Desalination and Water Treatment
www.deswater.com

doi:10.5004/dwt.2018.22164

107 (2018) 10–19
March

Fouling characterization of TFC forward osmosis membrane in a novel dynamic sludge anaerobic digestion reactor

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Received 23 October 2017; Accepted 4 March 2018

ABSTRACT

The low-solid sludge concentration in reactors limited application of anaerobic digestion. Forward osmosis technology could be innovatively used for adjusting total moisture content of anaerobic digestion (AD) process in order to achieve a higher solid concentration and smaller reactor volume of AD reactor, for which membrane fouling is a crucial problem and determines the water flux and system performance. Many factors such as flux and composition of feed solution might have influence on the membrane fouling. In high-solid anaerobic digester, total solid could be critical to membrane fouling. A simulative anaerobic digestion forward osmosis membrane bioreactor (adFO-MBR) system was designed for fouling investigation of thin-film composite (TFC) forward osmosis membrane. Anaerobic digestion sludge samples with different total solid (TS) content or with different particle size was used as feed solution in this study. Water flux, adsorption capacity of protein and polysaccharose, SEM-EDX and AIR-FTIR were used for characterization of the pristine and fouled membranes. Experimental results showed that feed solution with TS at 5.5–8.0% had the highest and relatively stable water flux (5.5–7.3 L/m^2/h) because of the skeleton support and water passage in sludge with high TS. Water flux with smaller particulates and narrow particle size distribution had a slow decline trend. Adsorption capacity of protein and polysaccharose were positively related with total solid content of AD sludge. The results of this study can suggest us the appropriate parameters of AD sludge in adFO-MBR in terms of membrane fouling control.

Keywords: Forward osmosis; Membrane fouling; Dewatering; Anaerobic digestion sludge

1. Introduction

Waste activated sludge is the byproduct of wastewater treatment process, and sludge handling is a costly operation that often accounts for 50% of the total operating cost in wastewater treatment plants (WWTPs) [1]. The amount of waste sludge generated in WWTPs are huge and expected to increase continuously in the next decade, due to increasing population connected to sewage networks, building of new WWTPs, and upgrading of existing plants to fulfill more stringent local effluent regulations. In China, about 30 million tons of sewage sludge (20% solid content) is generated annually, and almost 80% of waste sludge has not been stabilized sufficiently [2], making it a big burden to the society and the environment.

Anaerobic digestion (AD) is a useful technology to stabilize organic wastes and convert organic matter into renewable energy biogas simultaneously [3]. However, sludge retention time of anaerobic digestion is usually very long, and it always results in a huge reactor volume with high construction cost. To tackle this problem, high solid concentration anaerobic digestion was proposed around 20 years ago, which could be used to reduce the reactor volume. However, it is still not widely practiced due to complicated reasons [2].

The anaerobic digestion is usually carried out at low-solid state (TS < 10%) [4]. If the TS of sewage sludge could be increased from 2.5% to 10%, the volume of sludge would be decreased to about one-fourth [5]. Besides smaller reactor and lower construction cost, high-solid anaerobic digestion has many other advantages over low-solid anaerobic digestion, such as less energy input for heating, minimal material

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handling, and so on [6,7]. Conventionally, in order to realize high-solid anaerobic digestion process, sludge pre-dewatered to some extent was used as feed. The composition of waste sewage sludge is mainly organic compared with primary sludge [8], and it makes the dewatering of waste sewage sludge very difficult. Also, pre-dewatered sludge would cause some local mass transfer problems or increase mixing energy input after being fed into digestive reactor. Dewatering during anaerobic digestion course could alter these situations.

In this research, an anaerobic digestion osmotic membrane bioreactor (adFO-MBR) that uses highly selective forward osmosis (FO) membranes to allow water permeation from sludge feed solutions (denoted as FS) to saline draw solutions (denoted as DS), was proposed as a novel advanced sludge anaerobic digestion process to realize simultaneous anaerobic digestion and sludge dewatering.

Researches on FO (or direct osmosis) could be dated back to the early 1960s [9]. During the last decade, FO process has become a hot topic because of various advantages such as low membrane fouling potential, high ratio of water recovery, and economic efficiency over the conventional desalination technologies. FO process has been applied to many fields such as seawater desalination [10], wastewater treatment [11–14], food processing [15–17] and power generation [18].

In adFO-MBR, forward osmosis process could draw water content from anaerobic digestion reactor continuously, therefore, sludge from secondary sedimentation tank, rather than dewatered sludge, could be directly used as feeding sludge. This process thus might overcome the mass transfer problem in conventional high-solid anaerobic digestion process and be energy saving. Outflow of adFO-MBR is high quality water that can be directly reused because of the high rejection by forward osmosis.

Membrane fouling in FO is moderate since the hydraulic pressure in FO unit is relatively low to reverse osmosis (RO) [19]. Even though, fouling of membrane can still decrease the water flux across the membrane substantially, and therefore remains a critical issue in FO process [20,21].

Researchers have made their efforts to understand the mechanism and influence of fouling in FO system. E.R. Cornelissen investigated the membrane fouling of forward osmosis membranes in osmotic membrane bioreactor (OMBR, which uses forward osmosis membrane to separate the effluent and the activated sludge) when the feed solution was activated sludge [9]. Xinhua Wang investigated effects of SRT on membrane fouling of forward osmosis membranes in the OMBR using a laboratory-scale submerged OMBR treating synthetic wastewater [22]. It was reported that many factors could characterize fouling of the FO membrane. And under various conditions reversible fouling was the dominant forward osmosis membrane fouling.

