~Journal of Membrane Science~~J. Membr. Sci.* **504**, 2016, 113-132.

Wang Z., Zheng J., Tang J., Wang X. and Wu Z., A pilot-scale forward osmosis membrane system for concentrating low-strength municipal wastewater: performance and implications, *~~Scientific reports~~Sci. Rep.* **6**, 2016.

Wang X., Wang C., Tang C.Y., Hu T., Li X. and Ren Y., Development of a novel anaerobic membrane bioreactor simultaneously integrating microfiltration and forward osmosis membranes for low-strength wastewater treatment, *~~Journal of Membrane Science~~J. Membr. Sci.* **527**, 2017, 1-7.

WWAP, The United Nations World Water Development Report 2015, 2015, Water for a Sustainable World; Paris.

Xu J., Xu X., Liu Y., Li H. and Liu H., Effect of microbiological inoculants DN-1 on lignocellulose degradation during co-composting of cattle manure with rice straw monitored by FTIR and SEM, *~~Environmental Progress & Sustainable Energy~~Environ. Prog. Sustain. Energy* **35** (2), 2016, 345-351.

Xue W., Tobino T., Nakajima F. and Yamamoto K., Seawater-driven forward osmosis for enriching nitrogen and phosphorous in treated municipal wastewater: ~~E~~ffect of membrane properties and feed solution chemistry, *~~Water research~~Water Res.* **69**, 2015, 120-130.

York R., Thiel R. and Beaudry E., Full-scale Experience of Direct Osmosis Concentration Applied to Leachate Management, 1999.

Zarebska A., Nieto D.R., Christensen K.V. and Norddahl B., Ammonia recovery from agricultural wastes by membrane distillation: ~~F~~fouling characterization and mechanism, *~~Water research~~Water Res.* **56**, 2014, 1-10.

Zhao S., Zou L., Tang C.Y. and Mulcahy D., Recent developments in forward osmosis: opportunities and challenges, *~~Journal of Membrane Science~~J. Membr. Sci.* **396**, 2012, 1-21.

~~Appendix A.~~~~Appendix A.~~ **Appendix A. Supplementary data**

[Multimedia Component 1](#)

Supplementary material

alt-text: Image 1

Graphical abstract

Highlights

- Biomimetic FO membranes extract water from anaerobic digestion effluents.
- The membranes achieve high ammonia and orthophosphate rejection.
- No trade-off between methane yield and water flux and fouling propensity.
- Organic fouling and biological fouling ~~occured~~occurred on the active layer of the membrane.

Queries and Answers

Query:

Your article is registered as a regular item and is being processed for inclusion in a regular issue of the journal. If this is NOT correct and your article belongs to a Special Issue/Collection please contact p.das@elsevier.com immediately prior to returning your corrections.

Answer: Yes

Query:

Please confirm that given names and surnames have been identified correctly and are presented in the desired order, and please carefully verify the spelling of all authors' names.

Answer: Yes

Query:

The author names have been tagged as given names and surnames (surnames are highlighted in teal color). Please confirm if they have been identified correctly.

Answer: Yes

Query:

Please check whether the designated corresponding author is correct, and amend if necessary.

Answer: correct

Query:

Have we correctly interpreted the following funding source(s) and country names you cited in your article: "Innovation Fund Denmark, Denmark".

Answer: Yes

Query:

Supplementary caption was not provided. Please check the suggested data if appropriate, and correct if necessary.

Answer: Please replace the supplementary material with the updated version attached

Attachments: supplementary material_06082018.docx

Query:

Please provide the volume number and page range for the bibliography in Bell et al., 2016.

Answer: Volume 517, Pages 1-13

Query:

Please provide the volume number and page range for the bibliography in Liu et al., 2013.

Answer: vol. 2013, Article ID 595872, 10 pages

Query:

Please provide the volume number and page range for the bibliography in Morrow et al., 2017.

Answer: Volume 548, Pages 583-592