However, compared to aerobic sludge most researchers used before, AD sludge is of high concentration, high conductivity and high organic content [21,26]. Besides, the chemical structures of EPS extracted from both aerobic and anaerobic sludge were also found different [23]. To date, impacts of anaerobic sludge, especially anaerobic digested sludge, on membrane fouling in OMBR could hardly be found in the literatures.

In aerobic FO-MBR, water flux is considerably affected by properties of FS, such as solid content of the sludge. And in a like manner, total solid of AD sludge probably should also be a crucial factor that influences efficiency of anaerobic digestion process [24]. The total solids (TS) concentrations of anaerobic digestion sludge in publicly reported publications or practically operated projects were mainly among 3–10% [4], covering a wide range. Hence, it is indispensable to take a look into the relation of solid concentration of anaerobic digestion sludge and FO membrane fouling in an adFO-MBR to determine the most suitable TS concentration.

The objective of this study is to systematically investigate the fouling behavior and to verify the feasibility of forward osmosis membranes in adFO-MBR when anaerobic digestion sludge is used as feed solution. A series of experiments were thus carefully designed and conducted in order to evaluate the FO membrane performance with anaerobic digestion sludge, serving as a feasibility study for the operation and membrane fouling control of the novel adFO-MBR process.

2. Materials and methods

2.1. Feed sludge and draw solution

A lab-scale AD reactor was being maintained in lab to supply anaerobic digestion sludge as FS for the membrane fouling study. Dewatered sewage sludge from Yong Feng WWTP (Beijing, China) was used as substrate for the AD. The total solids (TS) of dewatered sludge ranged from 20% to 23% (w/w) and volatile solids (VS) accounted for 50–51% of TS. The substrate sludge was stored at 4°C and dissolved into deionized water (DI) before everyday feeding to make the TS around 5%. The mesophilic seed sludge collected from an anaerobic digester at an ecological garden (Beijing, China) had TS of 2.6% (w/w), among which VS made 56.8%.

The mesophilic (35°C) AD reactor with working volume of 6.0 L was equipped with a stirrer, which was set at a constant rate of 60 rpm with 10 min stirring and 10 min break alternate. Biogas produced was collected by air bag every single day.

In order to simulate FS sludge with different property, gravity thickening and ultrasonic generator were applied to AD sludge to make different solid concentration and particle size distribution.

The DS used in this study was 1 M sodium chloride solution (NaCl).

2.2. TFC forward osmosis membrane

Lab-made TFC forward osmosis membranes were used in this study. The fabrication method of FO membrane was based upon the work of Yip et al. [25], only that n-hexane was used as a substitute of Isopar-G.

2.2.1. Chemicals used for the fabrication of forward osmosis membrane and the detail process

All chemicals used for the fabrication of forward osmosis membrane are shown in Table 1. The brief procedure was described as below.

Table 1
Chemicals used in fabrication of Thin-film composite forward osmosis membrane

Chemicals	Specification	Manufacturer
Polysulfone (Psf)	Beads (Mn: 22,000 Da)	Sigma-Aldrich
N, N-dimethylformamide (DMF)	Anhydrous, 99.8%	Sigma-Aldrich
1-methyl-2-pyrrolidinone (NMP)	Anhydrous, 99.5%	Sigma-Aldrich
1,3-phenylenediamine (MPD)	>99%	Aladdin
1,3,5-benzenetricarbonyl trichloride (TMC)	98%	Sigma-Aldrich
n-hexane	Analytically pure	Baker
NaOCl	Analytically pure	Sinopharm
$NaHSO_3$	Analytically pure	Sinopharm

Polysulfone beads dried overnight were dissolved into a composite solvent in a 12 wt.% of DMF and NMP (1:3 on a weight basis) to prepare the membrane casting solution. Before being casted onto a clean glass plate with a casting knife at a 150 μm thickness, the membrane casting solution was stirred for at least 8 h and degassed 12 h. Immediately after casting, the thin as-cast liquid films were immersed in a precipitation bath with DI at room temperature. After 10 min, the membrane support layers formed in the precipitation bath were thoroughly rinsed with DI water and then kept in DI water until polyamide formation.

The polyamide active layer was fabricated via the interfacial polymerization of MPD dissolved in the liquid phase solution and TMC dissolved in the organic phase solution using the unified method. First, the membrane polysulfone support layers were immersed in a 3.4 wt.% MPD aqueous solution for 120 s. After removing excess MPD solution, the top surfaces of the MPD saturated membrane support layers were allowed to be in contact with 0.15 wt.% TMC in n-hexane for 60 s. Next, the fabricated TFC FO membranes were thoroughly rinsed by DI and stored in DI at 4°C for further use.

2.2.2. Morphology characterization of forward osmosis

The scanning electron microscopy (SEM) images of the TFC membrane fabricated in this study are shown in Fig. 1. The polysulfone supporting layer has finger-like voids close to top surface and sponge porous structure close to bottom surface (Fig. 1b). Physical parameters of the FO membranes were measured. The water flux and reverse salt flux values were determined using a laboratory-scale cross-flow reverse osmosis system. The effective membrane area was 42 cm^2. The DS and FS were 1 M NaCl and DI water respectively. The thickness of TFC membranes used in this study was 65 ± 2.

2.3. Experimental setup

A schematic diagram detailing the setup of a simulated adFO-MBR system is presented in Fig. 2. It consisted of a membrane cell, a peristaltic pump (YZ1515X, Longer Pump Corp., China), a draw solution tank and an electronic balance (PL2002, Mettler Toledo, Swit). The cell had dissymmetric channels on both sides of the membrane. On the active layer side, a FS tank with 100 mm long, 50 mm wide, and 75 mm deep was equipped with a stirrer; on the support layer side was a channel with dimensions of 100 mm long, 50 mm wide, and 2 mm deep, respectively. The peristaltic pump was used to circulate draw liquids at 25 rpm in a closed loop. The DS tank was placed on an electronic balance, which was connected to a computer in order to record the water variation volume (calculated from weight data).

Membrane fouling experiments were performed and the water flux as well as reverse salt flux was measured. Pristine membranes were used in each test in order to eliminate error brought by membrane properties. In each run, as water diffused across the membrane, DS was slowly diluted and the FS was thickened. In order to keep a relatively constant osmotic pressure, each test would run about 10 hours,

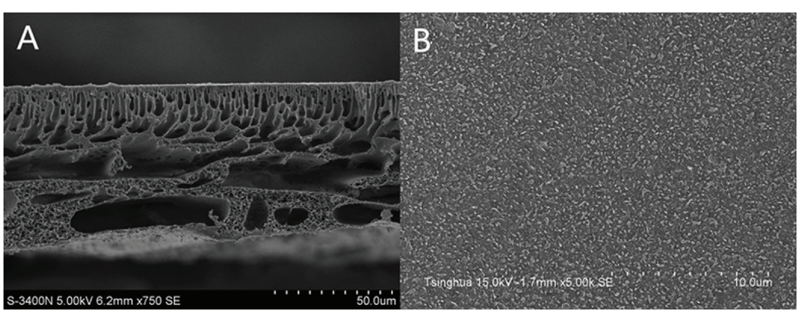

Fig. 1.

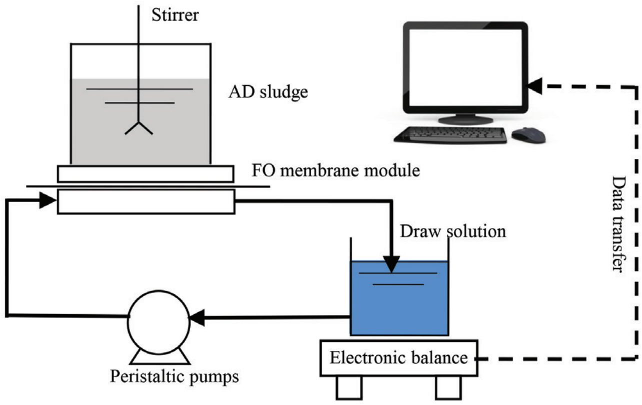

Fig. 2.

that was such a short time that the quality of AD sludge (FS) remained the same. For each run, the original volume of FS was 200 ml. Active layer faced the feed solution in order to minimize membrane fouling [9]. The experiment conditions are shown in Table. 2. A total of six kinds of sludge were used in adFO-MBR, all the tests were repeated three times. Unless stated otherwise, the experiments were conducted at constant room temperature of 20 ± 2°C.

2.4. Analytical methods

Thickness (T_s) and contact angle (CA) of pristine and fouled FO membrane were measured with a digital micrometer (293-240, Mitutoyo, Guangdong, China) and a contact angle meter (OCA20, Dataphysics, Germany), respectively. Before measurements, all samples were dried overnight in a desiccator. The surfaces of the pristine and fouled membrane were analyzed by field emission scan electronic microscopy (SEM, S-5500 Hitachi). For surface imaging, a small piece of a sample was air-dried and sputter coated with gold (Au) before being examined at 30 kV accelerating voltage for SEM analysis. All samples were dried in clean plastic bags at room temperature. The sample specimen was fixed on a slide glass and was measured with a suitable scan size. Data were collected and analyzed by the software. To characterize the organic pollution of FO membrane, AIR-FTIR (Vertex 70, Bruker, Germany) was used to analyze the change about functional group on the surface of FO membrane. The measurement of water flux and reverse salt flux of virgin membrane was similar with fouling experimental except the FS was DI water.

Conductivity, pH, particle size distribution, total organic carbon(TOC) and viscosity of the AD sludge were measured using a conductivity meter (DDSJ-318, Inesa, shanghai, China), pH meter (MP522, Sanxin, China), laser particle size analyzer (Mastersizer 3000, Malvern, UK), total organic carbon analyzer (TOC-V, CSN, Shimadzu) and rotational viscometer (NDJ-1, Changji Dizhi, Shanghai, China) respectively. TS and VS of AD sludge were measured by weight method. The soluble protein and carbohydrate concentrations were determined using the Lowry method and sulphuric acid-phenol method, respectively. The 4 cm^2 fouling membrane was cut into pieces and dissolved into 30 mL DI water and oscillated for 4h with the aid of oscillator (TS-100C, Tensuc, shanghai, China) to obtain a solution of fouling membrane (SFM). And the AD sludge and solution of fouling membrane were extracted for EPS for the further measurement.

Volatile fatty Acids(VFAs) of AD sludge was analyzed by a GC (6890N, Agilent, USA) with flame ionization detector. To analyze VFAs, the sludge samples from the reactors were centrifuged at 10,000 rpm for 10 min. Then the supernatant was passed through a microfiber filter (0.45 μm).

Table 2
Working conditions for each test (n = 3)

Test #	FS	TS (%)	Ultrasound power density (W/mL)	DS
1	AD sludge	3	–	1M NaCl
2	AD sludge	5	–	1M NaCl
3	AD sludge	7	–	1M NaCl
4	AD sludge	5	1	1M NaCl
5	AD sludge	5	2	1M NaCl
6	AD sludge	5	3	1M NaCl

3. Results and discussion

3.1. AD sludge (FS) composition

The characteristics of feeding sludge for lab-scale anaerobic digestion reactor and AD sludge (FS) for FO tests are shown in Table 3. The FS used in this study was neutral (pH = 7.00 ± 1.50). The total solid (TS) of the AD sludge was 5.0 ± 0.5%, which is regarded as conventional anaerobic digestion [4]. And the VS/TS (%) of AD sludge was about 48%.

3.2. Effects of TS on FO membrane fouling

3.2.1. Effects of TS on FO water flux drop

Fig. 3 shows the effects of different TS on the water flux decline under identical experimental conditions other than

Table 3
The characteristics of the feeding sludge for anaerobic digestion reactor and AD sludge for FO tests

Analyzed items	Units	Feeding sludge for lab-scale anaerobic digestion reactor	Anaerobic digestion sludge for FO tests
pH		6.89	7.00±1.50
TS	%	6.4±0.5	5.0±0.5
VS/TS	%	58±1	48±1
FAN	mg/L	280±3	625±20
SCOD	mg/L	4055±5	1098±100
TOC	mg/L	437	268±10
VFAs	mg/L	–	140±20
CH_4	%	–	60±10

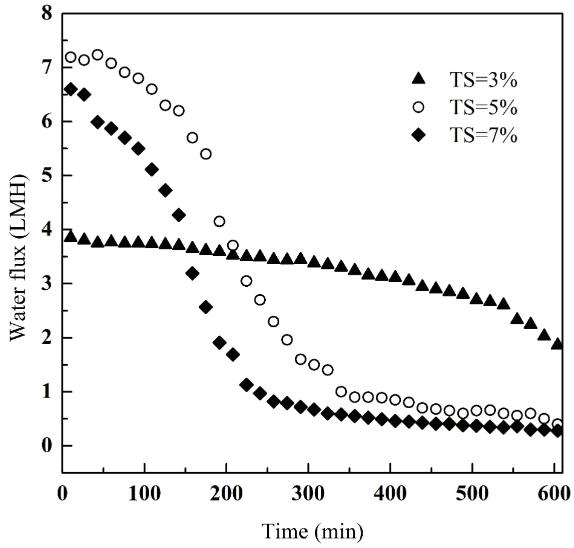

Fig. 3.

TS concentration. The variation of water flux was recorded for approximately 10 h. When the TS of feed sludge was 3%, the FO water flux declined slowly. During the first 7 h continuous operation, the flux only dropped about 20%. And the dropping curve became steep as time went by. By the end of 10 h, 40–45% of original flux was reduced as a result of the membrane fouling.

In terms of the flux decline rate, it was totally different in tests with higher TS feed sludge. When the TS of AD sludge were higher (i.e. TS = 5%, 7%), the water fluxes had drastic drops. Much higher initial water fluxes were also observed for 5% or 7% feed sludge compared with that of 3%. However, there were severe declines of water flux in the following 300 min until they were way below the curves of 3%. By 300 min operation, 80–90% of initial flux was reduced as a result of the membrane fouling.

This might be ascribed to the co-function of skeleton support of sludge particulates and moisture content of sludge. Some researchers reported that certain additives could be used as skeleton builder to enhance sludge dewaterability [27]. Many skeleton builders have been reported such as gypsum, coal fly ash, wood chips and wheat dregs [28–30]. These skeleton builders form a skeleton via the interaction to obtain a better dewaterability of sludge. In the filtration by membrane, the effect of skeleton support was observed from the dewater process. In sludge reactors, micro flocs with different sizes, i.e. the so called zooglea, might help to form the skeleton support when the hydraulic pressure is not too high. The size of these micro flocs is probably under the effects of TS of sludge because different micro-environment (e.g. F/M ratio) would lead to different EPS excretion which is very important in the forming of zooglea. Also, if the TS is as low as 3%, it means the chances for micro flocs to interact with each other in the mixed liquor are few, and very compact arrangement on the membrane surface would happen resulting in a high resistance and low flux. For 5% and 7% tests, the situations are different. As can be seen in Fig. 4, when TS of AD sludge was higher than 3%, the interaction force of sludge particulates could probably

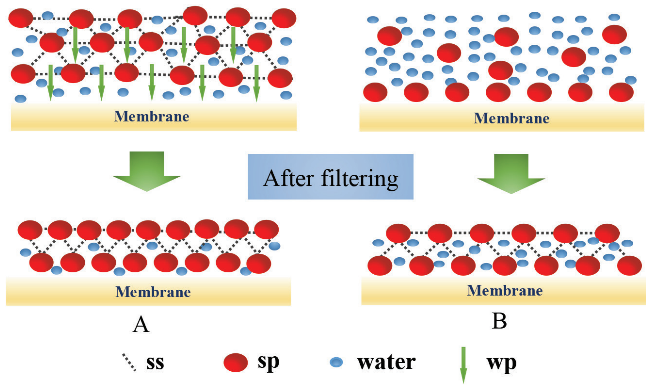

Fig. 4.

form a *skeleton*, in this process the sludge particulates was used as the skeleton builder. Therefore, the dewaterability of sludge was improved. To be specific, the water can flow past in a higher flux, which could be named as *water passage* (Fig. 4). In this study, the higher concentration of sludge particulates in 5% and 7% FS facilitate the formation of *skeleton* and *water passage*.

On the other hand, the form of skeleton and water passage could also be influenced by particle size of sludge. As shown in Fig. 5, the particle size distribution changed with TS of sludge. The volume density of larger particles rose when the TS concentration decreased from 7% to 3%. That is to say, 5% sludge has more large particles than 7% sludge. According to the mechanism shown in Fig. 4, the particle size of sludge can influence the form of water passage. Therefore, when the skeleton formed, there would be wider water passages in sludge with 5% TS. As described in Fig. 3, initial water flux of AD sludge with 5% TS was higher than that of 7%. However, though the 3% TS sludge has the largest particles, it was still probably below the

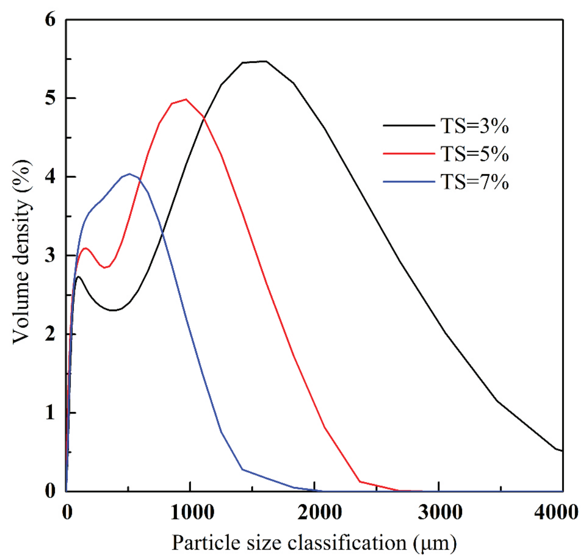

Fig. 5.

threshold of skeleton forming, hence the initial water flux was the lowest.

And It also should be noted that the final water flux of lowest solid content (TS = 3%) was highest in this study, confirming that moisture content could be as important as the skeleton support.

3.2.2. Viscosity and flux decline

The viscosity of AD sludge was measured by viscometer (data shown in Fig. 6). The viscosity was found to increase with the TS of AD sludge. When the TS of AD sludge increased from 3% to 5%, the latter sludge showed a viscosity nearly six times that of the former one. And, as TS increased from 5% to 7%, and then to 10%, the increase rate of viscosity was even higher. The increase in viscosity with TS was due to resistance offered by increased inter- and intra-particle interactions. Additionally, formation of flocs or other cell aggregates can also be mediated by extracellular polymeric substances (EPS), which is highly hydrated gel and bind microorganisms together. Probably, low EPS concentration at lower TS could have decreased the floc strength and then result in lower viscosity values as estimated in earlier studies [31]. The presence of a different viscosity seemed to affect the formation of skeleton support and water passage. A higher viscosity might increase the strength of skeleton as described in Fig. 4(A), so that water could flow through water passage easily. Based upon this point, an appropriate viscosity of AD sludge is significant for adFO-MBR process.

3.2.3. Water flux and average sludge moisture content

In order to show the way how the average TS of feed sludge influences the water flux, the variations of water flux with the real-time average TS of FS sludge (AD sludge) are shown in Fig. 7. Generally speaking, water fluxes in all three tests decreased as the average TS of feed sludge increased.

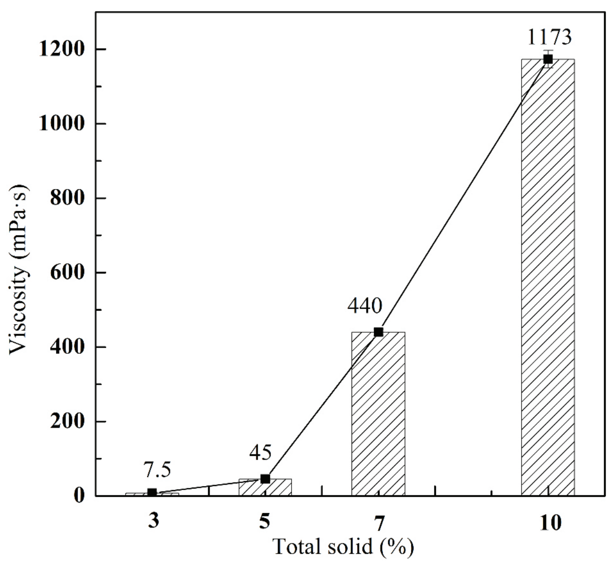

Fig. 6.

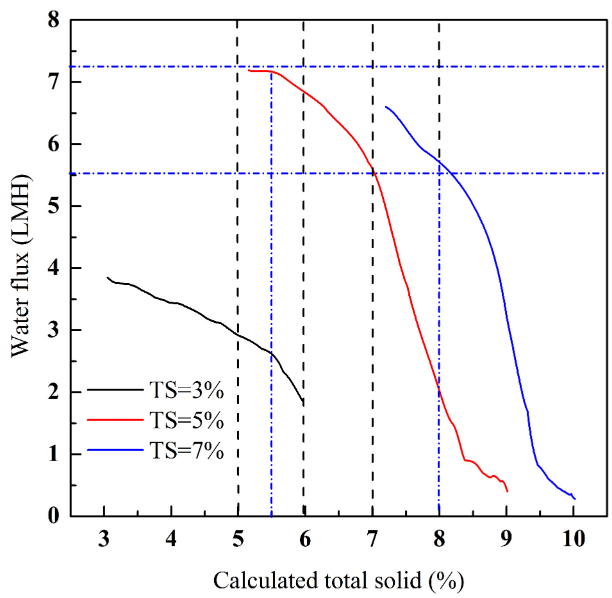

Fig. 7.

Another feature is that even the average real-time total solid concentrations are same the water fluxes were various in different tests. In test 3% and test 5%, water fluxes of each test vary a lot when TS of both tests were from 5% to 6%. Similarly, in test 5% and test 7%, water fluxes of each test also vary a lot from 7% to 9%. It could be conjectured that the average total solid concentration cannot determine the water flux alone. The water flux might also be determined by the structure of attached cake layer that is right on the membrane surface and formed during the process of dewatering. One point should be stressed here is the importance of initial total solid of AD sludge on the cake layer structure. As shown in Fig. 7, when the TS was 6%, there was a gap of water flux between two curves standing for initial TS 3% and 5% respectively. Apparently, it could be told from Fig. 7 that the TS concentration of feed sludge with each test had very important influences on the shape of the curves. The nominal TS concentration which was calculated based on the cumulative volume of water drawn out clearly cannot determine the real-time flux. Based upon our analysis, the structure of a very thin cake layer (the so-called skeleton support) which attached on the very top of the membrane surface might be the key reason controlling the actual flux. And its structure means a lot to the filtration resistance. This could be used for all gaps of water flux between different curves when TS was the same.

As shown in Fig. 7, when the TS of sludge in adFO-MBR were 5.5–7.0%, highest water flux (5.5–7.3 LMH) and comparatively lower water flux decline rate were received along red curve (5% TS test). Stabilized water flux in this reactor could be easily gained by adjusting the initial TS concentration of feed sludge to around 5%. And around this TS concentration, the viscosity of AD sludge was very low so that fouling of FO membrane could be not that severe. In the test 7% (the initial TS), the water flux was also high (5.6–6.6 LMH) when TS was between 7% and 8%. In practical application the influence of initial TS in sludge might be weakened. The results of test 7% could also be integrated to guide the practical application. Therefore, the TS of AD sludge could be adjusted to 5.5–8.0% during the operation of adFO-MBR to obtain stabilized levels of water flux and TS of sludge.

3.3. Organic and inorganic fouling of FO membrane

3.3.1. Effects of TS on organic concentration and adsorption capacity of FO membrane

A small piece (3 * 2 cm) of the fouling membrane after drying was cut off and cut up, then soaked into 15 mL of deionized water, after 10 min oscillation on a table shaker, the concentration of protein and polysaccharose in eluent was measured to obtain adsorbed capacity of FO membrane. Variations of concentration in protein, polysaccharose and adsorbed capacity of FO membrane are displayed in Fig. 8. It can be seen that organic concentration in sludge and the adsorbed capacity on FO membrane were positively correlated. The capacity proportions were almost constant among different TS values, even though concentrations of protein and polysaccharose increase along with the increment of TS in AD sludge. High organic concentration would increase the external concentration polarization (ECP) at the FO membrane surface, which could bring about more serious membrane fouling and flux decline [32].

Hence proved again, it is important to keep an appropriate solid content in a practical application to mitigate the membrane fouling. When TS is around 5.5–8.2%, concentrations of protein and polysaccharose are relatively low (600–1000mg/L; 300–550 mg/L) [33] so that fouling of FO membrane will be mild.

3.3.2. Air-FTIR of virgin and fouling FO membrane

Difference of FTIR spectra between pristine and fouled membranes could be used to describe the organic composition of the fouling layer, as shown in Fig. 9. The specific absorbing peaks of pristine TFC forward membrane were 1144, 1239, and 1485 cm^{-1}. However, all peaks of pristine membrane disappeared or were weakened when the fouling layers were present. On the fouled membranes, peaks near 2923 and 2852 cm^{-1} represent aliphatic methylene groups; proteins are indicated by the amide I and amide II peaks at 1637 and 1540 cm^{-1}; and peaks at approximately 1028 cm^{-1} represent C-O stretching of a polysaccharide. The peaks are same for all membranes fouled by AD sludge with different TS, and are in accord with peptidoglycans found in bacterial cell walls [34]. So, in conclusion, FTIR results show a predominance of bio-organic fouling such as soluble microbial products (SMP). Furthermore, FTIR signals of 3% sludge is less intense than that of 7% and 5%, suggesting positive correlation of FTIR intensity with flux decline rate (see Fig. 3 and Fig. 7).

3.3.3. Inorganic fouling of FO membrane

SEM-EDX analyses were used to compare the elemental content on the surface of pristine and fouled membranes (by AD sludge with TS 5%), and the results was shown

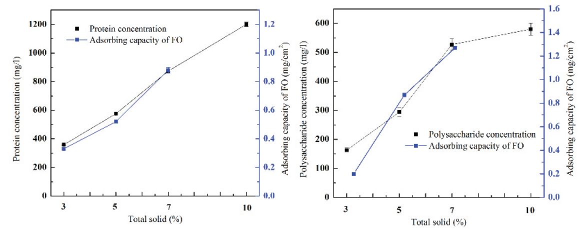

Fig. 8.

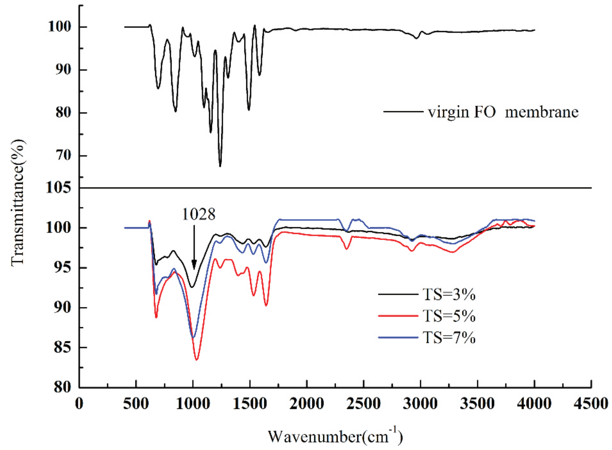

Fig. 9.

in Fig. 10. Large amount of sulfur was found on the pristine membrane and is probably because of its poly-sulfone support layer. The fouled membrane surface was covered with thick layer, and obvious inorganic particles could be observed on the surface of fouled membrane. SEM-EDX analyses revealed that calcium, iron, aluminum, sodium, and phosphorus were the dominant element in fouling membranes, followed by silicon, magnesium, sulfur, chlorine and potassium as shown in Table 4.

3.4. Effects of particle size on membrane fouling

Many factors could influence the particle size and its distribution of sludge, e.g., stirring and SRT. Fig. 11 shows the particle size distribution of AD sludge when pretreated with different ultrasonic intensity. With increment of ultrasonic intensity exerted on AD sludge, the volume density of smaller particles (i.e., particle size from 0 to 500 µm) rose while the volume density of larger particles (i.e., particle size from 500 to 3000 µm) went down. The fouling layers that developed on the membrane surfaces were visually examined with SEM and are shown in Fig. 12. The SEM images also suggest that AD sludge particles became smaller and smaller with the increase of ultrasonic intensity.

The effects of particle size on water flux are illustrated in Fig.13. Based upon Fig.13, the initial water flux declined with the increase of ultrasonic intensity. Water flux in tests with higher initial values decreased faster. Taking 3 W/mL test for an example, although the initial water flux was the lowest, it exhibited the most stable flux tendency.

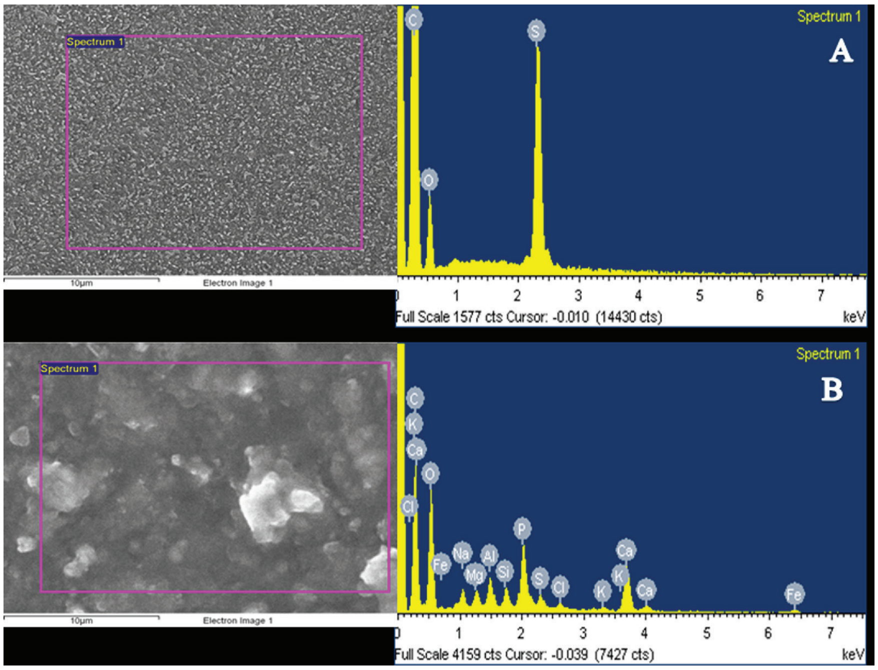

Fig. 10.

Table 4
Specific content of elements of fouled FO membrane

Element	Weight%	Atomic%
C	47.16	58.10
O	37.97	35.12
Na	1.26	0.81
Mg	0.96	0.58
Al	1.39	0.76
Si	0.98	0.52
P	3.55	1.70
S	0.64	0.29
Cl	0.35	0.15
K	0.25	0.09
Ca	4.00	1.48
Fe	1.49	0.39
Totals	100.00	

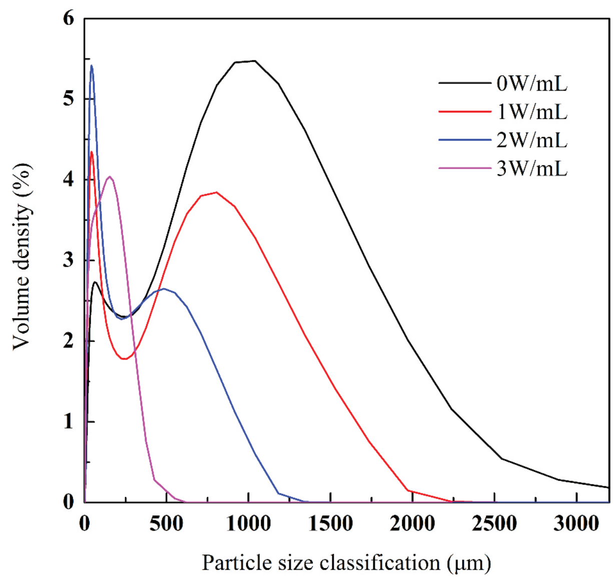

Fig. 11.

This phenomenon could also be attributed to the formation of water passage. Sludge with large particles seems to form water passage more easily, like AD sludge without ultrasonic pretreatment or with low ultrasonic intensity. However, high initial water flux doesn't mean it could last. Otherwise, like tests in Fig. 3, low initial water flux could sustain a relatively stable flux. This is a very interesting phenomenon, which means water passage might be effective only under certain circumstances. The formation of water passage enables fast transportation of water. And then, the relatively large water flow brings more stuff to block the water passage, and then result a quick decrease of water flux. In the test when 3 W/mL ultrasonic intensity was applied, there maybe only "micro-water passage" formed due to small and uniformed water particles. "Micro-water passage" results in slow water transportation, the drag force resulted by which is probably not enough to bring to the water passage as many stuff as the others do.

Another mechanism that might explain the phenomenon above could be different fouling cake layer structures formed by large particles and small particles. Chanhee Boo used two suspensions of silica particles of different sizes as model col-

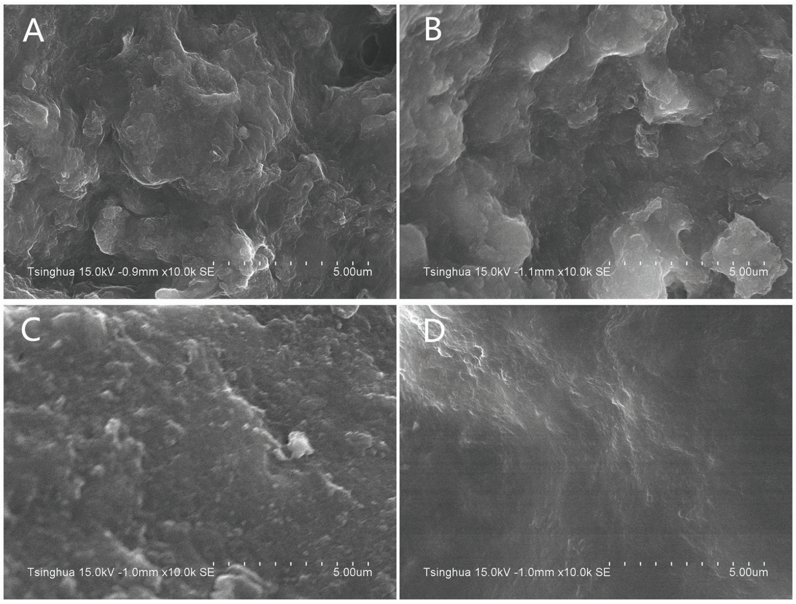

Fig. 12.

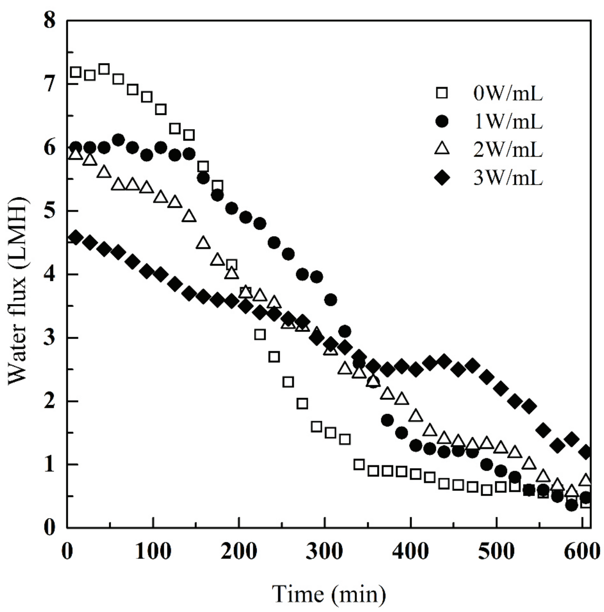

Fig. 13.

loidal foulants and much greater flux decline was observed with the large colloidal suspensions. They concluded that small particles that are transported to the membrane surface by the permeate flow could readily diffuse back into the bulk solution, and large particles had much lower back transport rate compared to the small particles [35].

4. Conclusion

Compared to 3%, high water flux was found at the starting period when the initial TS of AD sludge was 5%–7%, as a result of more sludge particulates in high concentration facilitating formation of *skeleton* and *water passage*. Higher viscosity could increase the strength of skeleton. Water flux decline faster when the initial sludge moisture content was low. Therefore, initial sludge TS was recommended as 5%, and when the calculated or average TS of sludge in adFO-MBR were 5.5–8.0%, high water fluxes (5.5–7.3 LMH) at the starting period could be obtained.

On account of the positive correlation of adsorption capacity of FO membrane and organic content of FS, it is important to maintain appropriate moisture content in practical applications to minimize membrane fouling. FTIR results showed significance of bio-fouling and organic fouling. The inorganic fouling analyses revealed that Calcium, phosphorus, iron, aluminum and sodium were the dominant elements on fouling membranes, followed by silicon, magnesium, sulfur, chlorine and potassium.

The particle size of AD sludge pretreated with ultra-sonication was smaller and homogeneous. When treating ultra-sonication pretreated sludge with FO, the initial water flux decreased, while it could relatively high flux longer than un-pretreated sludge or less pretreated sludge.

Acknowledgment

This research was supported by the Fundamental Research Funds for the Central Universities (2015ZCQ-HJ-01 and 2016ZCQ03).

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